# Phase diagram for the cubic model in the Kikuchi approximation

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(Received 3 May 1978)

A variant of the three-state Potts model known as the cubic model has been used to describe the critical behavior of some rare-earth pnictides. In the mean-field (MF) approximation this model has a tricritical-like phase transition, while in the Bethe-Peierls-Weiss (BPW) approximation the transition is first order. To obtain a better estimate of the phase diagram of this model we have used the Kikuchi cluster variational method with a tetrahedron as the basic cluster. By using the symmetry properties of the cubic model we were able to reduce the 36 coupled equations with 1296 variational parameters to 12 coupled equations and 58 distinct variational parameters. We were able to solve these equations and find the surfaces in the phase diagram where transitions occurred. The qualitative features of the phase diagram for the cubic model do not change too much in going from the BPW to the Kikuchi approximation. However, there are quantitative changes which become large for certain points and along symmetry directions in the phase diagram, i.e., for those values of the anisotropy ($\vec{D}$) for which several components of the spin become simultaneously critical. For these points the Kikuchi tetrahedron approximation takes much better account of the correlations between spins than the MF and BPW approximations. It follows that the estimates of the critical fields $\vec{D}_c$ and quadrupolar coupling $\eta_c$ necessary to drive the transition tricritical are substantially better in the Kikuchi tetrahedron approximation. At the origin $\vec{D}=0$ we find a first-order phase transition occurs at a temperature 13% lower than that found with the MF approximation and 5% lower than the BPW approximation.

## I. INTRODUCTION

To study the phase transitions in a series of cubic rare-earth compounds we recently introduced the model Hamiltonian[1]

$$H=-\mathcal{J}\sum_{\langle ij\rangle}\sigma_i\sigma_j\delta_{\alpha_i\alpha_j} \qquad (1)$$

where $\sigma$ takes on the value $\pm1$ and $\alpha=x$, $y$, and $z$. This Hamiltonian is a variant of the three-state Potts model [which is Eq. (1) without the Ising-like variables $\sigma_i\sigma_j$] and is called the cubic model. It was arrived at by projecting a bilinear isotropic spin interaction on to the sixfold-degenerate ground-state manifold of rare-earth ions in appropriate cubic crystal fields. The reader is advised to look at Refs. 1 and 2 to acquaint himself with the previous studies of the thermodynamic behavior of the cubic model.

In the mean-field (MF) approximation we found that the cubic model has a continuous phase transition with tricritical-like exponents.[1] To determine whether this is a property of the model or owing to the MF approximation, we used the Bethe-Peierls-Weiss (BPW) approximation. We found that the phase transition for the cubic model is first order, i.e., discontinuous.[2] However, by applying small single-ion anisotropy fields or by including suitable quadrupolar pair interactions we did find that the system can be made to undergo a continuous transition. This change from a discontinuous to a continuous transition is expected since for large enough anisotropies the cubic model reduces to an Ising model.[1] Therefore, in the temperature–single-ion-anisotropy, $T$-$\vec{D}$, space there exist lines of tricritical points about the temperature ($\vec{D}=0$) axis which form the boundary between regions of first-order and continuous transitions. We will use the term tricritical in this paper to mean the change from a first-order to continuous phase transition; indeed, the transition may be a multicritical one. We have previously used the BPW approximation to determine the magnitudes of the single-ion anisotropies and the quadrupolar pair interaction to drive the phase transition tricritical.[2]

To obtain a better idea of the nature of the phase diagram of the cubic model we have now determined it by using the Kikuchi cluster variational method.[3] With this method we obtain a *sequence* of approximate results from which we *extrapolate* reliable information about the transition region. As the size of the basic cluster used in these approximations increases, the results become increasingly accurate, i.e., approach those obtained from exact series analysis.[3]

The Hamiltonian for the cubic model in the presence of external fields and an isotropic quadrupolar pair interaction is given as[2]

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$$H = -\mathcal{J}\sum_{\langle ij\rangle}\vec{S}_i\cdot\vec{S}_j - \eta\mathcal{J}\sum_{\langle ij\rangle}\left(\frac{3S_{z_i}^2-1}{2\sqrt{3}}\right)\left(\frac{3S_{z_j}^2-1}{2\sqrt{3}}\right)+\tfrac{1}{4}(S_{x_i}^2-S_{y_i}^2)(S_{x_j}^2-S_{y_j}^2)-\vec{H}\cdot\sum_i\vec{S}_i$$
$$-D_1\sum_i\left(\frac{3S_{z_i}^2-1}{2\sqrt{3}}\right)-\tfrac{1}{2}D_2\sum_i(S_{x_i}^2-S_{y_i}^2)\ , \qquad (2)$$

where the $i$ and $j$ go over the $\frac{1}{2}zN$ nearest-neighbor pairs and we have written the quadrupolar coupling $\kappa$ as $\eta J$. The first term is equivalent to Eq. (1). To see this recall that the spin operator $S_\alpha$ has eigenvalues $\pm1$ when the spin points in the $\pm\alpha$ directions and zero otherwise.

The symmetry of the phase diagram of the cubic model with quadrupolar pair interactions in $T$-$\vec{H}$-$\vec{D}$ space follows from the invariance of the free energy under a group of symmetry transformations of the Hamiltonian Eq. (2). It is difficult to portray the phase diagram in the six-dimensional space $T$-$\vec{H}$-$\vec{D}$, and we have limited our preliminary studies to the subspace $\vec{H}=0$.[4] The phase diagram for $\vec{H}=0$ and for fixed temperature displays $C_{3v}$ symmetry, that is, for each point $(D_1,D_2)$ there are five other equivalent points with identical free energies. The various statistical mechanical approximations used to determine the nature of the phase transition, i.e., whether it is continuous or discontinuous, do not change the symmetry of the diagram.

For the case of no quadrupolar pair interactions ($\eta=0$) we found in the MF approximation that the transitions are continuous for all values of $\vec{D}$, including zero. For a fcc lattice in the BPW approximation, we found for $\eta=0$ that there is a triangular patch about the origin $\vec{D}=0$ for which the transitions are discontinuous, while outside the patch the transitions are continuous, see Fig. 1. In this paper we show that when we use successively larger clusters in the Kikuchi variational method the first-order patch becomes larger, particularly along the negative $D_1$ and symmetry related directions (see Fig. 2).

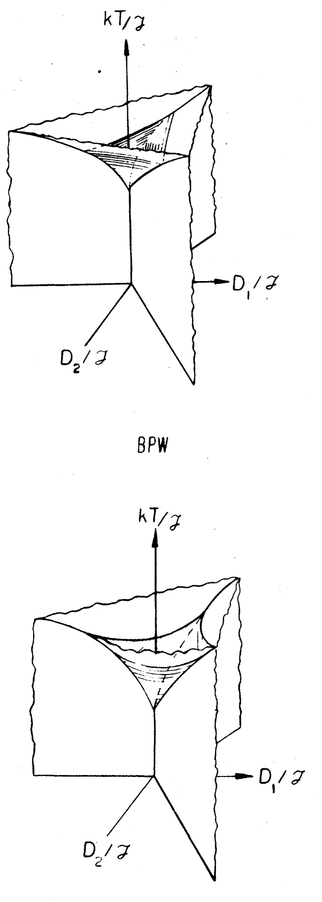



FIG. 1. Phase diagram for the cubic model in $T$-$\vec{D}$ space in the BPW and Kikuchi approximations. There is a surface of first-order phase transitions about the origin $\vec{D}=0$. Outside the line of tricritical points the surface consists of critical points. The regions below these surfaces are volumes where two phases exist in zero magnetic field $\vec{H}=0$. These volumes are separated by three vertical planes along which four phases coexist.

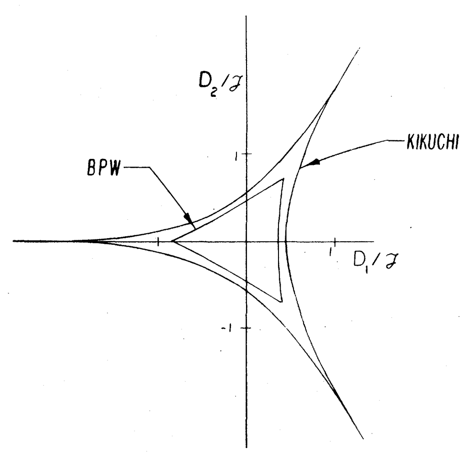



FIG. 2. Single-ion anisotropy fields $\vec{D}_c$ necessary to drive the phase transition tricritical in the BPW and Kikuchi approximations for a system described by Eq. (2) with $\eta=\vec{H}=0$. Points inside a curve represent first-order phase transitions; those outside represent continuous transitions. In this figure the anisotropy fields $\vec{D}$ are measured in units of the coupling constant $\mathcal{J}$.

We have also studied the phase diagram of the model Hamiltonian Eq. (2) in $T$-$\vec{D}$ space as a function of the quadrupolar coupling parameter $\eta$. For positive $\eta$ (ferroquadrupolar coupling) the region about the origin in $T$-$\vec{D}$ space where first-order phase transitions occur is enlarged, while for antiferroquadrupolar interactions ($\eta < 0$) this region shrinks until one reaches a critical value $\eta_c$ for which phase transitions for all regions in $T$-$\vec{D}$ space are continuous. In the MF approximation we found $\eta_c = 0$, i.e., the cubic model ($\eta = 0$) has a tricritical-like phase transition for $\vec{D} = 0$. For a fcc lattice we found $\eta_c = -0.092$ in the BPW approximation, while with a tetrahedron as a basic cluster in the Kikuchi method, we find it is much larger, $\eta_c = -0.452$.

The discrete-spin three-state Potts model is a special case of the cubic model Eq. (1) in which one removes the Ising variables $\sigma_i \sigma_j$. This is equivalent to letting in Eq. (2) $\mathcal{J}$ go to zero while keeping $\eta \mathcal{J}$ finite. Having developed the procedures and algorithms necessary to determine the phase behavior of the cubic model in the Kikuchi approximation, it was a simple extension to determine the phase diagram of the three-state Potts model in this approximation. Our results are presented elsewhere [Ref. (5)]. Here let us just mention that, in contrast to the behavior of the cubic model, we find that the region of first-order phase transition for the three-state Potts model shrinks considerably on going from the mean-field to the Kikuchi tetrahedron approximations; in fact it nearly disappears.

In Secs. II–IV we describe the application of the Kikuchi cluster variational approximation to the cubic model, then we present our results and a discussion of them.

## II. KIKUCHI APPROXIMATION

The basic tenet behind the Kikuchi approximation is that as the size of the cluster used to determine the entropy increases, the free energy becomes increasingly more accurate. This has been substantiated by Kikuchi and others[3] by expanding the free energy and its derivatives about the high-temperature limit ($1/T = 0$) and comparing the results of the various cluster approximations with the "exact" high-temperature series expansions. One finds that as the size of the cluster increases, more terms in the expansion obtained from the Kikuchi approximation to the free energy agree with the "exact" expansion.[3] In the following calculations we consider a fcc lattice; this is the lattice of magnetic ions for real systems to which the cubic model applies.

In the Kikuchi approximation the free energy of the cubic model is written as

$$F = E - TS \ , \tag{3}$$

where the energy, see Eq. (2), to within a constant is given as

$$E/N = -6\mathcal{J} \sum_{ij} (\sigma_i \sigma_j + \tfrac{1}{2}\eta)\,\delta_{\alpha_i \alpha_j} X_{ij} - \sum_i [H_{\alpha_i}\sigma_i + \tfrac{1}{2}\sqrt{3}D_1 \delta_{\alpha_i z} + \tfrac{1}{2}D_2(\delta_{\alpha_i x} - \delta_{\alpha_i y})] P_i \ . \tag{4}$$

The sums are over the six states of a site which we label one through six, and we use the following notation

$$\alpha_i = \begin{cases} z, & i = 1,2 \\ x, & i = 3,4 \\ y, & i = 5,6 \end{cases}$$

and

$$\sigma_i = \begin{cases} 1, & i = 1,3,5 \\ -1, & i = 2,4,6 \end{cases} \ . \tag{5}$$

This notation implies $i = \sigma_i \alpha_i$, e.g., for $i = 2 = -z$. The entropy depends on the cluster size. In the MF approximation where we consider a site as our cluster, the entropy is given as

$$S/kN = -\sum_i P_i \ln P_i \ . \tag{6}$$

With a tetrahedron as our basic cluster for a fcc lattice, we find

$$S/kN = 6 \sum_{ij} X_{ij} \ln X_{ij} - 2 \sum_{ijkl} W_{ijkl} \ln W_{ijkl} - 5 \sum_i P_i \ln P_i \ . \tag{7}$$

$P_i$ is the probability of finding a site in state $i$, i.e., $\sigma_i \alpha_i$. $X_{ij}$ is the probability of finding nearest-neighbor sites in states $i$ and $j$. $W_{ijkl}$ is the probability of finding a tetrahedron in the configuration $ijkl$. The probabilities $P_i$ and $X_{ij}$ are related to the $W_{ijkl}$ as follows

$$P_i = \sum_{j=1}^{6} X_{ij} = \sum_{j=1}^{6} \left( \sum_{kl=1}^{6} W_{ijkl} \right) \ . \tag{8}$$

We find the equilibrium values of these probabilities by minimizing the free energy, Eq. (3), with respect to variations of the tetrahedron probabilities $W_{ijkl}$. The variations are subject to the constraints of Eq. (8) and $\sum_i P_i = 1$. By using the symmetry properties of the cubic model we relate the 1,296 different tetrahedron probabilities (four spin sites, each with six possible orientations: $6^4$ sites) to 58

TABLE I. Anisotropy fields and antiferroquadrupolar pair coupling necessary to drive the cubic model tricritical in various approximations.

| | $D_1>0$ | | $D_1<0$ | | $\vec{D}=0$ | |
|---|---|---|---|---|---|---|
| | $kT_c/\mathcal{J}$ | $D_{1c}/\mathcal{J}$ | $kT_c/\mathcal{J}$ | $D_{1c}/\mathcal{J}$ | $kT_c/\mathcal{J}$ | $\eta_c$ |
| Mean-field | 4 | 0 | 4 | 0 | 4 | 0 |
| Bethe-Peierls-Weiss | 3.90 | 0.38 | 3.90 | −0.82 | 3.64 | −0.092 |
| Kikuchi tetrahedron | 3.77 | 0.44 | 4.07 | −2.70 | 3.12 | −0.452 |

distinct ones, and we reduce the 36 coupled equations to a set of 12 equations. These are solved self-consistently by the natural iteration process developed by Kikuchi.[6] With this procedure we find the energy, entropy, free energy, the order parameter $\langle S_z\rangle \equiv P_1-P_2$, and expectation values $\langle S_z^2-\frac{1}{3}\rangle \equiv (P_1+P_2)-\frac{1}{3}$ and $\langle S_x^2-S_y^2\rangle \equiv P_3+P_4-P_5-P_6$. By repeating these calculations over a range of temperatures we find the above properties as a function of temperature and locate the phase transition. In addition we obtain the specific heat by numerically differentiating the entropy.

To study the phase diagram of the three-state Potts model we have taken the limit of the cubic model in which $\mathcal{J}$ tends to zero (zero bilinear interaction), while holding $\eta\mathcal{J}$ finite. Details on the application of the Kikuchi approximation to these models and the solutions of the ensuing coupled equation by the natural iteration process are found elsewhere.[7] Here we summarize our results and draw some conclusions about the phase diagram for the cubic model.

## III. RESULTS

The qualitative features of the phase diagram for the cubic model do not change much as one goes from the BPW to the Kikuchi tetrahedron approximation. In Fig. 2 we show the projection of the tricritical lines on the $D_1$-$D_2$ plane. While the anisotropy field necessary to reach tricriticality does not change much along the positive $D_1$ axis (see Table I), there is a large change along the negative $D_1$ axis. Also, as seen from Table I and Fig. 3, the tricritical value of the quadrupolar pair interaction $\eta_c$ for $\vec{D}=0$ is considerably larger in the Kikuchi than in the BPW approximation.

A cross section of the cubic model's phase diagram in the $D_1$-$T$ plane is shown in Fig. 4. We note that as we go to successively better approximations the transition temperature $T(D_1)$ decreases. To ascertain the accuracy of the transition temperatures in the Kikuchi approximation, we took the Ising spin-$\frac{1}{2}$ limit of the cubic model, i.e., we let $D_1\rightarrow\infty$ and found $kT_c/\mathcal{J}=10.02$. This compares rather well with best estimates from series expansions of the transition temperature $kT_c/\mathcal{J}=9.76$. In the Bethe-Peierls-Weiss approximation we found $kT_c/\mathcal{J}=10.97$ for $D_1\rightarrow\infty$, while in the MF approximation it was $kT_c/\mathcal{J}=12$.

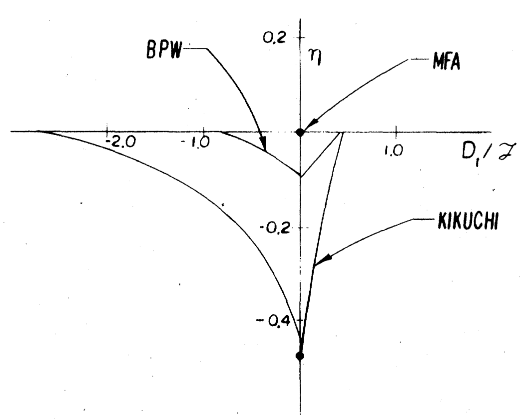



FIG. 3. Plot of the anisotropy fields $D_{1c}$ necessary to achieve tricriticality as a function of the quadrupolar coupling $\eta$ in the BPW and Kikuchi approximations.

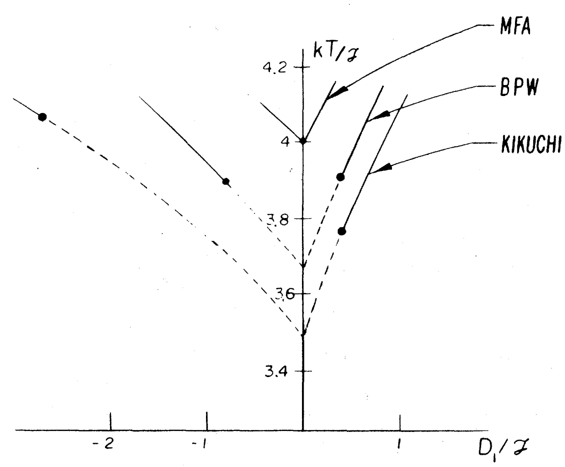



FIG. 4. Cross section of Fig. 1 with the single-ion anisotropy $D_2=0$. This shows the variation of the transition temperature with anisotropy $D_1$ for the cubic model with $\eta=0$ in the mean-field, BPW, and Kikuchi approximations. The temperature and anisotropy are measured in terms of the coupling constant $\mathcal{J}$. Solid lines represent continuous phase transitions; dashed lines, first-order phase transitions. The tricritical values of $kT/\mathcal{J}$ and $D_1/\mathcal{J}$ are given in Table I.

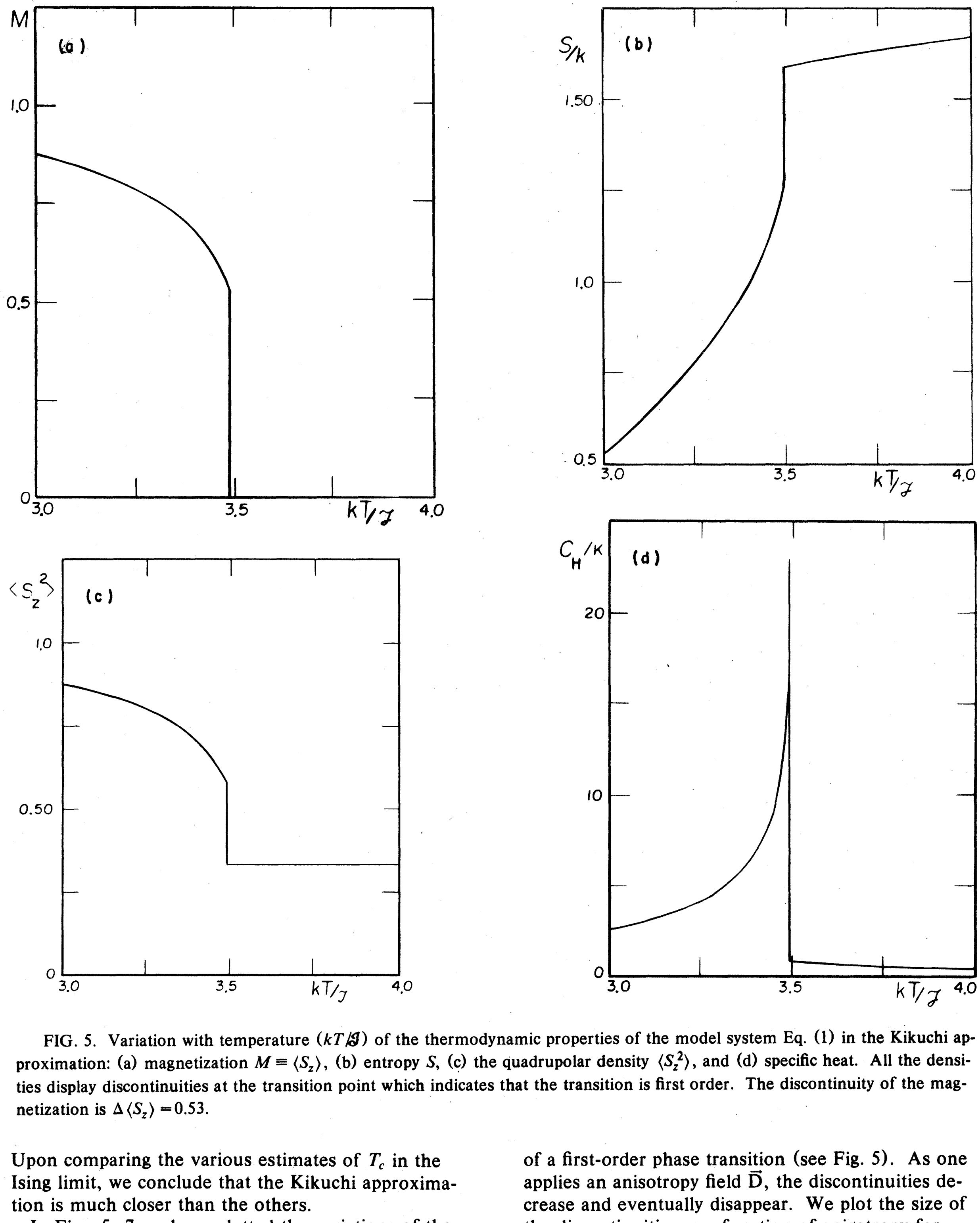



FIG. 5. Variation with temperature ($kT/\mathcal{J}$) of the thermodynamic properties of the model system Eq. (1) in the Kikuchi approximation: (a) magnetization $M \equiv \langle S_z \rangle$, (b) entropy $S$, (c) the quadrupolar density $\langle S_z^2 \rangle$, and (d) specific heat. All the densities display discontinuities at the transition point which indicates that the transition is first order. The discontinuity of the magnetization is $\Delta \langle S_z \rangle = 0.53$.

Upon comparing the various estimates of $T_c$ in the Ising limit, we conclude that the Kikuchi approximation is much closer than the others.

In Figs. 5–7 we have plotted the variations of the thermodynamic parameters with temperature and anisotropy. In zero field $\vec{D} = 0$, all the densities demonstrate sizable discontinuities which are the signatures of a first-order phase transition (see Fig. 5). As one applies an anisotropy field $\vec{D}$, the discontinuities decrease and eventually disappear. We plot the size of the discontinuities as a function of anisotropy for $D_1 > 0$ and $D_2 = 0$ in Fig. 6 and along the ray $D_1 = \sqrt{3} D_2 (D_1 > 0)$ in Fig. 7. The value at which the discontinuities disappear is the tricritical point. By re-

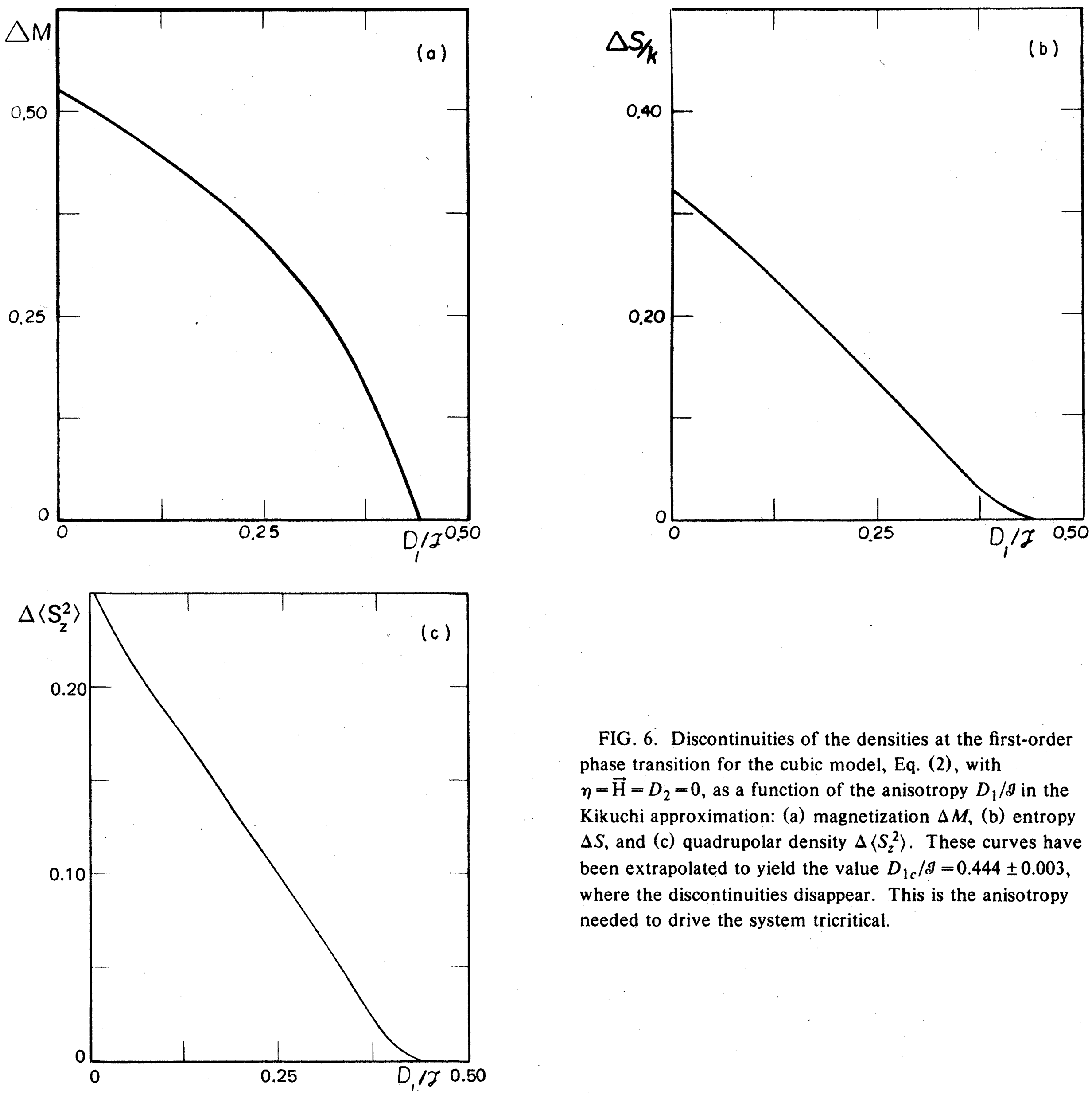



FIG. 6. Discontinuities of the densities at the first-order phase transition for the cubic model, Eq. (2), with $\eta=\vec{H}=D_2=0$, as a function of the anisotropy $D_1/\mathcal{J}$ in the Kikuchi approximation: (a) magnetization $\Delta M$, (b) entropy $\Delta S$, and (c) quadrupolar density $\Delta\langle S_z^2\rangle$. These curves have been extrapolated to yield the value $D_{1c}/\mathcal{J}=0.444\pm0.003$, where the discontinuities disappear. This is the anisotropy needed to drive the system tricritical.

peating the calculations for different directions in the $D_1$-$D_2$ plane we have been able to determine the line of tricritical points shown in Fig. 2. Owing to the threefold symmetry of the phase diagram[1] it is only necessary to investigate $\frac{1}{6}$ of the entire space, e.g., those directions with $D_1>0$ and between $D_2=0$ and $D_2=\sqrt{3}D_1$.[7] In Fig. 8 we show how the discontinuities of the densities decrease as one introduces antiferroquadrupolar coupling between the spins. When the ratio $\eta$ reaches the value $\eta_c=-0.452(\vec{D}=0)$ the discontinuities disappear and the phase transition is continuous. In Fig. 3 we show how the critical value of the ratio $\eta_c$ changes as one applies anisotropy.

The trend that emerges from our analysis of the cubic model by using the mean-field, Bethe-Peierls-Weiss, and Kikuchi approximations is as follows. As we increase the number of components of the spin which become simultaneously critical those approximations which do not take proper account of correlations between spin sites become increasingly poorer. This is substantiated from our work as can be seen from Fig. 2 and Table I. Along the positive $D_1$ axis ordering is favored along two directions $\pm z$; it is along these directions that critical fluctuations occur when the transition is critical or near tricriticality. The field $D_{1c}$ necessary to obtain tricriticality along

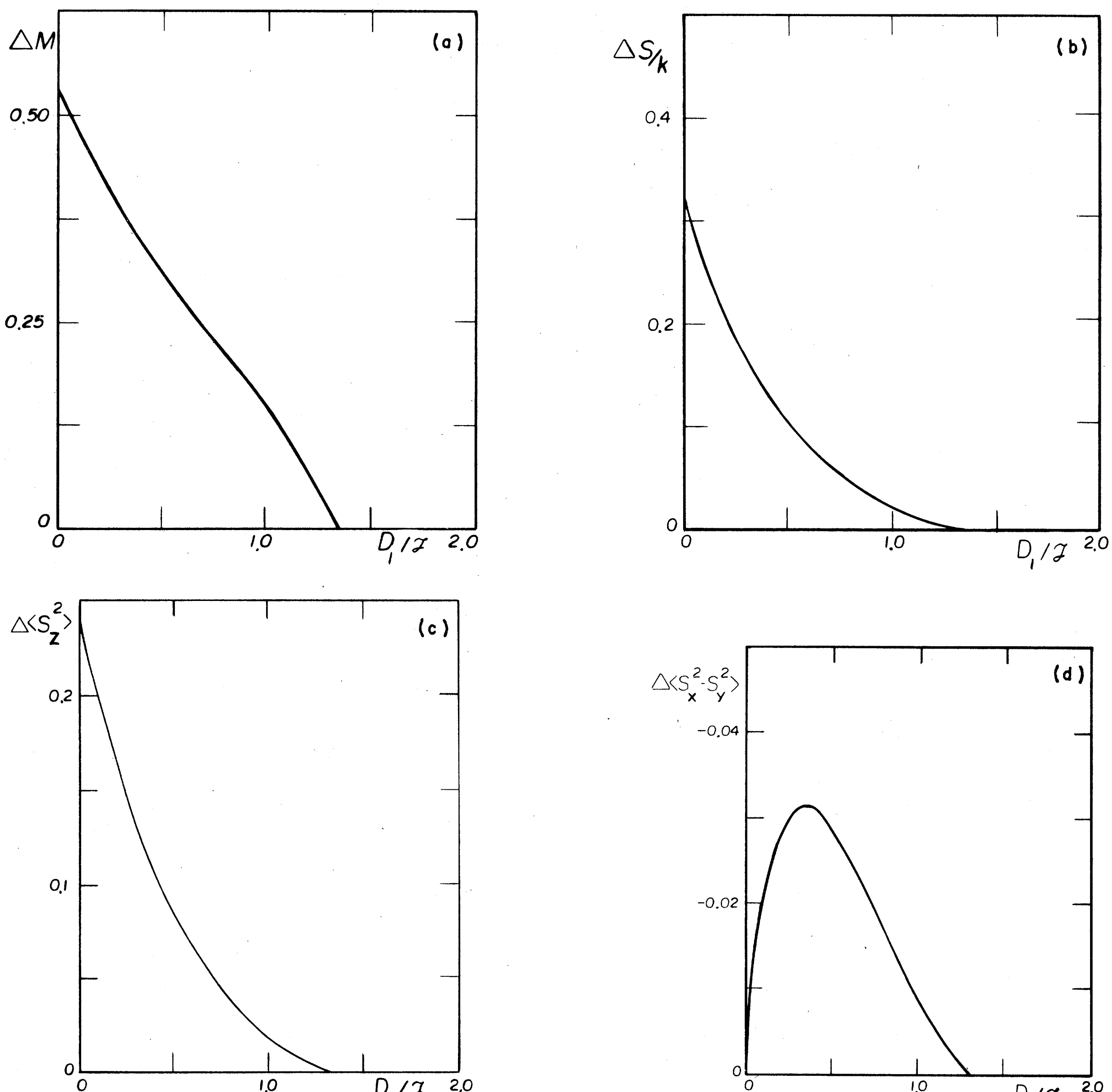
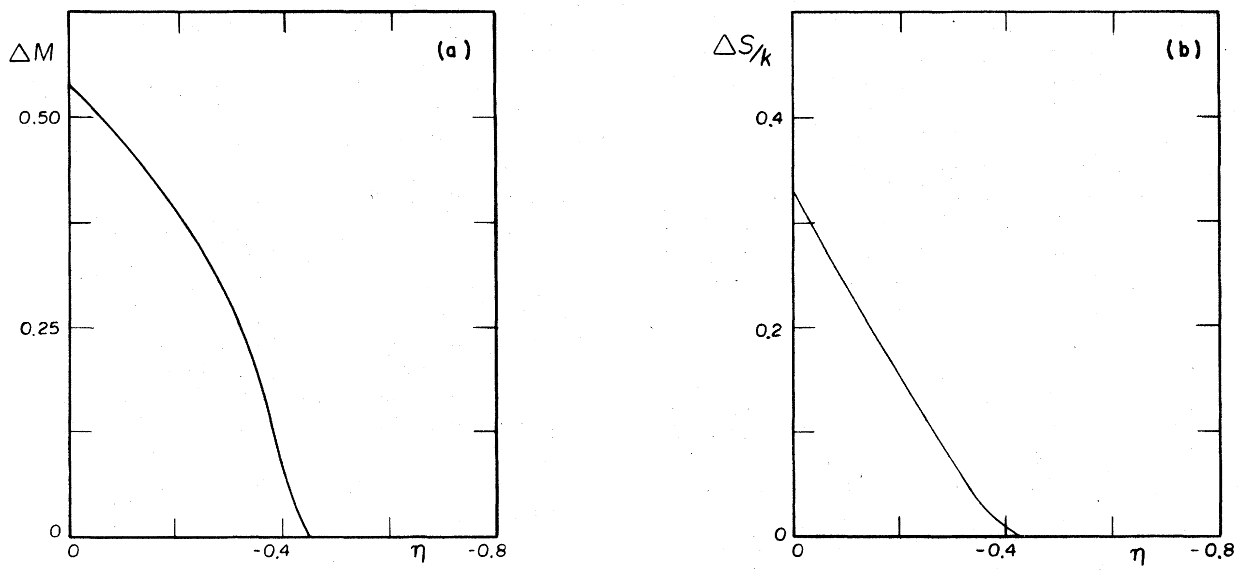



FIG. 7. Discontinuities of the densities at the first-order phase transition for the cubic model Eq. (2) with $\eta=\vec{H}=0$ and $D_2=\sqrt{3}D_1$, as a function of the anisotropy $D_1/\mathcal{J}$ in the Kikuchi approximation: (a) magnetization $\Delta M$, (b) entropy $\Delta S$, (c) quadrupolar density $\Delta\langle S_z^2\rangle$, and (d) the biaxial density $\Delta\langle S_x^2-S_y^2\rangle$. From these curves we extrapolate the tricritical value of $D_{1c}/\mathcal{J}=1.35\pm0.02$.

the positive $D_1$ axis does not change much in going from the BPW to the Kikuchi approximation. However, as we noted before, along the negative $D_1$ axis as well as along the equivalent rays $D_2=\pm\sqrt{3}D_1$, the fields necessary for tricriticality $\vec{D}_c$ change considerably on going from the BPW to the Kikuchi approximation, (see Fig. 2). The reason for this is that along these rays the cubic model spins can order in any of the *four* directions $\pm x$, $\pm y$, and there are twice as many directions along which critical fluctuations occur. Finally at the origin $\vec{D}=0$, critical fluctuations build up along *six* directions $\pm x$, $\pm y$, and $\pm z$ as we increase the size of the antiferroquadrupolar pair interaction $\eta<0$. For this extreme case, we find the critical value of the antiferroquadrupolar interaction $\eta_c$ is much larger in the Kikuchi approximation than



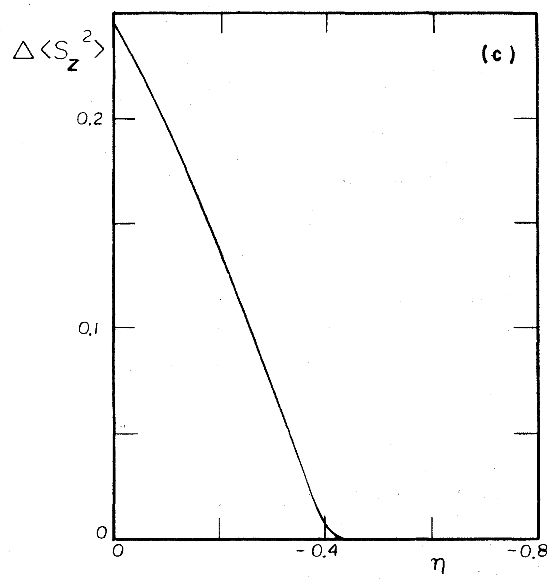




FIG. 8. Discontinuities of the densities at the first-order phase transition of the cubic model Eq. (2) with $\vec{H}=\vec{D}=0$ as a function of the quadrupolar coupling $\eta$ in the Kikuchi approximation: (a) magnetization $\Delta M$, (b) entropy $\Delta S$, and (c) quadrupolar density $\Delta\langle S_z^2\rangle$. From these curves we extrapolate the amount of antiferroquadrupolar coupling needed to drive the system tricritical to be $\eta_c=-0.452\pm0.002$.

in the BPW. As there are three times as many directions along which fluctuations build up at the origin, we readily understand why the MF and BPW approximations grossly underestimate the value of $\eta_c$. The Kikuchi approximation using a tetrahedron as the basic cluster provides a much better estimate of $\eta_c$ in this case.

These results on the *discrete*-spin cubic model are in agreement with the results on $n$-component *continuous*-spin models. For the continuous-spin models it has been shown that those with components $n\leqslant 3$ undergo continuous phase transitions while those with $n\geqslant 4$ do not, the conjecture being that they have discontinuous phase transitions.[8] Near the origin $\vec{D}=0$, the cubic model has six components which undergo critical fluctuations, and we expect a region of first-order phase transitions. As one goes out from the origin along the negative $D_1$ axis or along the rays $D_2=\pm\sqrt{3}D_1(D_1>0)$, two of the six fluctuating components of the spin are suppressed, and one expects a continuous transition as one approaches an effective $n=4$ model (which for the discrete-spin model reduces to two spin-$\frac{1}{2}$ Ising models). If one goes out from the origin along the positive $D_1$ axis or along the rays $D_2=\pm\sqrt{3}D_1$ $(D_1<0)$, four components of the fluctuating spins are suppressed, and past a point the model system has effectively $n=2$. In this case one expects, and finds in all the approximations used, a continuous phase transition.

When one includes an antiferroquadrupolar pair interaction and keeps $\vec{D}=0$, there is eventually a crossover from a discontinuous to a continuous phase transition at $\eta_c$. In this case the changeover is not caused by a reduction in the number of fluctuating components but rather by the competition between the ferromagnetic bilinear interaction which produces the *ferromagnetically* ordered phase and the antiferroquadrupolar pair interaction, which, if large enough, stabilizes an antiferroquadrupolar ordered state.[9]

## IV. SUMMARY

We have seen that although the qualitative features of the phase diagrams do not drastically change on going from the BPW to the Kikuchi approximation, the details do. Most striking are the changes in anisotropy fields and biquadratic coupling energy necessary to drive the first-order phase transition of the cubic model to tricriticality. As we have shown, the size of the change is proportional to the number of components of the spin undergoing critical fluctuations. More detailed information about the phase transition and critical properties of the cubic model can be obtained from a real-space renormalization-group analysis of the model. Up to the present, this has been done only for one- and two-dimensional lattices.[10] For three dimensions the analysis is indeed difficult.

The three-state Potts model can be viewed as the cubic model in the limit $\mathcal{J}\rightarrow 0$, while holding $\kappa=\eta\mathcal{J}$ finite. On comparing the phase transition behavior of these two models, it is interesting to note that for the cubic model the region of first-order phase transitions (coexistence volume) increases as one goes to better approximations, while for the three-state Potts model it decreases.[5]

Finally, the cubic model was developed to study the phase transitions in a series of rare-earth compounds.[1] These compounds have rather unique single-ion ground states which are nearly sixfold degenerate. In addition these compounds have type-II antiferromagnetic order on a fcc lattice. Our investigations up to the present have only dwelt on the unique nature of the single-ion states and have not considered the additional complication of the antiferromagnetic ordering. We are presently studying the cubic model on the fcc lattice with antiferromagnetic interactions.

## ACKNOWLEDGMENTS

We would like to thank Professor M. E. Fisher for pointing out the usefulness of the Kikuchi approximations. The hospitality and use of the computing facilities of the Laboratoire de Physique des Solides of the Université de Paris-Sud in Orsay is gratefully acknowledged. Research supported in part by the NSF under Grant No. DMR 76-82363. This study is based in part on a thesis presented by J. J. S. in partial fulfillment of the requirements for the degree of Doctor of Philosophy at New York University.

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