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Sequences of second-order, first-order, and reentrant phase transitions in anisotropic systems with cubic symmetry

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The infinite-range magnetization equation is solved for a three-component spin system involving cubic anisotropy of fourth, sixth, eighth, and tenth degree. Six possible types of ordered phases are found. Two of them, (X) and $(X=Y=Z)$ are well known and are characterized by ordering along an axis and along a body diagonal, respectively. In one of the four new phases, $(X=Y)$, the resultant magnetization is directed along a face diagonal, say, in the XY plane, while in the remaining three "generic" phases, (XY), $(X=Y,Z)$, and (XYZ), the magnetization vector rotates continuously in a plane and in space, respectively, upon varying the temperature. It is shown that the longest possible nonreentrant sequences of second-order transitions for appropriate choices of parameters are $I\rightarrow(X)\rightarrow(XY)\rightarrow(X=Y)\rightarrow(X=Y,Z)\rightarrow(X=Y=Z)$ and $I\rightarrow(X)\rightarrow(XY)\rightarrow(XYZ)\rightarrow(X=Y=Z)$, for the eighth- and tenth-order Hamiltonians, respectively. The feasibility of reentrant sequences, in which each one of the above-mentioned phases appears twice, at two different temperature (and total magnetization) ranges, as well as of first-order transitions from the isotropic phase into each one of the six ordered phases, were demonstrated.

I. INTRODUCTION

Spin Hamiltonians of increasing complexity keep appearing in a large variety of contexts.[1-3] An impressive number of approaches to the treatment of systems including higher than bilinear terms in the spin Hamiltonian as well as of certain classes of anisotropic spin Hamiltonians have been developed. While the Landau theory[4-6] and microscopic mean-field theory[7-18] have been primarily used to determine the global structure of the phase diagram, the renormalization group (RG) technique has been exploited for a detailed description of the critical point and calculation of the critical exponents.[4,19-24] In most of these works only the low-order anisotropic terms (quadratic and quartic) have been taken into account. The basis for neglecting the terms of higher order lies in the concept of universality and irrelevant variables, which is central to the RG theory: it is well known that including these terms in the Landau-Ginsburg-Wilson Hamiltonian does not usually affect the critical properties.[19,21,25,26] One of the most intensively studied systems, in this context, is the n-component quartic-spin model with cubic anisotropy. Applying the Landau theory it is found that only one ordered phase is possible for a system with a given set of coefficients in the Landau expansion. The type of ordering depends on the sign of the anisotropic term. When this sign is positive the ordering is along an axis, while for a negative sign the resultant spin is directed along the main diagonal. In both cases Landau theory predicts the transition from the disordered phase to be of second order. RG theory confirms the structure of the phase diagram but predicts the transition to occur via a tetracritical and a bicritical point for these two types of ordering, respectively.[4,5,22]

For a long time these results were considered as complete and reliable and the n-component quartic model served as a classical testing ground for modern methods in the theory of phase transitions and critical phenomena. However, very recently, the completeness of the picture presented above has been seriously challenged. Studying the two-component vector model with cubic symmetry in the framework of the "old fashioned" Landau theory, Galam and Birman have shown that including sixth- and eighth-degree terms in the free-energy expansion can have drastic effects on the phase diagram.[27-30] These terms generate an additional symmetry breaking, giving rise to a new low-symmetry phase, with an order parameter continuously rotating in the XY plane as a function of the temperature. This phase was called by Galam and Birman "generic." By analyzing the free-energy expression they concluded that for some ranges of parameters of the Landau expansion the "generic" phase is the most stable ordered phase. The transition from the isotropic to the generic phase is always of first order. Applying group theoretical considerations,[31] Galam and Birman[29] argued that terms of order higher than eighth do not create new symmetry breakings and therefore do not change the general structure of the phase diagram. It was found[30] that experimental data[32,33] on the first-order ferroelectric transition in tetragonal rare-earth molybdate $Tb_2(MoO_4)_3$ fit the proposed theory. This was taken by Galam and Birman as an argument that disagreement between predictions of RG and Landau theory in this case should be resolved in favor of the latter.

An analysis of the types of phases arising in the three-component system of cubic symmetry with terms up to eighth order, was carried out within the framework of Landau theory, by Gufan and Sakhnenko.[35-37] They pointed out the existence of five phases which we denote by symbols, listing the nonvanishing magnetization components and specifying equalities among them, when they exist. These five phases are as follows:

(X), in which the magnetization is directed along one of the three Cartesian axes.

($X=Y$), in which the magnetization is directed along one of the face diagonals.

(XY), in which the magnetization points in an arbitrary direction within a face.

($X=Y=Z$), in which the magnetization is directed along a body diagonal.

($X=Y,Z$), in which the magnetization points in an arbitrary direction within one of the diagonal planes.

Note that the cubic symmetry is not totally broken in any of the above five phases. One can show that the sixth possible phase (XYZ), in which the cubic symmetry is completely broken, cannot be obtained as the stable phase of the eighth-order Landau model, for any set of Hamiltonian parameters.

In the present paper we examine the three-component spin system with cubic anisotropy containing terms up to tenth order. Our primary aims are to obtain the (XYZ) phase and to investigate some characteristic sequences of phases arising upon variation of the temperature. This cannot be done in a fully consistent way within the Landau theory because this theory involves a truncation of the entropy[15] and a postulation of the temperature dependence of the expansion coefficients which is only valid in the vicinity of the highest second-order transition temperature. Our analysis is therefore carried out using the microscopic mean-field theory (MMFT) which is free of these drawbacks and enables the description of ordered phases in the whole range of change of the order parameter.

The extension of standard MMFT to study spin Hamiltonians more complex than the isotropic Heisenberg Hamiltonian is based on the equivalence between MMFT and the exact treatment of the appropriate infinite-range spin Hamiltonian. The general isotropic spin Hamiltonian was considered in Ref. 7. The general anisotropic Heisenberg Hamiltonian was investigated by Gilmore.[34] Lee and co-workers[8–11] studied the static and dynamic properties of the infinite-range anisotropic Heisenberg Hamiltonian with uniaxial symmetry. These studies were extended in Refs. 12 and 13 to the general uniaxial infinite-range Hamiltonian.

A generalization of the magnetization equation to an arbitrary anisotropic spin Hamiltonian was derived in Ref. 14. The general infinite-range Hamiltonian is written in the form

$$\mathcal{H}=NH(\hat{S}_x,\hat{S}_y,\hat{S}_z)\ , \tag{1}$$

where N is the number of particles and

$$\hat{S}_i=\sum_{j=1}^{N}\hat{S}_{ij}/N,\quad i=x,y,z\ .$$

The magnetization equation for the Hamiltonian in Eq. (1) was shown to be

$$S=-\frac{\nabla_S H}{|\nabla_S H|}\sigma B_\sigma(\beta\sigma|\nabla_S H|)\ . \tag{2}$$

Here $S_i=\langle\hat{S}_i\rangle$ is the thermal average of $\hat{S}_i$,

$$\nabla_S H=\mathbf{i}\frac{\partial H}{\partial S_x}+\mathbf{j}\frac{\partial H}{\partial S_y}+\mathbf{k}\frac{\partial H}{\partial S_z}\ ,$$

σ is the elementary spin, B_σ is Brillouin's function, and $\beta=1/kT$. An equivalent form of Eq. (2) is

$$\frac{1}{S_x}\frac{\partial H}{\partial S_x}=\frac{1}{S_y}\frac{\partial H}{\partial S_y}=\frac{1}{S_z}\frac{\partial H}{\partial S_z}\ , \tag{3a}$$

$$S=\sigma B_\sigma(\beta\sigma|\nabla_s H|)\ , \tag{3b}$$

where $S^2=S_x^2+S_y^2+S_z^2$.

The solution of the magnetization equation (3) in its general form was discussed in Ref. 16 and the procedure was applied to a class of anisotropic spin Hamiltonians of monoclinic symmetry, containing both quadratic and quartic terms. The types of phases and the location and nature of the phase transitions were determined and the corresponding phase diagrams were constructed. This work was extended in Ref. 17 to study long sequences of order-order transitions for the same class of Hamiltonians. It was found that sequences containing as many as six different phases can exist for a certain range of Hamiltonian parameters.

In the present paper we apply the technique developed in Refs. 16 and 17 to study ordered phases and phase sequences in spin systems with high-order cubic anisotropy. In the next section the magnetization equation associated with the system studied is formulated and solved and the possible ordered phases are established. Using this formalism we show that for the spin Hamiltonian with cubic symmetry it is necessary to include terms of tenth order in the spin operators in order to obtain the phase (XYZ) in which the symmetry is completely broken. The ($X=Y,Z$) phase is introduced by the sixth-order anisotropy, while (XY) (the "generic" two-dimensional phase[30]) by eighth-order anisotropy. The choices of Hamiltonian parameters giving rise to the various sequences of second-order transitions are discussed in Sec. III. The longest nonreentrant sequences involving second-order transitions are $I\rightarrow(X)\rightarrow(XY)\rightarrow(XYZ)\rightarrow(X=Y=Z)$ and $I\rightarrow(X)\rightarrow(XY)\rightarrow(X=Y)\rightarrow(X=Y,Z)\rightarrow(X=Y=Z)$. The feasibility of reentrant sequences as well as of sequences involving first-order transitions from the isotropic phase into each one of the ordered phases, is demonstrated. The analysis is illustrated by means of an appropriate set of numerical results.

II. THE MAGNETIZATION EQUATION FOR THE VARIOUS PHASES

There are five groups O_h, T_h, O, T_d, and T allowed by the Lifshitz condition[35] corresponding to all the crystallographically possible transitions with a three-component order parameter. From these only the group O_h allows for six-ordered phases: (X), ($X=Y$), (XY), ($X=Y=Z$), ($X=Y,Z$), and (XYZ). The entire rational basis of invariants constructed from the components of the order parameter which in our case is the magnetization vector, consists of three functions

$$I_1=S_x^2+S_y^2+S_z^2=S^2 ,$$

$$I_2=S_x^2S_y^2+S_y^2S_z^2+S_z^2S_x^2, \quad I_3=S_x^2S_y^2S_z^2 .$$

Writing the Hamiltonian under consideration in terms of these invariants we obtain

$$H=aS^2+bS^4+cS^6+dS^8+fS^{10}+e(S)I_2 +g(S)I_3+h(S)I_2^2+nI_2I_3 , \tag{4}$$

where $e(S)$, $g(S)$, and $h(S)$ are polynomials in S^2 of the appropriate order. Using the formalism discussed in Ref. 16 we derive

$$\frac{1}{S_x}\frac{\partial H}{\partial S_x}=2a+4bS^2+6cS^4+8dS^6+10fS^8+e'(S)/SI_2+2e(S)(S_y^2+S_z^2) +g'(S)/SI_3+2g(S)S_y^2S_z^2+h'(S)/SI_2^2+4h(S)I_2(S_y^2+S_z^2)+2nS_y^2S_z^2(2S_x^2S_y^2+2S_x^2S_z^2+S_y^2S_z^2) , \tag{5}$$

with equivalent expressions in the y and z directions. Corresponding to Eq. (3a) we get the following two equations:

$$(S_y^2-S_x^2)[2e(S)+2g(S)S_z^2+4h(S)(S_x^2S_y^2+S_x^2S_z^2+S_y^2S_z^2)+2nS_z^2(2S_x^2S_y^2+S_x^2S_z^2+S_y^2S_z^2)]=0 , \tag{6a}$$

$$(S_z^2-S_x^2)[2e(S)+2g(S)S_y^2+4h(S)(S_x^2S_y^2+S_x^2S_z^2+S_y^2S_z^2)+2nS_y^2(2S_x^2S_z^2+S_x^2S_y^2+S_y^2S_z^2)]=0 . \tag{6b}$$

The solutions for phases with three nonzero components are obtained using Eqs. (6a), (6b), and 3(b). The (XY) phases are determined from (6a), (3b), and $S_z^2=0$. The (X) phase is obtained by solving Eq. (3b) with $S_y^2=S_z^2=0$. Depending on the degree of the Hamiltonian one gets different phase diagrams.

(A) Sixth-degree anisotropy. Here

$$e(S)=e_0+e_1S^2, \quad g=g_0\neq 0, \quad d=f=h=n=0 .$$

Out of the six possible phases, the following four are obtained:

$$\begin{aligned} &(X=Y=Z)\text{:} \quad S_x^2=S_y^2=S_z^2=S^2/3 , \\ &(X=Y,Z)\text{:} \quad S_x^2=S_y^2=-e(S)/g, \quad S_z^2=S^2-2s_x^2 , \\ &(X=Y)\text{:} \quad S_x^2=S_y^2=S^2/2, \quad S_z^2=0 , \\ &(X)\text{:} \quad S_x^2=S^2, \quad S_y^2=S_z^2=0 . \end{aligned} \tag{7}$$

(B) Eighth-degree anisotropy. In this case

$$e(S)=e_0+e_1S^2+e_2S^4, \quad g(s)=g_0+g_1S^2 ,$$

$$h=h_0\neq 0, \quad f=n=0 .$$

Comparing to case (A) one new phase, (X,Y) is introduced. In this phase

$$S_x^2=S^2/2(1+\sqrt{1+2e(S)/hS^4}, \quad S_y^2=S^2-S_x^2, \quad S_z^2=0 . \tag{8}$$

Furthermore, the expressions for the components in the $(X=Y,Z)$ phase are modified into

$$\begin{aligned} &S_x^2=S_y^2=\frac{g(S)+4hS^2}{12h}\left[1+\left[1+\frac{24he(S)}{[g(S)+4hS^2]^2}\right]^{1/2}\right] , \\ &S_z^2=S^2-2S_x^2 , \end{aligned} \tag{9}$$

while the $(X=Y)$, $(X=Y=Z)$, and (X) phases are defined as above.

(C) Tenth-degree anisotropy. In this case

$$e(S)=e_0+e_1S^2+e_2S^4+e_3S^6 ,$$

$$g(S)=g_0+g_1S^2+g_2S^4 ,$$

$$h(S)=h_0+h_1S^2 ,$$

$$n=n_0\neq 0 .$$

Tenth-degree anisotropic terms introduce a sixth, lowest-symmetry phase, (X,Y,Z). The three components are the three solutions of

$$S_i^6+S_i^4(-S^2)+S_i^2\beta(S)+\delta(S)=0 , \tag{10}$$

which is a cubic equation in S_i^2. The coefficient of S_i^4 guarantees that $S_x^2+S_y^2+S_z^2=S^2$. The other two coefficients are given by

$$\begin{aligned} &\beta(S)=-g(S)/n , \\ &\delta(S)=[e(S)+2h(S)\beta(S)]/n . \end{aligned} \tag{11}$$

From the known properties of the solution of the cubic equation it follows that

$$\begin{aligned} &\beta(S)=S_x^2S_y^2+S_y^2S_z^2+S_z^2\cdot S_x^2 , \\ &\delta(S)=-S_x^2S_y^2S_z^2 . \end{aligned} \tag{12}$$

The components of the $(X=Y,Z)$ phase satisfy $S_x^2=S_y^2$ and

$$S_x^6+S_x^4[3/(5n)](2h-nS^2) -S_x^2(g+4hs^2)/(5n)-e/(5n)=0 . \tag{13}$$

The (XY), $(X=Y)$, $(X=Y=Z)$, and (X) phases are defined as above.

III. PHASE SEQUENCES FOR DIFFERENT CHOICES OF THE HAMILTONIAN PARAMETERS

We take advantage of the fact that S increases monotonically upon lowering the temperature, to carry out the analysis of the phase sequences generated by different choices of the Hamiltonian parameters as follows. Choosing S we determine the orientation of the magnetization using the appropriate equations of the previous section, discarding the nonphysical solutions with $S_i^2<0$; this enables the evaluation of $|\nabla_S H|$ from which the temperature is obtained by inverting Eq. (3b). When more than one type of solution is obtained for a given temperature the equilibrium state is determined by means of the free energy. Before embarking on a detailed numerical computation we present a qualitative discussion to motivate the choices of the Hamiltonian parameters.

A. Nonreentrant second-order phase sequences

In the present section we concentrate on generating nonreentrant sequences involving second-order transitions among all the phases which can arise as a consequence of the competition between the different anisotropic terms in the Hamiltonian.

We shall assume that $a<0$. b is assumed to be positive. It could also be taken to be negative provided that it is large enough to guarantee that the transition from the isotropic state, at $kT_c=|a|/2$, is of second order.

Retaining only the lowest- (fourth-) order anisotropic term in the Hamiltonian it has been found that for $e=e_0>0$ the only possible ordered phase is (X), for which the anisotropy energy is zero, whereas for $e_0<0$ the lowest possible anisotropy energy is obtained for the $(X=Y=Z)$ phase.

Since the lowest-order anisotropic term is dominant at high temperatures (for which all the magnetization components are small), a continuous transition from the isotropic phase will always be into either the (X) or the $(X=Y=Z)$ phase, depending on the sign of e_0.

The sixth-order Hamiltonian contains two types of anisotropic terms, one of which has an S-dependent coefficient. Thus, one might expect the opportunity for competition, giving rise to different phases at different temperatures. In order to specify the possible sequences of continuous transitions it is useful to consider the $T\to 0$ limit, in which the stable phase is determined by the Hamiltonian itself. At this limit the magnetization is saturated, $S=\frac{1}{2}$. The energies of the various phases are

$$E_{(X)}=0\ ,$$

$$E_{(X=Y)}=e/64\ ,$$

$$E_{(X=Y,Z)}=-(e/g)^2(e+g/4)\ ,$$

$$E_{(X=Y=Z)}=(e+g/36)/48\ ,$$

where $e=e(S=1/2)$. The requirement that $S_i^2\geq 0$ introduces the restriction $0\leq -e/g\leq\frac{1}{8}$ at the $(X=Y,Z)$ phase. No similar restrictions apply to the other three phases. Comparing the energies of the phases and taking into account the restriction concerning the $(X=Y,Z)$ phase we find that the (g,e) plane is divided into three sectors, as presented in Fig. 1. One important conclusion is that the lowest-temperature phase cannot be $(X=Y,Z)$.

In view of these remarks, the longest continuous sequences that one might anticipate are

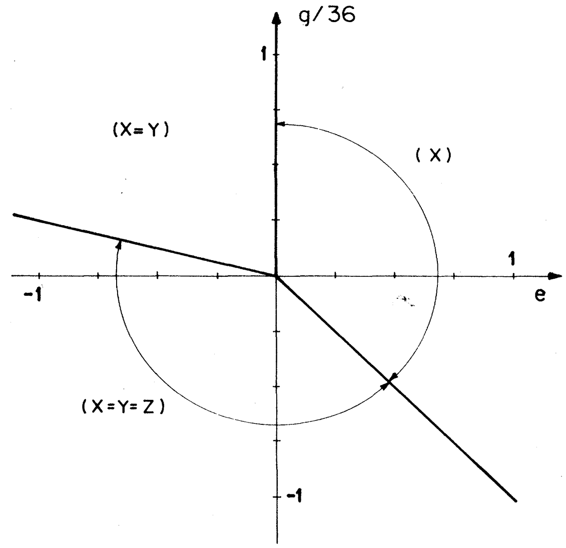

FIG. 1. Phase diagram for the sixth-order Hamiltonian, at $T=0$.

$$(X=Y=Z)\to(X=Y,Z)\begin{smallmatrix}\nearrow (X=Y)\\ \searrow (Z)\end{smallmatrix}$$

or

$$(Z)\to(X=Y,Z)\begin{smallmatrix}\nearrow (X=Y=Z)\\ \searrow (X=Y)\end{smallmatrix}\ .$$

An extensive numerical search has provided no set of Hamiltonian parameters which gives rise to either one of these sequences as the equilibrium solution.

For the eighth-order Hamiltonian the longest nonreentrant sequence involves the phases (X), (XY), $(X=Y)$, $(X=Y,Z)$, and $(X=Y=Z)$. Inspection of Eq. (6) for $n=0$ indicates that assuming the existence of a solution with complete breaking of symmetry we immediately obtain the contradictory result $S_y^2=S_z^2$. This does not happen when $n\neq 0$, showing that the tenth-order term is required to generate the (XYZ) phase.

Assuming that the highest-temperature ordered phase is (X) $(e_0>0)$ the longest possible sequence of continuous phase transitions will be of the form

$$(X)\xrightarrow{1}(XY)\xrightarrow{2}(X=Y)\xrightarrow{3}(X=Y,Z)\xrightarrow{4}(X=Y=Z)\ . \tag{14}$$

This sequence describes the appearance of a magnetization directed along the x axis at the highest critical temperature. This magnetization grows to a value S_1, at which a y component appears and starts growing; the magnetization vector rotates in the xy plane. At a lower temperature T_2 the magnetization vector points along a face diagonal (in the xy plane). The value of the resultant magnetization at this point is S_2. The magnetization keeps growing in magnitude, without change of direction, until it reaches the value S_3 (at T_3). Upon further reduction of the temperature the magnetization rotates within a diagonal plane, reaching the direction of the body diagonal at T_4, with a resultant magnetization S_4. Below this temperature the magnetization keeps growing without change of direction.

An attempt will now be made to provide a simple choice of Hamiltonian parameters giving rise to this sequence.

The fourth- and eighth-order anisotropic terms both distinguish between the (X), (XY), and (XYZ) phases. The S dependence of e, the coefficient of the fourth-order term, enables it to change sign upon variation of the temperature. This by itself would only give rise to a transition between the (X) and $(X=Y=Z)$ phases, but the interference of the eighth-order term can result in stabilization of the intermediate phases (XY), $(X=Y)$, and $(X=Y,Z)$.

The sixth-order anisotropic term does not distinguish among the phases (X), (XY), and $(X=Y)$. This suggests that the complete sequence can be generated by means of the interplay of the fourth- and eighth-order terms only. Consequently, we set $g(S)=0$.

The following conditions are obtained for the various phase transitions.

(i) At the $(X)\rightarrow(XY)$ transition $S_y^2=0$ and $S_x^2=S^2$. Inspection of Eq. (8) indicates that this condition is satisfied when $e(S_1)=0$. At this point we note that if $h<0$, the equilibrium phase for $e(S)\geq 0$ will be $(X=Y=Z)$, at which the sequence terminates. A richer variety of phases is obtained for $h>0$, which is the choice we make from now on.

(ii) The $(X=Y)$ phase is obtained when $S_x^2=S_y^2=S^2$, i.e., $e(S_2)=-hS_2^4/2$ [Eq. (8)].

(iii) $(X=Y,Z)$ phase appears when $e(S_3)=-hS_3^4/2$ [Eq. (9)]. Comparing with the condition for the appearance of the $(X=Y)$ phase we find that $S_2=S_3$, i.e., the Z component starts to grow as soon as the Y component equals X. For a different choice of $g(S)$ one could obtain a range of temperatures for which the stable phase is $(X=Y)$. We have not explored this possibility.

(iv) The $(X=Y=Z)$ phase is obtained when $e(S_4)=-2hS_4^4/3$.

To generate the sequence (14) $e(S)$ has to start with a positive value at $S=0$, vanish at S_1, and obtain the values $-hS_2^4/2$ and $-2hS_4^4/3$ at S_2 and S_4, respectively, where $S_1<S_2<S_4$. The simplest choice which satisfies these requirements is

$$e(S)=e_0+e_1S^2 ,$$

where

$$e_0=hS_2^2S_4^2[(2S_4^2/3)-(S_2^2/2)]/(S_4^2-S_2^2) , \tag{15}$$

and

$$e_1=h[(-2S_4^4/3)+(S_2^4/2)]/(S_4^2-S_2^2) . \tag{16}$$

Note that the inequality $S_2<S_4$ suffices to make $e_0>0$ and $e_1<0$. With this choice of parameters $S_4^2=-e_0/e_1$.

A sequence of phases starting with the $(X=Y=Z)$ phase at the highest-transition temperature and developing in the opposite direction to sequence (14) can be obtained by making the following choice of Hamiltonian parameters: To start with the $(X=Y=Z)$ phase we require $e_0<0$. In order to obtain the (X) phase, $e(S_1)$ should vanish, i.e., $e_1>0$. Since in this case $S_1>S_2>S_4$ it follows from Eq. (15) that the inequalities $e_0<0$ and $e_1>0$ require that $S_2^2>S_4^2>\sqrt{3}S_2^2/2$.

We shall now introduce the tenth-order terms in the spin Hamiltonian and obtain a sequence incorporating the (XYX) phase, in which a complete breaking of the cubic symmetry is achieved. One such sequence is of the form

$$(X)\xrightarrow{1}(XY)\xrightarrow{2}(XYZ)\xrightarrow{3}(X=Y=Z) . \tag{17}$$

Inspection of Eqs. (11) and (12) indicates that we have to choose $g(S)\neq 0$ in order to obtain the (XYZ) phase. It can be shown that the choice

$$g(S)=-2h(S)S^2 , \tag{18}$$

suffices to allow the various phases in the sequence (17). To start with the (X) phase we set $e_0>0$. The condition for the transition into the (XY) phase at T_1 is the same as before, i.e., $e(S_1)=0$. In order to present a set of Hamiltonian parameters for which the transition into the (XYZ) phase takes place we point out that the magnetization components for this phase are the three solutions of the cubic equation (10). We choose the three components to be of the form

$$\begin{aligned} S_x^2&=S^2/3+q(S) ,\\ S_y^2&=S^2/3 ,\\ S_z^2&=S^2/3-q(S) , \end{aligned} \tag{19}$$

such that $q(S_2)=S_2^2/3$. At the transition into the body diagonal phase $(X=Y=Z)$ we should have $q(S_3)=0$. Substituting the expressions in Eq. (19) into Eq. (12) we obtain

$$\beta(S)=S^4/3-q^2(S) . \tag{20}$$

It follows from the above that

$$\beta(S_2)=2S_2^4/9 ,$$

and

$$\beta(S_2)=2S_2^4/9 ,$$

Using Eqs. (11) and (18) we obtain $h(S_2)=nS_2^2/9$ and $h(S_3)=nS_3^2/6$. From Eq. (12) $\delta(S_2)=0$ and $\delta(S_3)=-S_3^6/27$. Using Eq. (11) we obtain $e(S_2)=-\frac{4}{81}nS_2^6$ and $e(S_3)=-\frac{4}{27}nS_3^6$. We can write $e(S)$ and $h(S)$ in the form

$$e(S)=e_0+e_1S^2 ,$$

$$h(s)=h_0+h_1S^2 ,$$

with the constants e_0, e_1, h_0, and h_1 chosen so as to satisfy the above relations. One can check that the sign of $h(S)$ remains unchanged over all the order-order phase transitions. Since the signs of h and n are the same a competition among the various anisotropic terms requires that $n>0$.

The reversed sequence, starting with the $(X=Y=Z)$ phase as the highest temperature ordered phase, is obtained by choosing $S_1^2>S_2^2>S_3^2>S_2^2/3^{1/3}$, the last inequality resulting from the requirements $e_0<0$ and $e_1>0$.

B. Reentrant sequences

In the previous section we considered nonreentrant sequences involving second order phase transitions. The S-dependent Hamiltonian coefficients $e(S)$, $g(S)$, and $h(S)$ were taken to vary monotonically in the whole range $0\le S\le\frac{1}{2}$. In the present section we consider the possibility that nonmonotonic variation of these coefficients can give rise to reentrant behavior.

As a starting point we consider the longest sequences obtained in the previous section. For the eighth-order Hamiltonian the sequence presented by Eq. (14) involves a monotonic decrease of $e(S)$ with increasing S, i.e., decreasing temperature. If, after the low-temperature phase $(X=Y=Z)$ is reached, $e(S)$ starts increasing upon further increase of S, the sequence of phases can be reversed, eventually reaching one of the three equivalent axial phases (X), (Y), or (Z). The complete sequence obtains the form

$$(X)\xrightarrow{1}(XY)\xrightarrow{2}(X=Y)\xrightarrow{3}(X=Y,Z)\xrightarrow{4}(X=Y=Z)$$
$$\xrightarrow{4'}r(X=Y,Z)\xrightarrow{3'}r(X=Y)\xrightarrow{2'}r(XY)\xrightarrow{1'}r(X)\ , \qquad (21)$$

where r indicates a reentrant phase.

As before, each symbol represents any one of a set of symmetry-equivalent phases. Moreover, the particular phase which $r(X=Y,Z)$ stands for need not be the same as the one represented by $(X=Y,Z)$. A similar remark applies to the pair (XY) and $r(XY)$ and the pair (X) and $r(X)$. At S_1, S_2, S_3, and S_4, $e(S)$ should obtain the values derived in connection with Eq. (14). At the reentrant transition points $e(S)$ should obtain values given by expressions of the same form, with S_i replaced by S_i'. All these conditions can be satisfied by

$$e(S)=e_0+e_1S^2+e_2S^4\ ,$$

where

$$e_2>2h\ ,$$
$$e_1=-2e_2S_2^2\ ,$$
$$e_0=S_2^4(e_2-K)\ ,$$

and K is in some neighborhood of $h/2$. Choosing $\frac{1}{4}h<e_2<2h$ we obtain a sequence in whose $(X=Y,Z)$ phase S_z^2 increases upon lowering the temperature up to some value smaller than $S^2/3$, and starts decreasing back to zero, at which point the $r(X=Y)$ phase is reached. By choosing K sufficiently smaller than $\frac{1}{2}h$, the $(X=Y,Z)$ phase is not reached. The sequence reduces to $(X)\rightarrow(XY)\rightarrow r(X)$. For the tenth-order Hamiltonian the longest reentrant sequence containing the (XYZ) phase, that can be obtained when both $e(S)$ and $g(S)$ are quadratic in S^2, is of the form

$$(X)\xrightarrow{1}(XY)\xrightarrow{2}(XYZ)\xrightarrow{2'}r(XY)\xrightarrow{1'}r(X)\ .$$

An analysis along the lines presented above results in the following expressions for the Hamiltonian parameters,

$$e_1=-[S_2^2+(S_2')^2](4h/9+e_2)\ ,$$
$$e_0=-(4hS_2^4/9+e_1S_2^2+e_2S_2^4)$$
$$g_2<-2n/3\ ,$$
$$g_1=2g_2S_2^2\ ,$$
$$g_0=-S_2^2(2n/9+g_2)\ ,$$

where

$$(S_2')^2=S_2^2\left[g_2+\frac{2n}{9}\right]\Big/\left[g_2-\frac{2n}{9}\right]\ .$$

C. First-order transitions

It was pointed out above that only the axial and body-diagonal phases can be reached by a second-order transition from the isotropic phase. On the other hand, there are no symmetry restrictions concerning first-order transitions from one phase to another. In particular, each one of the six types of ordered phases can be reached via a first-order transition from the isotropic phase. To demonstrate the feasibility of such first-order transitions we point out that the value of the ordered phase magnetization at a first-order transition depends on the value of b, the coefficient of the S^4 term in the spin Hamiltonian. By making appropriate choices of b we obtained each one of the phases (XY), $(X=Y,Z)$, and (XYZ), which are not accessible via a second-order transition from the isotropic phase.

IV. ILLUSTRATIVE RESULTS

The qualitative results presented in the previous section will now be illustrated by means of appropriate numerical computations for a system with an elementary spin $\sigma=\frac{1}{2}$. The presentation follows the subdivision of the previous section.

A. Nonreentrant sequences of second-order transitions

For the eighth order Hamiltonian we obtain the $(X)\rightarrow(XY)\rightarrow(X=Y)\rightarrow(X=Y,Z)\rightarrow(X=Y=Z)$ sequence presented in Fig. 2 for the choice of Hamiltonian parameters

$$a=-15,\ b=1,\ \ c=0,\ \ d=0\ ,$$
$$e_0=2.37\times10^{-2},\ \ e_1=-0.255,\ \ h=1\ .$$

The phases shown are always those corresponding to the lowest free energy at each temperature. The reversed se-

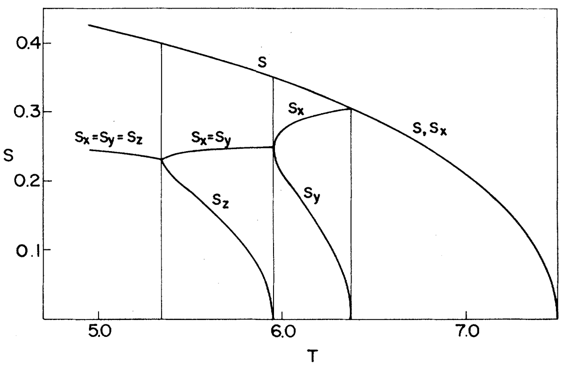

FIG. 2. The sequence $(X)\rightarrow(XY)\rightarrow(X=Y,Z)\rightarrow(X=Y=Z)$ for the eighth-order Hamiltonian.

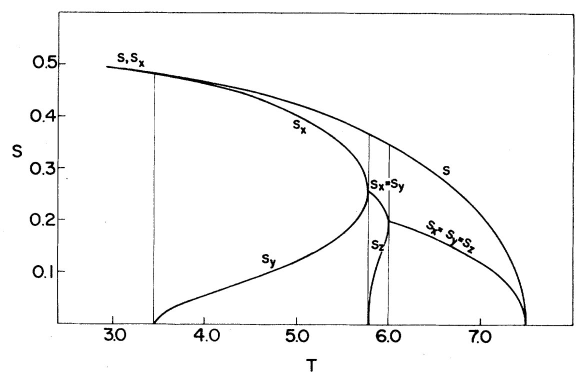

FIG. 3. The sequence $(X=Y=Z)\rightarrow(X=Y,Z)\rightarrow(XY)\rightarrow(X)$ for the eighth-order Hamiltonian.

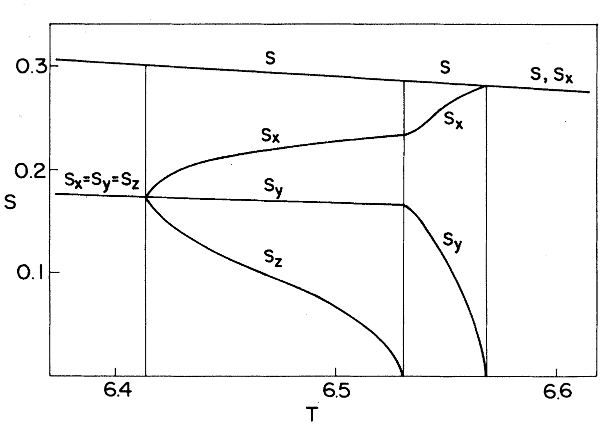

FIG. 4. The sequence $(X)\rightarrow(XY)\rightarrow(XYZ)\rightarrow(X=Y=Z)$ for the tenth-order Hamiltonian.

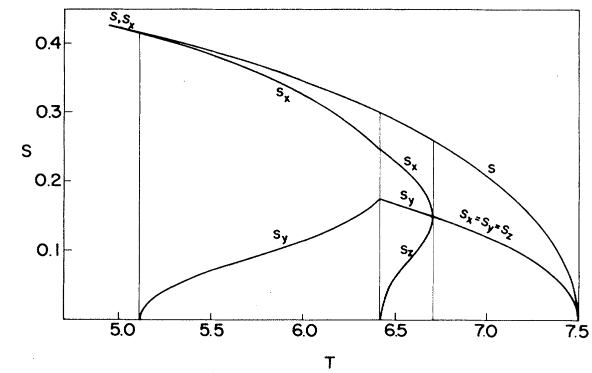

FIG. 5. The sequence $(X=Y=Z)\rightarrow(XYZ)\rightarrow(XY)\rightarrow(X)$ for the tenth-order Hamiltonian.

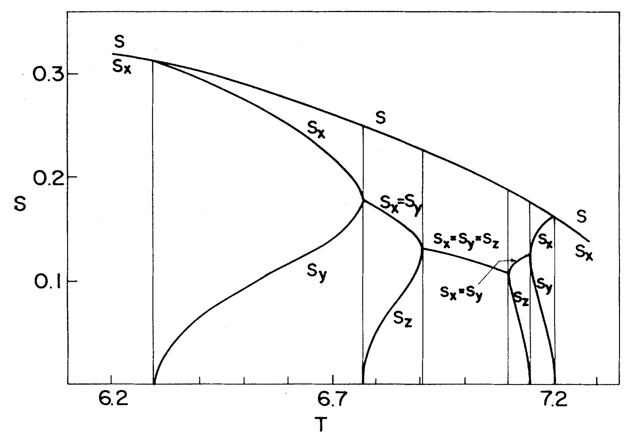

FIG. 6. The sequence $(X)\rightarrow(XY)\rightarrow(X=Y,Z)\rightarrow(X=Y=Z)\rightarrow r(X=Y,Z)\rightarrow r(XY)\rightarrow r(X)$ for the eighth-order Hamiltonian.

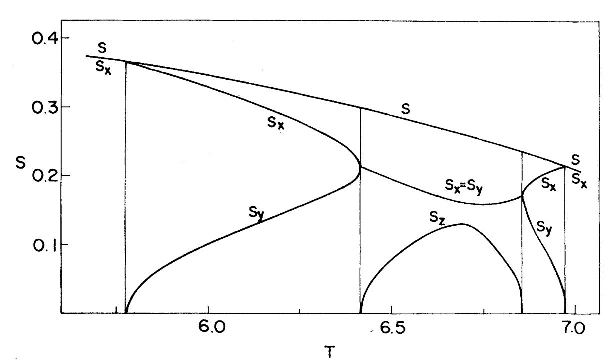

FIG. 7. The sequence $(X)\rightarrow(XY)\rightarrow(X=Y,Z)\rightarrow r(XY)\rightarrow r(X)$ for the eighth-order Hamiltonian.

quence, presented in Fig. 3, is obtained by choosing

$$e_0=-2.10\times10^{-2} \text{ and } e_1=9.01\times10^{-2} .$$

For the tenth-order Hamiltonian we obtain the sequence $(X)\to(XY)\to(XYZ)\to(X=Y=Z)$ for

$$f=0 ,$$

$$e_0=7.28\times10^{-4},\quad e_1=-9.29\times10^{-3} ,$$

$$h_0=-4.63\times10^{-2},\quad h_1=0.681 ,$$

$$g_0=0,\quad g_1=9.26\times10^{-2},\quad g_2=-1.362 ,$$

$$n=1 ,$$

and the reversed sequence for

$$e_0=-7.52\times10^{-5},\quad e_1=4.36\times10^{-4} ,$$

$$h_0=1.51\times10^{-2},\quad h_1=-5.65\times10^{-2} ,$$

$$g_0=0,\quad g_1=-3.02\times10^{-2},\quad g_2=0.113 .$$

These sequences are presented in Figs. 4 and 5, respectively.

B. Reentrant sequences

The longest sequence obtained for the eighth-order Hamiltonian

$$I\to(X)\to(XY)\to(X=Y,Z)\to(X=Y=Z)$$
$$\to r(X=Y,Z)\to r(XY)\to r(X)$$

is presented in Fig. 6 for the choice of Hamiltonian parameters $e_0=3.91\times10^{-3}$, $e_1=-0.1875$, $e_2=1.5$, $h=1$. All other parameters retain the values presented in connection with Fig. 2. For the choice of parameters $e_0=0.01296$, $e_1=-0.378$, and $e_2=2.1$ we obtain the sequence,

$$I\to(X)\to(XY)\to(X=Y,Z)\to r(XY)\to r(X)$$

which avoids the formation of the $(X=Y=Z)$ phase.

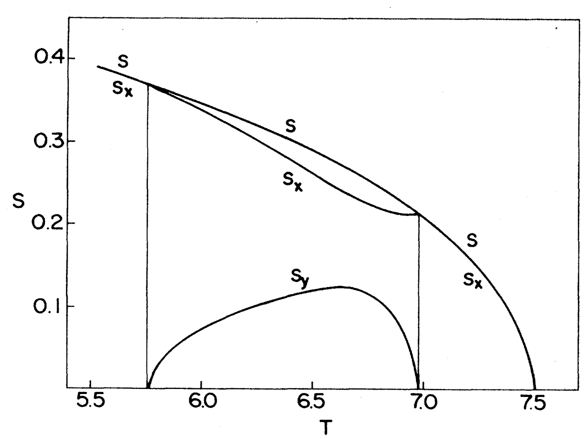

FIG. 8. The sequence $(X)\to(XY)\to r(X)$ for the eighth-order Hamiltonian.

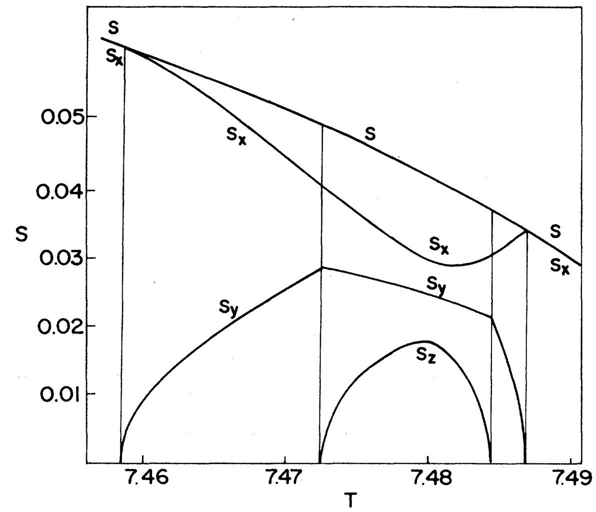

FIG. 9. The sequence $(X)\to(XY)\to(XYZ)\to r(XY)\to r(X)$ for the tenth-order Hamiltonian.

This sequence is presented in Fig. 7. The shortest reentrant sequence $I\to(X)\to(XY)\to r(X)$ is presented in Fig. 8. The parameters used are $e_0=6.075\times10^{-3}$, $e_1=-0.18$, and $e_2=1$.

For the tenth-order Hamiltonian the longest reentrant sequence containing the (XYZ) phase is

$$I\to(X)\to(XY)\to(XYZ)\to r(XY)\to r(X) .$$

This sequence is presented in Fig. 9 for the Hamiltonian parameters

$$e_0=0.447\times10^{-5},\quad e_1=-0.496\times10^{-2},\quad e_2=1 ,$$

$$g_0=0.361\times10^{-5},\quad g_1=-0.4\times10^{-2},\quad g_2=0.8 ,$$

$$h=0.6,\quad n=1 .$$

C. First-order transitions

The sequences whose highest temperature transition is a first-order transition from the isotropic phase into the

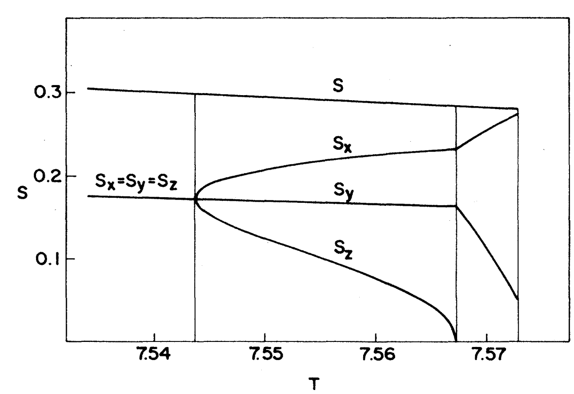

FIG. 10. The first-order transition into the (XY) phase.

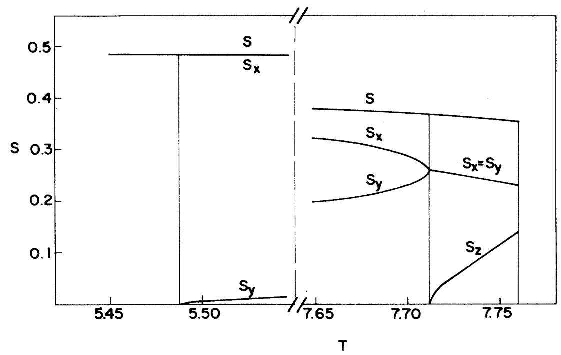

FIG. 11. The first-order transition into the $(X=Y,Z)$ phase.

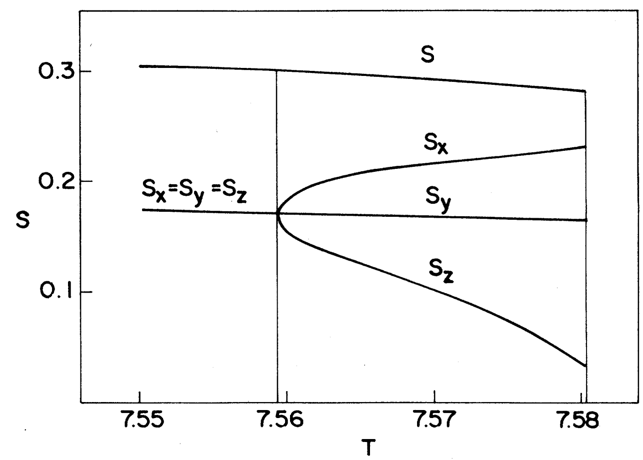

FIG. 12. The first-order transition into the (XYZ) phase.

(XY), $(X=Y,Z)$, and (XYZ) phases are presented in Figs. 10, 11, and 12, respectively. The sequences presented in Figs. 10 and 12 were obtained with the tenth-order Hamiltonian, with the parameter values equal to those in Fig. 4, except that $b=-13.5$ in Fig. 10 and $b=-13.7$ in Fig. 12. Figure 11 was obtained for the eighth-order Hamiltonian, with the parameter values equal to those in Fig. 3, except that $b=-17.4$.

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