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Low-Temperature Analysis of the ANNNI Model in an External Magnetic Field: Cascades of Phase Transitions, Complete Devil's Staircases

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The behavior of the axial next-nearest-neighbor Ising (ANNNI) model in an external magnetic field is investigated using a low-temperature expansion of the free energy. Unusual cascades of phase transitions and "complete devil's staircases," unexpected for the ANNNI model, are found.

KEY WORDS: Low-temperature expansion; phase diagram; first-order phase transition; linear programming method; "complete devil's staircase"; cascade of phase transitions.

1. INTRODUCTION

Four years ago Bak and von Boehm[1] suggested the ANNNI model as a candidate for an interpretation of the numerous spatially modulated magnetic structures of CeSb. The numerical analysis of the relevant mean-field equations at low temperatures revealed a similarity between the sequence of phases predicted by the ANNNI model and that observed experimentally. Recent experimental data on CeSb[2–6] can be understood well in the framework of the ANNNI model.[1,7–11] This substance consists of ferromagnetic layers of spins with an easy axis normal to the layers. At low enough temperatures, magnetic moments of whole layers display the periodic structure "up–up–down–down" along an easy axis. If an external magnetic field or the temperature changes, the magnetic structure turns into other modifications through first-order phase transitions.

However, some of the experimental data do not fit the framework of the conventional ANNNI model. The origin of this discrepancy is due to an oversimplified approximation of the real system by the Ising model. A

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general Hamiltonian must include in this case the competition of the magnetic and paramagnetic states of layers.

The ANNNI model is attractive to theorists since it is the simplest one with nontrivial competitive behavior. Now the phase diagram of the three-dimensional (3D) ANNNI model is known to exhibit cascades of phase transitions of the first order (see the aforementioned papers and Refs. 12 and 13). Note that on the analogous diagram of the 2D ANNNI model the infinite discrete sets of commensurate phases existing in the 3D model are replaced by floating phases.[14,15]

In the present work we investigate the phase diagram of the 3D ANNNI model in an external magnetic field emphasizing the topological structure of the phase diagram and using for this a low-temperature expansion for the free energy. This analytic method has been applied by Fisher and Selke[8,9] to describe the phase diagram of the ANNNI model in the vicinity of the competition point at low temperatures and in zero magnetic field. Their systematic analysis demonstrated an infinite number of spatially modulated phases with periodicities $2j+1$ for all integer j's. A detailed analysis of the 3D ANNNI model in an external magnetic field has been carried by Pokrovsky and Uimin.[10,11] They assumed a strong anisotropy of intra- and interlayer couplings. This condition, which is not invoked in the present paper, makes it possible to apply a high-temperature expansion of the free energy with respect to interlayer couplings over a wide region of existence of the ordered phases. The phase diagram pattern found in the framework of two above-mentioned approaches are topologically identical in zero magnetic field. In a finite field the cascades of phase transitions become more complicated (cf. Refs. 10, 11 and 8, 9). A low-temperature expansion of the free energy versus a high-temperature expansion makes it possible to investigate the phase diagram in a wider region of coupling constants. In the present paper the so-called "complete devil's staircases," unexpected for the ANNNI model, are found in a form of asymptotic expansions.

The phase diagram of the "ANNNI + field" model without the assumption of a strong anisotropy of couplings has been partly found in Refs. 12 and 13. The construction of the phase diagram between the limiting AF and $\langle 2.\bar{1}\rangle$ states[12] and between F and $\langle 2.\bar{2}\rangle$ states[13] has been described. The splitting of the last boundary, however, is found to be more complicated than described by Smith and Yeomans.[13] They did not pay attention to the existence of nontrivial excitations above the degenerate ground state. For example, in the first order of the free energy expansion double-spin flips can be most favored (see Section 7.1 for a detailed consideration).

In the next section we construct the phase diagram of the "ANNNI + field" model at $T = 0$. Also Section 2 can be considered as an introduction to the linear programming method used throughout this paper. General features of a low-temperature expansion of the free energy in systems with competing interactions are discussed in Section 3. In Sections 4.1–4.3 the pattern of the boundary F-$\langle 2.\bar{1}\rangle$ splitting is obtained. The existence of the infinite cascades of phase transitions and several "complete devil's staircases" are demonstrated in Sections 5.1–5.2, 6.1–6.2, 7.1–7.3.

2. PHASE DIAGRAM OF THE GROUND STATE

The Hamiltonian of the model considered is

$$\mathscr{H} = -v^{-1}J_0 \sum_{l,\mathbf{r},\mathbf{r}'} \sigma_{\mathbf{r},l}\sigma_{\mathbf{r}',l} + \tfrac{1}{2}\sum_{l,\mathbf{r}} (J_1\sigma_{\mathbf{r},l}\sigma_{\mathbf{r},l+1} + J_2\sigma_{\mathbf{r},l}\sigma_{\mathbf{r},l+2}) - h\sum_{l,\mathbf{r}} \sigma_{\mathbf{r},l} \quad (1)$$

where $\sigma_{\mathbf{r},l}$ is the Ising spin ($\sigma = 1$) in layer l at site $(\mathbf{r}, l)$. It interacts ferromagnetically with its v nearest neighbors $\sigma_{\mathbf{r}',l}$ of the same layer. We suppose also the coordination number of the lattice is equal to $v + 2$.

There exist several approaches to construction of the phase diagram at zero temperature. Here we employ the so-called linear programming method. This choice is not an accidental one, because this method will be applied throughout the calculations of the present work.

Since J_0 is positive, the ground state of N_l spins of a layer is specified by the ferromagnetic spin orientation "up" or "down" (+ or −). The ground-state energy, i.e., the interlayer part of it, can be written as

$$\begin{aligned} E/N_l = &-h(N(+) - N(-)) \\ &+ 0.5J_1(N(++) + N(--) - N(+-) - N(-+)) \\ &+ 0.5J_2(N(+++) + N(+++) + N(-+-) + N(---) \\ &\qquad - N(++-) - N(+--) - N(-++) - (N--+)) \end{aligned} \quad (2)$$

where $N(+)$ $[N(-)]$ denotes the total number of layers with a positive (negative) magnetization; $N(\sigma_1, \sigma_2)$ is the total number of pairs of adjacent layers with magnetizations per site being equal to σ_1 and σ_2, etc. Dividing this number by the total number of layers N_0 defines the corresponding probabilities of magnetic configurations. A convenient notation for probabilities is $(+ - \cdots + \cdots)$, for instance, $(+-+) = N(+-+)/N_0$. An integer D is the cardinality of a probability space $(\sigma_1, \sigma_2, \ldots, \sigma_D)$. The energy (2) depends on probabilities for $D \leqslant 3$. They cannot be varied independently. A convenient reduced set of independent probabilities can be selected as

$$\begin{aligned} (++) &= P, \qquad (--) = Q \qquad (D = 2) \\ (+++) &= p, \qquad (---) = q \qquad (D = 3) \end{aligned} \quad (3)$$

The other probabilities ($D \leqslant 3$) can be calculated using the obvious sum rules:

$$
\begin{aligned}
(++-) &= (-++) = (++) - (+++) = P - p \\
(--+) &= (+--) = (--) - (---) = Q - q \\
(+-) &= (-+) = (1 - (++) - (--))/2 = (1 - P - Q)/2 \\
(+-+) &= (+-) - (+--) = (1 - P - 3Q + 2q)/2 \\
(-+-) &= (-+) - (-++) = (1 - Q - 3P + 2p)/2 \\
(+) &= (+-) + (++) = (1 + P - Q)/2 \\
(-) &= (-+) + (--) = (1 + Q - P)/2
\end{aligned}
\tag{4}
$$

Equations (4) imply the following inequalities:

$$P \geqslant p \geqslant 0, \qquad Q \geqslant q \geqslant 0$$
$$P + 3Q - 2q \leqslant 1, \qquad Q + 3P - 2p \leqslant 1$$

and define a convex polytope P_4. We need to know vertices of P_4 because any linear form defined on a convex polytope achieves its extremal value on vertices of a polytope. As the energy (2) is a linear function of probabilities P, Q, p, and q, its extremal points coincide with the vertices $(P, p;\ Q, q)$:

$$(0,0;0,0); \qquad (1,1;0,0) \text{ and } (0,0;1,1);$$
$$(1/3,0;0,0) \text{ and } (0,0;1/3,0); \qquad (1/4,0;1/4,0)$$

A brief calculation shows that these extremal points correspond to the periodic structures AF, F, and $\bar{F}$, $\langle 2.\bar{1}\rangle$ and $\langle \bar{2}.1\rangle$, $\langle 2.\bar{2}\rangle$, respectively.

Hereafter we denote $\langle A\rangle$ the periodic structure with the elementary cell A. The symbol $\langle AB\rangle$ denotes the periodic structure constructed as the dimeric sequence of A and B elements. For instance, $\langle 2.\bar{1}\rangle$ has an elementary cell $++-$. The antiferromagnetic structure AF can be represented also by the symbol $\langle 1.\bar{1}\rangle$; the usual ferromagnetic state is denoted by F.

The phase diagram of the model at zero temperature and in a positive magnetic field is depicted in Fig. 1, where the aforementioned phases are shown to be separated by phase boundaries. The ground state along any phase boundary is infinitely degenerate. As an example, consider the degenerate ground state along the boundary F–$\langle 2.\bar{2}\rangle$. Here the probabilities of the sequences $---$, $+-+$, and $-+-$ vanish. Hence, this situation can be described by introducing one degeneracy parameter, or, for brevity, parameter, p, via the relations

$$q = 0, \qquad Q = (1 - p)/4, \qquad P = (1 + 3p)/4$$

Along the boundary the ground-state energy cannot depend on p.

It will be shown that the boundaries presented in Fig. 1 split with the formation of sets of intermediate phases. A splitting is specified by the

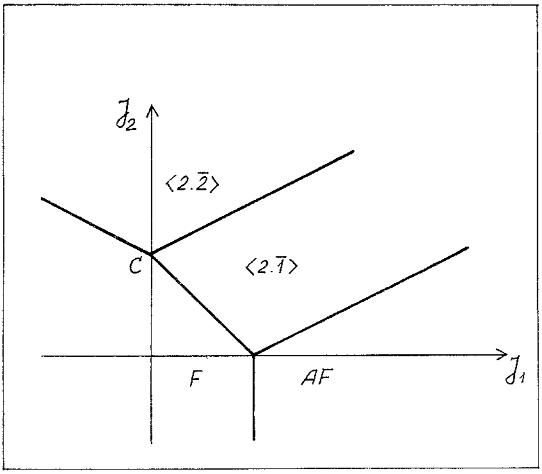

Fig. 1. Phase diagram of the model at $T = 0$. Equations for the phase boundaries are given in the following sections.

coupling relationship. However, the boundary F–AF is an exception to the general rule. There is no splitting along this boundary. In its vicinity the probability space contains three nonvanishing probabilities of cardinality $D = 3$, namely,

$$(+++) = p, \qquad (+-+) = (-+-) = (1-p)/2$$

To pass from the sequence $+++$ to $+-+$ or to $-+-$ we need to include the intermediate sequence $++-$. In our convention the probability of the last sequence vanishes, i.e., the interface energy F–AF does not vanish as in the case of a genuine competition.

3. STRATEGY FOR ATTACKING THE PROBLEM

To analyze the model described by the Hamiltonian (1), the low-temperature expansion of the free energy will be used. For the sake of simplicity we suppose the following inequality to hold:

$$J_0 \gg T \tag{5}$$

This ensures the convergence of the series of the free energy.

In the vicinity of some boundary the ground-state energy is a function of a degeneracy parameter. Excitations above a ground state are associated with spin–flips, which can be divided into two groups. These are "connected" and "disconnected" configurations, shown in Fig. 2. The energy of the spin–flip at the lattice site $(\mathbf{r}, l)$ depends in our model on the orientations of the nearest spins in the layer l and of five spins, arranged

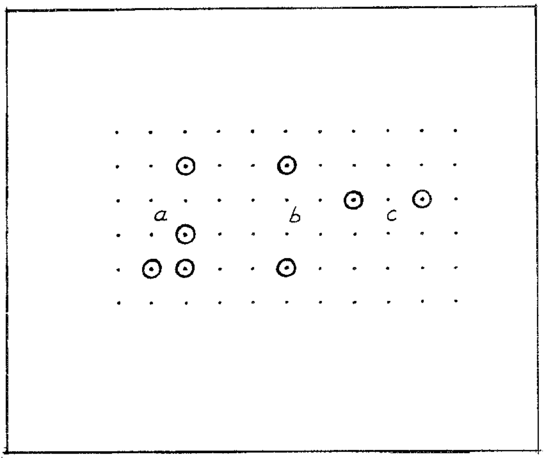

Fig. 2. Excited states. A flipped spin is denoted by a circle. The spin–flip (a) is "connected," while the other two, (b) and (c), are "disconnected."

along the chain from the site $(\mathbf{r}, l-2)$ to $(\mathbf{r}, l+2)$. Therefore, the cardinality of a probability space should be extended from three to five. One might expect the increase of the cardinality to generate a new degeneracy parameter in addition to the basic one, the former and the latter to be denoted by q and p.

To first order of the low-temperature expansion one finds

$$f = \epsilon_0 - T \sum_{\lambda} w_{\lambda} \exp(-\epsilon_{\lambda}/T) \tag{6}$$

where f is the reduced free energy per spin, ϵ_0 is the energy of the degenerate ground state per spin, and the possible five-spin sequence λ is characterized by its probability w_{λ} and by the excitation energy, caused by the spin–flip in the middle of this sequence.

As mentioned before, ϵ_0 is a linear function of p:

$$\epsilon_0 = \xi p \tag{7}$$

where ξ is a deviation from some boundary and may be computed from Eq. (2). The equation

$$\xi = 0$$

defines the position of that boundary in Fig. 1. It is convenient to rearrange (6), absorbing p-dependent components of w_{λ} into the ground-state energy ϵ_0. This only shifts the boundary slightly renormalizing ξ by terms of the order of Tw_0, where $w_0 = \exp(-2J_0/T) \ll 1$ in view of inequality (5). The new parameter q plays a more significant role. In addition to two competing vertices, defined by p, the parameter q generates new vertices. In the vicinity of the boundary a new state may have the lowest free energy. It leads to a splitting of the ground boundary. If this happens a new basic parameter p' can be introduced instead of p along a new boundary. Otherwise, the boundary remains stable against the higher orders of low-temperature expansion, because the energy gap between original and new phases is of the order of Tw_0. Higher-order terms of a series for the free energy include higher powers of w_0 and practically do not change this gap.

One extra situation is worth explaining. Sometimes the first-order terms of the low-temperature expansion do not generate a new parameter, leading instead to a small shift of the corresponding boundary. But in the next orders a new parameter may still be generated.

Higher orders of the low-temperature expansion correspond to multispin flips. By the "linked-cluster theorem" (see Refs. 16 and 9), one should take into account all "connected" configurations of flipped spins and select that contribution of "disconnected" configurations into the free-energy expansion which is proportional to N.

Most of the important features like the stability of the boundary, the pattern of the boundary splitting into a set of striped intermediate phases, and the widths of these phases on the phase diagram, can be found by restricting ourselves to the most significant in-chain "connected" spin–flips only. More detailed considerations are needed only to obtain the exact shape of the boundary lines.

To outline our convention of a choice of leading spin–flips we compare the statistical weights of two configurations (a and b) of flipped spins. They consist of the same "connected" configuration of spin–flips along a chain. But an extra in-layer "linked" bond is contained in the configuration a. The degeneracy parameters coincide, but the Boltzmann factor of the a differs from that of b by a factor $\exp[-2(1-v^{-1})J_0/T] \ll 1$. Therefore, the contribution of a can be neglected. In addition, only in-chain spin–flips can produce new degeneracy parameters in the vicinities of the phase boundaries considered; conversely any "disconnected configuration" cannot generate new parameters.

Henceforth, in accordance with our convention we shall select only "connected" configurations of flipped spins to pick up essential ones among them. This program will be carried out below.

4. THE BOUNDARY F–$\langle 2.\bar{1}\rangle$

4.1. Low-Order Expansion

The ground-state equation of this line has a form:

$$J_1 + J_2 = h \qquad (J_1 > 0, J_2 > 0)$$

A convenient choice of the basic parameter is as follows:

$$p = (+++)$$

There exist three more nonzero probabilities ($D = 3$):

$$(++-) = (+-+) = (-++) = (1-p)/3$$

The ground-state energy (2) can be represented in the vicinity of this boundary by the equations [cf. Eq. (7)]

$$f = \xi p, \qquad \xi = \tfrac{2}{3}(J_1 + J_2 - h) \tag{8}$$

There are two favored single-spin–flips with a vanishing in-chain part of the excitation energy.

If we denote the flipped spin by a circle, they can be depicted as $++\oplus++$ and $++\ominus++$. Here the relevant surroundings are also shown. The probability of the latter configuration is given by a simple relation

$$(++-++) = (-++) = (1-p)/3$$

which depends on p only. The first configuration generates new parameters. In this case the probability space ($D = 5$) implies the following additional parameters:

$$q = (++++) \quad \text{and} \quad r = (+++++)$$

The remaining probabilities for $D = 4$ and $D = 5$ can be written as certain linear combinations of p, q, and r. The corresponding polytope is specified by the following inequalities:

$$p \geqslant q \geqslant r \geqslant 0, \qquad p - 2q + r \geqslant 0, \qquad 4p - 3q \leqslant 1 \tag{9}$$

The first of inequalities (9) is obvious. The others are based on the set of equalities

$$\begin{aligned}(-+++-) &= (+++-) - (++++-)\\ &= (+++) - (++++) - (++++) + (+++++)\\ &= p - 2q + r \geqslant 0\\ (+-++-) &= (-++-) = (++-) - (+++-)\\ &= (++-) - (+++) + (++++)\\ &= (1 - 4p + 3q)/3 \geqslant 0\end{aligned}$$

The vertices (p, q, r) of the polytope and the periodic structures corresponding to them can easily be calculated:

$(1, 1, 1)$	F
$(0, 0, 0)$	$\langle 2.\bar{1}\rangle$
$(1/4, 0, 0)$	$\langle 3.\bar{1}\rangle$
$(2/5, 1/5, 0)$	$\langle 4.\bar{1}\rangle$

In first order of the low-temperature expansion the contributions of the leading spin-flips $+ + \oplus + +$ and $+ + \ominus + +$ result in the following expression of the free energy:

$$f = \xi p - T w_0 r \tag{10}$$

where $w_0 = \exp(-2J_0/T)$ is the Boltzmann factor of these spin-flips. The deviation ξ from the boundary in Eq. (10) differs from that in Eq. (8) by a small term of the order of Tw_0. It is a consequence of the configuration $+ + \ominus + +$.

Minimization of the reduced free energy (10) is illustrated in Fig. 3. To this order the boundary F–$\langle 2.\bar{1}\rangle$ is stable, so that the extra extremal phases $\langle 3.\bar{1}\rangle$ and $\langle 4.\bar{1}\rangle$ cannot compete with the original ones.

Near the point C (see Fig. 1) this conclusion does not hold. If the coupling J_1 becomes of the order of T, the new single-spin-flips

$$+ + \oplus - + \quad \text{and} \quad + - \oplus + + \tag{11}$$

begin to compete with the previous configurations. A convenient param-

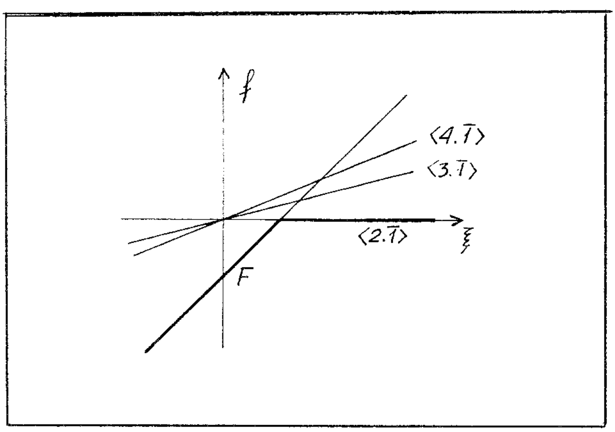

Fig. 3. Free energy (10) as a function of ξ. Four different lines correspond to four relevant vertices (p, q, r).

etrization of the boundary in the vicinity of the point C is

$$J_1 = j \cdot T, \qquad J_2 = h - j \cdot T$$

The Boltzmann factors of the excitations (11) are defined by the equation

$$Z = \exp(-2j)$$

and the probabilities of the spin–flips (11) are connected with the additional parameter q via

$$(+-+++) = (+++-+) = (+++-) \\ = (+++) - (++++) = p - q$$

Now the reduced free energy takes a form

$$f = \xi p - Tw_0(-2qz + r) \tag{12}$$

To determine what kind of phases can survive we must compare magnitudes of the free energy (12) at the vertices. Such an analysis leads to the following conclusions:

1. There is no splitting at $z < 1/2$ (or $j > \frac{1}{2}\ln 2$)
2. The intermediate phase $\langle 3.\bar{1}\rangle$ appears in between F $\langle 2.\bar{1}\rangle$, if $z > 1/2$. The phase $\langle 4.\bar{1}\rangle$ is energetically unfavorable elsewhere; therefore the phase transition F–$\langle 3.\bar{1}\rangle$ in the sequence F–$\langle 3.\bar{1}\rangle$–$\langle 2.\bar{1}\rangle$ will be stable against higher orders of the low-temperature expansion.

4.2. The Boundary F–$\langle 2.\bar{1}\rangle$ (Continued)

Here we consider the stability of the new boundary $\langle 2.\bar{1}\rangle$–$\langle 3.\bar{1}\rangle$ produced by the original boundary F–$\langle 2.\bar{1}\rangle$.

Generally, in a vicinity of any new boundary new values of a basic parameter p', additional parameters $q', r', \ldots,$ and deviations ξ' from a boundary must be introduced instead of the original values p, q, r, and ξ. It is convenient not to proliferate notation and to retain the old one.

Two in-chain spin–flips generate the new extended probability space ($D = 7$). The probabilities, depending on the additional parameter, are

$$\begin{aligned} &(+++-+++) = q \\ &(+++-++-) = (-++-+++) = p - q \\ &(-++-++-) = (-++-) - (-++-+++) \\ &\qquad = (1 - 7p + 3q)/3 \end{aligned} \tag{13}$$

The basic parameter is defined by $p = (+++)$. The set of the significant two-spin–flips is

$$++\oplus-\oplus++, \quad ++\oplus-\oplus+-, \quad \text{and} \quad -+\oplus-\oplus+-$$

with respective in-chain contributions to the excitation energy $\Delta_{\mathrm{ch}} = 2h$, $4h$, and $6h$. Therefore, the free energy can be written as

$$f = \xi p - T w_0^2 w_h (1 - w_h)^2 q \tag{14}$$

where $w_h = \exp(-2h/T)$.

The competitive vertices (p, q) can easily be derived from Eqs. (13) as

$$\begin{array}{ll} (0,0) & \langle 2.\bar{1} \rangle \\ (1/4, 1/4) & \langle 3.\bar{1} \rangle \\ (1/7, 0) & \langle 3.\bar{1}.2.\bar{1} \rangle \end{array}$$

The second term on the right-hand side of Eq. (14) is negative, thus making the intermediate phase $\langle 3.\bar{1}.2.\bar{1} \rangle$ energetically unfavorable.

Our conclusion holds at $h < J_0$. If $h > J_0$, then the Boltzmann factors of the previous two-spin–flips are much smaller than those of the following three-spin–flips

$$- \oplus + \ominus + \oplus - \quad \text{and} \quad + + \oplus \ominus + \oplus -$$

whose in-chain contributions to the excitation energy are $\Delta_{\mathrm{ch}} = 4j \cdot T$ and $2j \cdot T$, respectively. The corresponding free energy can be written as

$$f = \xi p + T w_0^3 (2 - z) z q \tag{15}$$

In contrast to Eq. (14) the second term on the right-hand side of Eq. (15) is positive. Hence, the intermediate phase appears in between the phases $\langle 3.\bar{1} \rangle$ and $\langle 2.\bar{1} \rangle$, i.e.,

$$\langle 2.\bar{1} \rangle – \langle 2.\bar{1}.3.\bar{1} \rangle – \langle 3.\bar{1} \rangle$$

A comparison of Eqs. (14) and (15) demonstrates an important feature of the free energy expansion. A term, depending linearly on the additional parameter, may be either positive or negative depending on whether the significant probabilities are "diagonal" or "off-diagonal." For example, any degenerate state along the boundary $\langle 2.\bar{1} \rangle – \langle 3.\bar{1} \rangle$ can be constructed as an arbitrary sequence of the "particles" $A = + + -$ and $B = + + + -$, like $\ldots A^{n_1} B^{m_1} A^{n_2} B^{m_2} \ldots$.[2] The concentration of "particles" A and B are connected with the basic parameter linearly. An extension of the probability space by introducing the following probabilities: (BB), (BA), (AB), and (AA), leads simultaneously to the appearence of an additional parameter q. Hereafter we use the compact form of (13). Of these probabilities two, (AA) and (BB), may be called "diagonal." They yield the main negative contri-

[2] We add the Appendix, where all abbreviations of spin sequences and configurations of flipped spins are listed for easy rapid reference.

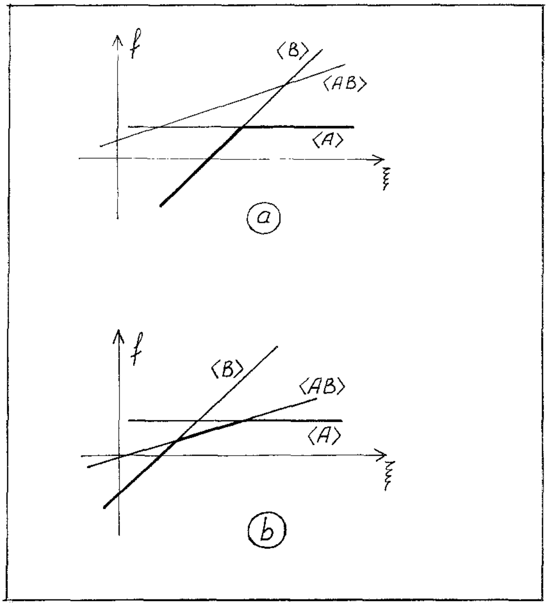

Fig. 4. Two possibilities concerning the splitting of the boundary $\langle A\rangle$–$\langle B\rangle$: (a) The case of a positive q term: the free energy of the intermediate phase $\langle AB\rangle$ is higher than that of either of the phases $\langle A\rangle$ and $\langle B\rangle$. (b) The reverse possibility: there exists a stability region of the phase $\langle AB\rangle$.

bution to the q term in Eq. (14). Conversely, the contribution to the q term in Eq. (15) is positive when caused by the "off-diagonal" probabilities (AB) and (BA). Figure 4 illustrates these possibilities.

Furthermore, we shall use this change of sign of the additional degeneracy parameter term of the free-energy expansion to find the phase diagram.

4.3. The Boundary F–$\langle 2.\bar{1}\rangle$: Resulting Splitting

Firstly we show that the boundary $\langle 3.\bar{1}.2.\bar{1}\rangle$–$\langle 3.\bar{1}\rangle$ (or the boundary $\langle BA\rangle$–$\langle B\rangle$ in the notation of Section 4.2) is stable.

We choose the basic degeneracy parameter in the following form:

$$p = (BB) = (+++-+++-) = (+++-+++)$$

Along the boundary in question any degenerate sequence $\ldots A^{n_1}B^{m_1} A^{n_2}B^{m_2}\ldots$ implies that either all of the n_i are 0 or 1. There arises an additional parameter q defined so that

$$\begin{aligned}(BBB) &= (+ + + - + + + - + + + -) \\ &= (+ + + - + + + - + + +) = q \\ (BBA) &= (+ + + - + + + - + + -) = p - q \\ (ABB) &= (- + + - + + + - + + +) = p - q \\ (ABA) &= (- + + - + + + - + + -) = (1 - 11p + 7q)/7\end{aligned} \tag{16}$$

To prove the last equation here, we note that (AB) vanishes in the "pure" phase $\langle B \rangle$ and $(AB) = 1/7$ in the phase $\langle AB \rangle$. Therefore the linear dependence

$$(BA) = (+ + + - + + -) = (1 - 4p)/7$$

allows one to obtain the probability (ABA) from the simple equation

$$(ABA) = (BA) - (BBA)$$

In the range of the coupling constants pointed out in Section 4.2 five-spin–flips are significant:

$$\begin{array}{lll} & + + \oplus \ominus + \oplus + \ominus \oplus + + & (\Delta_{\text{ch}} = 0) \\ \text{and} & \left.\begin{array}{l} + + \oplus \ominus + \oplus + \ominus + \oplus - \\ - \oplus + \ominus + \oplus + \ominus \oplus + + \end{array}\right\} & (\Delta_{\text{ch}} = 2j \cdot T) \\ & - \oplus + \ominus + \oplus + \ominus + \oplus - & (\Delta_{\text{ch}} = 4j \cdot T) \end{array} \tag{17}$$

The in-chain excitation energies are quoted in parentheses.

The main contribution to the free energy expansion

$$f = \xi p - T w_0^5 (1 - z)^2 q \tag{18}$$

is due to the diagonal probabilities (BBB) and (ABA). One concludes that the boundary $\langle AB \rangle$–$\langle B \rangle$ is stable.

The analogous conclusion can be made about the stability of the boundary $\langle A^{n+1}B \rangle$–$\langle A^n B \rangle$ ($n \geqslant 1$). The probabilities connected with the additional parameter are as follows:

$$(A^{n+1}BA^{n+1}), \quad (A^{n+1}BA^nB), \quad (BA^nBA^{n+1}) \quad \text{and} \quad (BA^nBA^nB)$$

There are four leading spin–flips [cf. (17)]:

$$+ + \oplus \ominus + \oplus \ominus \cdots + \oplus \ominus + \oplus + \ominus \oplus + \ominus \cdots \oplus + \ominus \oplus + + \qquad (\Delta_{\mathrm{ch}} = 0)$$

$$+ + \oplus \ominus + \oplus \ominus \cdots + \oplus \ominus + \oplus + \ominus + \oplus \ominus \cdots + \oplus \ominus + \oplus - \qquad (\Delta_{\mathrm{ch}} = 2j \cdot T)$$

$$- \oplus + - \oplus + \ominus \cdots \oplus + \ominus + \oplus + \ominus \oplus + \ominus \cdots \oplus + \ominus \oplus + + \qquad (\Delta_{\mathrm{ch}} = 2j \cdot T)$$

$$- \oplus + \ominus \oplus + \ominus \cdots \oplus + \ominus + \oplus + \ominus + \oplus \ominus \cdots + \oplus \ominus + \oplus - \qquad (\Delta_{\mathrm{ch}} = 4j \cdot T)$$

The relevant q term of the reduced free energy

$$f = \xi p + \alpha q \tag{19}$$

is again negative, as in Eq. (18). This ensures the stability of the boundary $\langle A^{n+1}B\rangle$–$\langle A^nB\rangle$.

We often shall refer to Eq. (19)—more concretely, to the second term on the right-hand side of Eq. (19), which for brevity will be called the q term.

We must also control the stability of the boundary $\langle A\rangle$–$\langle AB\rangle$. The probabilities, containing the additional parameter, can be written by using the convenient notation

$$(AAA), \qquad (AAB), \qquad (BAA), \qquad (BAB)$$

The main competition is caused by the following five-spin–flips:

$$- \oplus + \ominus + \oplus \ominus + \oplus - \qquad [\Delta_{\mathrm{ch}} = 4j \cdot T, (AAA)]$$

$$\left.\begin{array}{l} - \oplus + \ominus \oplus + \ominus + \oplus - \\ + + \oplus \ominus + \oplus \ominus + \oplus - \end{array}\right\} \qquad [\Delta_{\mathrm{ch}} = 2j \cdot T, (BAA)]$$

$$- \oplus + \ominus \oplus + \ominus \oplus + + \qquad [\Delta_{\mathrm{ch}} = 2j \cdot T, (AAB)]$$

where the in-chain excitation energies and the corresponding probabilities are quoted in brackets. Here the general expression for the reduced free energy (19) is also correct and includes the coefficient α, proportional to the positive multiplier $(2 - 2z)$. Hence the boundary splits according to the scheme

$$\langle A\rangle\text{–}\langle A^2B\rangle\text{–}\langle AB\rangle$$

To determine conditions for the instability of the boundary $\langle A\rangle$–$\langle A^2B\rangle$ we employ our usual approach. The main "connected" spin–flips contain seven individual flipped spins:

$$\left.\begin{array}{l} -\oplus+\ominus\oplus+\ominus\oplus+\ominus+\oplus- \\ -\oplus+\ominus\oplus+\ominus+\oplus\ominus+\oplus- \\ -\oplus+\ominus+\oplus\ominus+\oplus\ominus+\oplus- \end{array}\right\} \qquad [\Delta_{\mathrm{ch}} = 4j \cdot T,\ (AAAA)]$$

$$++\oplus\ominus+\oplus\ominus+\oplus\ominus+\oplus- \qquad [\Delta_{\mathrm{ch}} = 2j \cdot T,\ (BAAA)]$$

$$-\oplus+\ominus\oplus+\ominus\oplus+\ominus\oplus++ \qquad [\Delta_{\mathrm{ch}} = 2j \cdot T,\ (AAAB)]$$

The coefficient α [see Eq. (19)] is proportional here to $(2 - 3z)$. Hence, the splitting of the boundary $\langle A\rangle$–$\langle A^2B\rangle$ takes place only at $z < 2/3$. We recall also that $z > 1/2$. As soon as one has $2/3 < z < 1$ the boundary becomes stable. The boundary between the phases $\langle A\rangle$ and $\langle A^3B\rangle$ is stable at any z.

It is worthwhile to cite the resulting phase transitions along the original boundary F–$\langle 2.\bar{1}\rangle$. In a "weak" magnetic field ($h < J_0$) the single splitting takes place in the vicinity of the point C (see Fig. 1):

$$F\text{–}\langle 2.\bar{1}\rangle \qquad (z < 1/2)$$

$$F\text{–}\langle 3.\bar{1}\rangle\text{–}\langle 2.\bar{1}\rangle \qquad (z > 1/2)$$

A region of a "strong" magnetic field ($h > J_0$) exhibits the more compli-

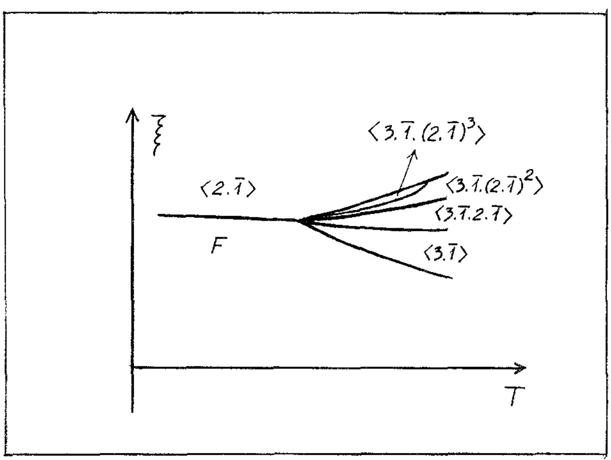

Fig. 5. Phase diagram in the (ξ, T) plane in the vicinity of the C point (Fig. 1).

cated sequence of phase transitions:

$$F\text{–}\langle 2.\bar{1}\rangle \qquad (z < 1/2)$$

$$F\text{–}\langle 3.\bar{1}\rangle\text{–}\langle 3.\bar{1}.2.\bar{1}\rangle\text{–}\langle 3.\bar{1}.(2.\bar{1})^2\rangle\text{–}\langle 3.\bar{1}.(2.\bar{1})^3\rangle\text{–}\langle 2.\bar{1}\rangle \qquad (1/2 < z < 2/3)$$

$$F\text{–}\langle 3.\bar{1}\rangle\text{–}\langle 3.\bar{1}.2.\bar{1}\rangle\text{–}\langle 3.\bar{1}.(2.\bar{1})^2\rangle\text{–}\langle 2.\bar{1}\rangle \qquad (2/3 < z < 1)$$

These are qualitatively shown in Fig. 5.

5. THE BOUNDARY $\langle 2.\bar{1}\rangle$–AF

5.1. Low-Order Expansion

The stability of this boundary has been investigated in a short note.[12] Here we present the technicalities of calculations.

The equation of this boundary is as follows:

$$J_1 = h + 2J_2 \qquad (J_2 > 0)$$

The basic parameter can conveniently be introduced using the following relationships:

$$(+ + -) = (- + +) = p, \quad (+ - +) = (1 - p)/2, \quad (- + -) = (1 - 3p)/2$$

There are two distinct possibilities. The first one corresponds to the inequality $J_0 > J_2$. Then the single-spin–flips are most-favored ones:

$$+ + \ominus + + \qquad (\Delta_{ch} = 6J_2)$$

$$+ + \ominus + - \quad \text{and} \quad - + \ominus + + \qquad (\Delta_{ch} = 4J_2)$$

$$- + \ominus + - \qquad (\Delta_{ch} = 2J_2)$$

As usual, the free-energy expansion in the vicinity of the boundary has a form of Eq. (19) with

$$\alpha = -Tw_0(1 - w_2)^2 < 0$$

where $w_2 = \exp(-2J_2/T)$. The boundary $\langle 2.\bar{1}\rangle$–AF (or $\langle 2.\bar{1}\rangle$–$\langle 1.\bar{1}\rangle$) is stable.

The reverse inequality ($J_2 > J_0$) leads to the different most-favored spin–flips:

$$+ \oplus \ominus + - \quad \text{and} \quad - + \ominus \oplus +$$

Their in-chain excitation energies vanish. The coefficient α in the free-energy expansion is positive, being

$$\alpha = 2Tw_0^2$$

This circumstance can be understood in another manner. The main contribution to the q term of the reduced free energy is caused by the off-diagonal spin–flip configurations with the probabilities (AC) and (CA), where the "particle" C is defined below:

$$C = + -$$

So, the first step of the boundary $\langle 2.\bar{1}\rangle$–$\langle 1.\bar{1}\rangle$ splitting is such that

$$\langle 2.\bar{1}\rangle\text{–}\langle 2.\bar{1}.1.\bar{1}\rangle\text{–}\langle 1.\bar{1}\rangle$$

or

$$\langle A\rangle\text{–}\langle AC\rangle\text{–}\langle C\rangle$$

5.2. Resulting Splitting

The degenerate structure, constructed by using the particles A and C as bricks, can be written in a general form:

$$\ldots A^{n_1}C^{m_1}A^{n_2}C^{m_2}\ldots$$

As shown in Section 5.1 there exists a region of the stability of the dimerized phase $\langle AC\rangle$ with all of n's and m's being equal to unity. A further analysis has to be made along the coexistive curves $\langle A\rangle$–$\langle AC\rangle$ and $\langle AC\rangle$–$\langle C\rangle$. The general forms of degenerate structures along these boundaries can be represented, respectively, by the following sequences:

$$\ldots A^{n_1}CA^{n_2}C\ldots$$

and

$$\ldots C^{m_1}AC^{m_2}A\ldots$$

Concerning the stability of the boundaries considered, two observations can be made:

1. The boundary $\langle A\rangle$–$\langle A^{k-1}C\rangle$ splits with an appearance of a strip of the intermediate phase $\langle A^kC\rangle$.
2. The boundary $\langle A^kC\rangle$–$\langle A^{k-1}C\rangle$ is stable.

They can be proven as follows:

The basic parameter p along the boundary $\langle A\rangle$–$\langle A^{k-1}C\rangle$ can conveniently be represented as

$$p = (A^k)$$

In the p space there are two extremal points or vertices corresponding to the stable phases $\langle A\rangle$ and $\langle A^{k-1}C\rangle$. The degeneracy parameter q is defined by the following probabilities:

$$
\begin{aligned}
(A^{k+1}) &= q \\
(A^kC) = (CA^k) &= p - q \\
(CA^{k-1}C) &= (A^{k-1}C) - (A^kC) \\
&= \left[1 - (3k+2)p + (3k-1)q\right]/(3k-1)
\end{aligned}
\tag{20}
$$

It is worthwhile to explain the origin of the equality

$$(A^{k-1}C) = (1-3p)/(3k-1) \tag{21}$$

Obviously the basic parameter p is equal to $1/l_A$ and to $1/[(k-1)l_A + l_C]$ in the case of "pure" structures $\langle A\rangle$ and $\langle A^{k-1}C\rangle$, respectively. Here $l_A = 3$ and $l_C = 2$ are the periods of phases $\langle A\rangle$ and $\langle C\rangle$. These extremal values of p directly lead to Eq. (21).

To establish a relevant q term of the free-energy expansion [see Eq. (19)] we must consider only those spin–flips, which are connected with a new parameter q and whose Boltzmann weights are largest among them. These leading spin–flips correspond to the vanishing of their in-chain excitation energies. This vanishing is consistent with two possible spin–flip configurations within particles A

$$A_1 = \oplus + \ominus \quad \text{and} \quad A_2 = + \oplus \ominus$$

The flip of negative spins is essential, otherwise the following three-spin sequence may appear: $\oplus - \oplus$, which corresponds to too high an excitation energy. The possible in-chain orderings of elements A_1 and A_2 are determined by three allowed transitions between the neighboring elements:

$$A_1 \to A_1, \qquad A_2 \to A_2, \qquad A_2 \to A_1$$

whereas $A_1 \to A_2$ is forbidden, because this transition produces the excited spin sequence $+++$ having too high an in-chain energy.

Let us present now the "connected" spin–flip sequences, having vanishing in-chain excitation energy, which induce a new parameter q and contain the minimal number of the individual spin–flips:

$$+\oplus\ominus A_2A_2 \cdots A_1 + - \quad \text{and} \quad - + \ominus A_1A_1 \cdots A_1 \oplus + \tag{22}$$

Consequently, among all the relevant configurations these two have the lowest excitation energy.

If at least one C-particle is included in the sequence, consisting of A-particles, then the indirect transition $A_1 \to A_2$ becomes possible. Indeed, spin–flips within C-particle of the form

$$\overline{C} = \oplus\ominus$$

usually lead to transitions $A_1 \rightarrow \bar{C}$ and $\bar{C} \rightarrow A_2$. Therefore, the indirect $\bar{C}$-mediated transitions $A_1 \rightarrow \bar{C} \rightarrow A_2$ become possible.

The minimal number of spin–flips in a relevant sequence with a single "impurity" (C-particle) corresponds to the following combination:

$$- + \ominus A_1 \ldots A_1 \bar{C} A_2 \ldots A_1 + - \tag{23}$$

These sequences must be taken into account to find the stability condition of the boundary $\langle A^{k-1}C\rangle$–$\langle A^kC\rangle$. The relevant probabilities including the additional parameter q, can be written

$$\begin{aligned} (A^kCA^k) &= q \\ (A^kCA^{k-1}C) &= (CA^{k-1}CA^k) = p - q \\ (CA^{k-1}CA^{k-1}C) &= (CA^{k-1}C) - p + q \end{aligned} \tag{24}$$

where the basic parameter $p = (A^{k-1}CA^{k-1})$ and

$$(CA^{k-1}C) = [1 - (3k+2)p]/(3k-1)$$

In the leading spin–flips (22) the end-point particles are different. This ensures that the main contribution into the q-term of the reduced free energy comes from the off-diagonal probabilities (A^kC) and (CA^k). The boundary $\langle A\rangle$–$\langle A^{k-1}C\rangle$ splits according to the scheme $\langle A\rangle$–$\langle A^kC\rangle$–$\langle A^{k-1}C\rangle$. In the vicinity of the boundary between $\langle A^{k-1}C\rangle$ and $\langle A^kC\rangle$ phases the diagonal probability $(CA^{k-1}CA^{k-1}C)$ is significant [compare the labeling of the relevant probabilities (24) and spin–flip configurations (23)]. In this case the boundary remains stable against all highest orders of the free-energy expansion.

As usual, in both these cases the reduced free energy has the general form (19). The coefficients α are equal to $2Tw_0^{2k}$ and $-Tw_0^{4k-1}$, respectively.

A brief consideration allows one to establish the sequence of phase transitions as

$$\langle C\rangle\text{–}\langle AC\rangle\text{–}\langle A^2C\rangle - \cdots - \langle A^nC\rangle - \cdots - \langle A\rangle$$

or in an alternative notation

$$\langle 1.\bar{1}\rangle\text{–}\langle 2.\bar{1}.1.\bar{1}\rangle\text{–}\langle (2.\bar{1})^2.1.\bar{1}\rangle - \cdots - \langle (2.\bar{1})^n.1.\bar{1}\rangle\text{–} \cdots \text{–}\langle 2.\bar{1}\rangle \quad (J_2 > J_0)$$

and

$$\langle 1.\bar{1}\rangle\text{–}\langle 2.\bar{1}\rangle \qquad (J_2 < J_0)$$

These cascades are illustrated in Fig. 6.

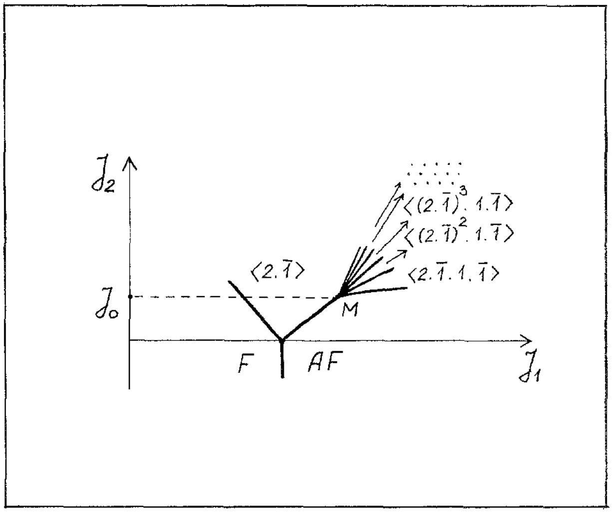

Fig. 6. Splitting of the boundary AF–$\langle 2.\bar{1} \rangle$. There is a multiphase point, M.

6.1. The Boundary $\langle 2.\bar{1} \rangle$–$\langle 2.\bar{2} \rangle$

The equation of this line at $T = 0$ has the form

$$2J_2 = J_1 + 2h \qquad (J_1 > 0)$$

There exist five nonzero probabilities ($D = 3$)

$$(+--) = (--+) = p$$
$$(++-) = (-++) = (1-p)/3$$
$$(+-+) = (1-4p)/3$$

The peculiarity of this boundary consists in appearance of the new parameter q for $D = 6$:

$$(--++--) = q$$
$$(--++-+) = (+-++--) = p - q \tag{25}$$
$$(+-++-+) = (1 - 7p + 3q)/3$$

The vertices in the (p, q)-space, which can also be labeled by the conventional notation for the corresponding phases, are

$(0, 0)$	$\langle 2.\bar{1} \rangle$
$(1/4, 1/4)$	$\langle 2.\bar{2} \rangle$
$(1/7, 0)$	$\langle 2.\bar{2}.2.\bar{1} \rangle$

To the first order in the free-energy expansion there is no splitting, because the single-spin–flips merely generate an extension of the probability space to $D = 5$. Therefore, they may only shift the boundary without any splitting. However, this line is not stable in the next order in the free-energy expansion. The second order is connected with the two double-spin–flips, whose excitation energy vanishes, namely,

$$+ - \oplus + \ominus - \quad \text{and} \quad - \ominus + \oplus - +$$

These spin–flips yield the negative q term in the reduced free energy, consequently the boundary $\langle 2.\bar{1}\rangle$–$\langle 2.\bar{2}\rangle$ is unstable. The intermediate phase here is $\langle 2.\bar{2}.2.\bar{1}\rangle$.

To extend our analysis, we make use of the simplified notations

$$E = + + - -, \qquad A = + + -$$

Firstly, we consider the boundary $\langle 2.\bar{1}\rangle$–$\langle 2.\bar{2}.2.\bar{1}\rangle$, or $\langle A\rangle$–$\langle EA\rangle$. The basic parameter can be defined by the equation

$$p = (AA)$$

The new parameter q is connected with four probabilities (AAA), (EAA), (AAE), and (EAE). The significant three- and four-spin–flips are presented below with the corresponding probabilities and the in-chain excitation energies:

$$+ - \oplus + \ominus + \oplus - + \qquad (AAA)$$
$$- \ominus + \oplus \ominus + \oplus - + \qquad (EAA)$$
$$+ - \oplus + \ominus \oplus + \ominus - \qquad (AAE)$$

The contributions to the q term of the three- and four-spin–flips become comparable to each other, if $J_1 = 2J_0$. In the region $J_1 < 2J_0$ the boundary is stable. At $J_1 > 2J_0$ the boundary splits into the following sequence:

$$\langle EA\rangle\text{–}\langle EA^2\rangle\text{–}\langle A\rangle$$

Along the other boundary $\langle E\rangle$–$\langle EA\rangle$ three-spin–flips are not possible and four-spin–flips,

$$+ - \oplus + \ominus - \oplus + \ominus - \quad \text{and} \quad - \ominus + \oplus - \ominus + \oplus - +$$

correspond to the off-diagonal probabilities (AEE) and (EEA). This ensures the instability and splitting of this boundary with an appearance of the intermediate phase $\langle E^2A\rangle$. In contrast to the case of the boundary $\langle EA\rangle$–$\langle A\rangle$ the last splitting does not depend on the relationship between J_1 and J_0.

6.2. The Boundary $\langle 2.\bar{1}\rangle$–$\langle 2.\bar{2}\rangle$ (Continued)

After introducing the A and E particles a general approach in the spirit of Section 5.2 can be accomplished. Firstly, the following spin–flips within particles A and E are consistent with the vanishing of the in-chain excitation energy:

$$A_1, A_2, A_3 = \oplus\oplus\ominus \quad \text{and} \quad E_1 = \oplus + \ominus -, \qquad E_2 = + \oplus - \ominus$$
$$E_3 = + \oplus\ominus\ominus, \qquad E_4 = \oplus\oplus\ominus\ominus$$

The elements A_3, E_3, E_4 will not be taken into account since the extra flipped spin yields an extra small Boltzmann factor. Transitions from one element to another, like

$$\begin{pmatrix} E_1 \\ A_1 \end{pmatrix} \to \begin{pmatrix} E_1 \\ A_1 \end{pmatrix}, \qquad \begin{pmatrix} E_2 \\ A_2 \end{pmatrix} \to \begin{pmatrix} E_2 \\ A_2 \end{pmatrix}$$

are allowed.

The arguments of Section 6.1 lead to the splitting

$$\langle E\rangle\text{–}\langle EA\rangle\text{–}\langle A\rangle$$

Along the first new boundary any degenerate structures include A particles, as "impurities," surrounded by E particles. In principle, the existence of the following periodic structure does not lead to a direct contradiction along the boundary $\langle E\rangle$–$\langle EA\rangle$:

$$\langle G\rangle = \langle E^{n_1}AE^{n_2}A \ldots E^{n_k}A\rangle$$

Suppose this phase and

$$\langle G_1\rangle = \langle E^{n_1+1}AE^{n_2}A \ldots E^{n_k}A\rangle = \langle EG\rangle$$

have a common boundary. The probabilities

$$(E\tilde{G}E), \quad (E\tilde{G}A), \quad (A\tilde{G}E), \quad \text{and} \quad (A\tilde{G}A) \tag{26}$$

where $\tilde{G} = GE^{n_1}$, describe the competition along the boundary. As a result of such a competition there may appear a new intermediate phase

$$\langle EG^2\rangle$$

Instead of two competitive phases

$$\langle G\rangle \quad \text{and} \quad \langle EG\rangle$$

one can consider the phases $\langle G\rangle$ and $\langle EG^2\rangle$. Then the role of $\tilde{G}$ is played by the following sequence:

$$\tilde{G}' = G^2E^{n_1} = G\tilde{G}$$

and so on. Therefore, we can consider the probabilities (26) as competitive probabilities of a general case.

To compare the labelings of the significant spin–flips with those of their probabilities, we rewrite the set (26) in more detail as

$$
\begin{aligned}
(E\tilde{G}E) &= (--\tilde{G}++--)\\
(E\tilde{G}A) &= (--\tilde{G}++-+)\\
(A\tilde{G}E) &= (+-\tilde{G}++--)\\
(A\tilde{G}A) &= (+-\tilde{G}++-+)
\end{aligned}
\tag{26'}
$$

The "connected" sequences of flipped spins which correspond to vanishing of the in-chain excitation energies of the resulting spin-sequences, which generate the probabilities (26′) and which include the minimal number of individual spin–flips, are given by

$$
\begin{aligned}
&-\ominus E_2^{n_1}A_2E_2^{n_2}A_2\ldots E_2^{n_k}A_2E_2^{n_1}+\oplus-+\\
&+-E_1^{n_1}A_1E_1^{n_2}A_1\ldots E_1^{n_k}A_1E_1^{n_1}\oplus+\ominus-
\end{aligned}
\tag{27}
$$

Both the probabilities $(E\tilde{G}A)$ and $(A\tilde{G}E)$ of these spin–flips are off-diagonal. Hence the striped intermediate phase $\langle EG^2\rangle$ exists in between the phases $\langle G\rangle$ and $\langle EG\rangle$.

The analogous considerations apply to the boundary $\langle A\rangle$–$\langle EA\rangle$. The counterparts of (27) and (26) for this boundary are now

$$
\begin{aligned}
&-\ominus A_2^{m_1}E_2A_2^{m_2}E_2\ldots A_2^{m_k}E_2A_2^{m_1}+\oplus-+\\
&+-A_1^{m_1}E_1A_1^{m_2}E_1\ldots A_1^{m_k}E_1A_1^{m_1}\oplus+\ominus-
\end{aligned}
\tag{28}
$$

and

$$
\begin{aligned}
(A\tilde{\tilde{G}}A) &= (+-\tilde{\tilde{G}}++-+)\\
(A\tilde{\tilde{G}}E) &= (+-\tilde{\tilde{G}}++--)\\
(E\tilde{\tilde{G}}A) &= (--\tilde{\tilde{G}}++-+)\\
(E\tilde{\tilde{G}}E) &= (--\tilde{\tilde{G}}++--)
\end{aligned}
\tag{29}
$$

where

$$\tilde{\tilde{G}} = \bar{G}A^{m_1} = A^{m_1}EA^{m_2}E\ldots A^{m_k}EA^{m_1}$$

The probabilities of the spin–flip sequences (28) are off-diagonal ones again. This circumstance leads to a splitting of any boundary generated from the original boundary $\langle A\rangle$–$\langle EA\rangle$.

This splitting picture is an unexpected one for the ANNNI model. The total scheme of the branching is as follows. The first step corresponds to the splitting

$$
\begin{array}{c}
\langle A\rangle - \langle E\rangle\\
\swarrow \quad \searrow\\
\langle A\rangle - \langle AE\rangle - \langle E\rangle
\end{array}
$$

the second step corresponds to

$$\begin{array}{ccccccccc} \langle A\rangle & & \text{———} & & \langle AE\rangle & & \text{———} & & \langle E\rangle \\ & \swarrow & & \searrow & & \swarrow & & \searrow & \\ \langle A\rangle & \text{–} & \langle A^2E\rangle & \text{–} & \langle AE\rangle & \text{–} & \langle AE^2\rangle & \text{–} & \langle E\rangle, \quad \text{etc.} \end{array}$$

Any two phases $\langle \tilde{A}\rangle$ and $\langle \tilde{E}\rangle$, which become neighboring in this branching hierarchy, create the intermediate phase $\langle \tilde{A}\tilde{E}\rangle$ in between them. This sequence is well known as a "complete devil's staircase," which has been theoretically observed in the lattice gas model with a long-range interaction (see Refs. 17–19).

Our considerations hold at low enough temperature provided $J_1 > 2J_0$. The reverse inequality $J_1 < 2J_0$ leads to the destruction of this curious sequence of phase transitions. Indeed, at $J_1 = 2J_0$ the total excitation energy of any spin–flip sequence formed from sequences (27) by replacing one of the "normal" transitions $A_1 \to E_1$ by the "wrong" one, $A_1 \to E_2$, would be identical and equal to the excitation energy of the original sequences (27). There would exist k new spin–flips, like

$$+ - E_1^{n_1} A_1 \ldots E_1^{n_j} A_1 E_2^{n_{j+1}} A_2 \ldots A_2 E_2^{n_1} + \oplus - + \tag{30}$$

This competition occurs along the original boundary $\langle EA\rangle$–$\langle E\rangle$. Along the other boundary $\langle EA\rangle$–$\langle A\rangle$ one more "wrong" transition $A_1 \to A_2$ becomes allowed. The relevant spin–flips, competing with those of (28), look like

$$+ - A_1^{m_1} E_1 \ldots A_1^{m} A_2^{m_j - m} E_2 \ldots E_2 A_2^{m_1} + \oplus - + \tag{31}$$

The total number of sequences like (31) is equal to

$$\sum_{j=1}^{k} m_j + m_1 - 1$$

Note that the probabilities of the relevant spin–flips from the sets (30) and (31) are usually diagonal. Hence, almost all of the possible boundaries in the region $J_1 < 2J_0$ become stable. There exists one exception to the general rule, because the spin–flips

$$+ - E_1^n \oplus + \ominus - \quad \text{and} \quad - \ominus E_2^n + \oplus - +$$

correspond to off-diagonal probabilities and belong to the sets (27) and (30) simultaneously. Consequently, the conventional approach leads first to the splitting of the boundary $\langle E\rangle$–$\langle A\rangle$, then the boundary $\langle E\rangle$–$\langle EA\rangle$ splits, etc. The resulting cascade looks like

$$\langle A\rangle\text{–}\langle EA\rangle\text{–}\langle E^2A\rangle\text{–} \cdots \text{–}\langle E^nA\rangle\text{–} \cdots \text{–}\langle E\rangle \tag{32}$$

The crossover of the two regimes when

$$J_1 - 2J_0 \sim T$$

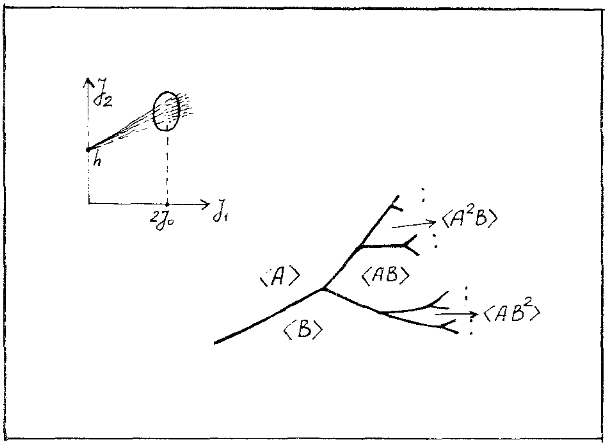

Fig. 7. Schematic representation of "complete devil's staircase" branching. It takes place in the crossover region along the boundary $\langle 2.\bar{1}\rangle$–$\langle 2.\bar{2}\rangle$. Any boundary $\langle A\rangle$–$\langle B\rangle$ from this branching family splits at some triple point generating two new boundaries. Inset: position of the crossover region (encircled) in the (J_1, J_2) plane.

contains an infinite number of triple points. To find them we must compare the contributions of configurations (27) and (30) [or (28) and (31)] to the free-energy expansion. They differ by the factor

$$\frac{K}{2}\exp\left(\frac{J_1 - 2J_0}{T}\right)$$

where K is the total number of representatives of a set (30) [or (31)]. Hence, the simple cascade of phase transitions (32) splits inhomogeneously into the "complete devil's staircase." This situation is depicted qualitatively in Fig. 7.

7. THE BOUNDARY F–$\langle 2.\bar{2}\rangle$

7.1. Low Orders of the Free-Energy Expansion

The splitting pattern of this boundary is the most complicated one among those considered. The boundary equation at the zero temperature is

$$2J_2 = |J_1| + 2h \qquad (J_1 < 0) \tag{33}$$

The basic parameter is connected with the probabilities

$$(+\,+\,+) = p, \qquad (+\,+\,-) = (+\,-\,-) = (-\,-\,+) = (-\,+\,+) = (1-p)/4$$

As usual, to take into consideration a contribution of single-spin–flips into the reduced free energy, we must extend the probability space up to $D = 5$. According to this extension we need to introduce two more parameters

$$q = (+ + + +) \quad \text{and} \quad r = (+ + + + +)$$

The relevant, energetically favored single-spin–flips can easily be found; they are

$$+ + \oplus + + \qquad (\Delta = |J_1| + 2J_0) \tag{34}$$

$$+ + \oplus - - \quad \text{and} \quad - - \oplus + + \qquad (\Delta = 2h + 2J_0) \tag{35}$$

Their total energies are quoted in parentheses. There exist also the double-spin–flips

$$\begin{aligned} &+ + \oplus - \ominus \quad \text{and} \quad \ominus - \oplus + + && (\Delta = 4J_0) \\ &+ + \oplus \oplus + + && (\Delta = 4J_0) \end{aligned} \tag{36}$$

whose in-chain excitation energies vanish. Comparing the excitation energies (34)–(36) we recognize three distinct possibilities:

$$|J_1| < 2h, \qquad |J_1| < 2J_0 \tag{34'}$$

$$2h < |J_1|, \qquad h < J_0 \tag{35'}$$

$$2J_0 < |J_1|, \qquad J_0 < h \tag{36'}$$

If the inequalities (34′) hold, the significant spin–flips (34) yield the following expression of the reduced free energy:

$$f = \xi p - T w_0 w_1 r$$

where $w_1 = \exp(-|J_1|/T)$. The probability space (p, q, r) contains four vertices. Two of them, $(0,0,0)$ and $(1,1,1)$, correspond to the phases $\langle 2.\bar{2}\rangle$ and F, respectively. Only they are stable in this case. Hence, condition (34′) leaves the boundary F–$\langle 2.\bar{2}\rangle$ stable.

In contrast, spin–flips (35) lead to the reduced free energy

$$f = \xi p + 2T w_0 w_h q \tag{37}$$

which has a positive q term. Equation (37) can be reproduced using the probability of the significant spin–flips (35) which are

$$(+ + + - -) = p - q$$

The nature of the free energy (37) implies the splitting of the boundary in question. The new phase $\langle 3.\bar{2}\rangle$ appears in between F– and $\langle 2.\bar{2}\rangle$–phases. Along the boundary F–$\langle 3.\bar{2}\rangle$ some of the periodic phases, for example, $\langle 4.\bar{2}\rangle$, belong to the family of degenerate structures. A further analysis of the low-temperature expansion shows that the boundary F–$\langle 3.\bar{2}\rangle$ is stable owing to the spin–flips:

$$+ + \oplus + + \quad \text{or} \quad + + \oplus \oplus + +$$

In the region (36′) the original boundary F–$\langle 2.\bar{2}\rangle$ splits, thus generating the intermediate phase $\langle 3.\bar{2}\rangle$. One of the new boundaries, namely, F–$\langle 3.\bar{2}\rangle$, is also stable as in the case considered before.

Now we concentrate our attention on the analysis of the splitting of the boundary $\langle 2.\bar{2}\rangle$–$\langle 3.\bar{2}\rangle$. As above, introduce an abbreviated notation:

$$H = + + + - -$$

The additional parameter along the boundary $\langle E\rangle$–$\langle H\rangle$ is connected with the following probabilities:

$$(EE), \qquad (EH), \qquad (HE), \qquad (HH)$$

The significant spin–flips, which generate the additional parameter include three and four individual spin–flips:

$$+ + \oplus - \ominus + \oplus - \quad \text{and} \quad - \oplus + \ominus - \oplus + + \qquad (\Delta = 2h + 6J_0)$$

$$+ + \oplus - \ominus + \oplus - \ominus \quad \text{and} \quad \ominus - \oplus + \ominus - \oplus + + \qquad (\Delta = 8J_0)$$

The probabilities of these pairs of spin–flips are both off-diagonal: (HE) and (EH). The boundary $\langle E\rangle$–$\langle H\rangle$ splits in both regions (35′) and (36′).

A more complicated situation arises along the new boundary $\langle EH\rangle$–$\langle H\rangle$. Here the additional parameter is connected with probabilities of the form

$$(HHH), \qquad (EHH), \qquad (HHE), \qquad (EHE)$$

The competitive spin–flips, which generate the additional degeneracy parameter, are written

$$+ + \oplus - \ominus + \oplus + \ominus - \oplus + + \qquad (\Delta = 10J_0 + |J_1|)$$

$$\left.\begin{array}{l} - - \oplus + \ominus - \oplus\oplus + \ominus - \oplus + + \\ + + \oplus - \ominus + \oplus\oplus - \ominus + \oplus - - \end{array}\right\} \qquad (\Delta = 12J_0 + 4h)$$

$$\left.\begin{array}{l} \ominus - \oplus + \ominus\ominus\oplus\oplus + \ominus - \oplus + + \\ + + \oplus - \ominus + \oplus\oplus\ominus\ominus + \oplus - \ominus \end{array}\right\} \qquad (\Delta = 16J_0)$$

Comparing the total excitation energies we find that if $|J_1| < 2J_0 + 4h$ and $|J_1| < 6J_0$ the most significant spin–flip corresponds to the diagonal probability (HHH) and, consequently, the boundary $\langle EH\rangle$–$\langle H\rangle$ is stable. In contrast, the probabilities of other pairs of spin–flips are off-diagonal. So, in the region defined by either of inequalities $|J_1| > 2J_0 + 4h$ and $|J_1| > 6J_0$, the boundary $\langle EH\rangle$–$\langle H\rangle$ splits according to the scheme

$$\langle EH\rangle\text{–}\langle EH^2\rangle\text{–}\langle H\rangle$$

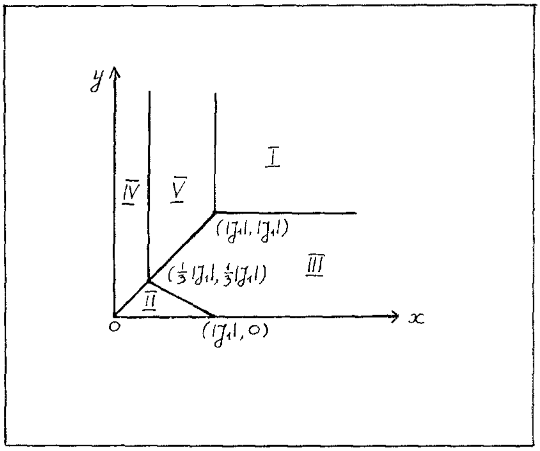

Fig. 8. Regions of different types of phase transition along the original boundary F–$\langle 2.\bar{2}\rangle$ are shown at fixed coupling constant $|J_1|$ (with $x = 2J_0, y = 2h$).

To consider a splitting of the boundary $\langle E\rangle$–$\langle EH\rangle$ we need to take into account the significant spin–flips which generate the additional parameter corresponding to this boundary, namely,

$$\left.\begin{array}{l} + + \oplus - \ominus + \oplus - \ominus + \oplus - - \\ - - \oplus + \ominus - \oplus + \ominus - \oplus + + \end{array}\right\} \quad (\Delta = 10J_0 + 2h)$$

$$\left.\begin{array}{l} + + \oplus - \ominus + \oplus - \ominus + \oplus - \ominus \\ \ominus - \oplus + \ominus - \oplus + \ominus - \oplus + + \end{array}\right\} \quad (\Delta = 12J_0)$$

In spite of the spin–flips of different kinds presented above, these correspond to the same off-diagonal probabilities (EEH) and (HEE). Thus, the boundary splits everywhere, excluding the region (34′). Figure 8 displays the various regions of splitting of the original boundary.

7.2. The Boundary F–$\langle 2.\bar{2}\rangle$ for $2h < |J_1|$, $h < J_0$

Following the approach of Section 6.2 we introduce the elementary spin–flip sequences constructed from the original "particles" E and H. According to the relationships (35′) between the coupling constants and the magnitude of the magnetic field the most significant spin–flips must include the following elements:

$$E_1, \qquad E_2, \qquad H_1 = \oplus\oplus + \ominus - \qquad H_2 = + \oplus\oplus - \ominus \tag{38}$$

The excitation energies of elements E and H (Δ_E and Δ_H) satisfy the inequalities

$$\Delta_E \geqslant 4J_0, \qquad \Delta_H \geqslant 6J_0 + 2h$$

These inequalities saturate, if the elements E and H are situated according to the ordering, defined by transition rules

$$\begin{pmatrix} E_1 \\ H_1 \end{pmatrix} \to \begin{pmatrix} E_1 \\ H_1 \end{pmatrix}, \qquad \begin{pmatrix} E_2 \\ H_2 \end{pmatrix} \to \begin{pmatrix} E_2 \\ H_2 \end{pmatrix}$$

First, consider the boundary $\langle E \rangle$–$\langle EH \rangle$. Along this boundary any possible degenerate periodic configuration $\langle G \rangle$ can be written as

$$\ldots E^{n_1}HE^{n_2}H \ldots E^{n_k}HE^{n_1} \ldots$$

or

$$\langle G \rangle = \langle E^{n_1}HE^{n_2}H \ldots E^{n_k}H \rangle \tag{39}$$

As explained in Section 6.2 we have to find the most significant spin–flips which generate the additional parameter along the boundary $\langle EG \rangle$–$\langle G \rangle$. They look like

$$\begin{matrix} + + \oplus - \ominus E_2^{n_1}H_2 \ldots E_2^{n_k}H_2E_2^{n_1} + \oplus - - \\ - - \oplus + \ominus - E_1^{n_1}H_1 \ldots E_1^{n_k}H_1E_1^{n_1} \oplus + + \end{matrix} \tag{40}$$

The probabilities of spin–flips (40) have the off-diagonal form

$$(H\tilde{G}E) \quad \text{and} \quad (E\tilde{G}H), \qquad \text{where} \quad \tilde{G} = GE^{n_1}$$

Hence the boundary $\langle EG \rangle$–$\langle G \rangle$ splits.

Arguments analogous to those presented in Section 6.2 imply the existence of a "complete devil's staircase" sequence of phase transitions.

The above considerations hold if $|J_1| > 2J_0 + 4h$ (region II in Fig. 8). The reverse inequality leads to significant spin–flips of a new kind, in which the indirect H_0-mediated transition

$$E_2 \to H_0 \to E_1$$

becomes allowed. Here the elementary sequence H_0 is $+ \oplus + \ominus -$. The excitation energy of the element H_0 in the allowed sequence is $4J_0 + |J_1|$. One representative of the set of most significant spin–flips is

$$+ + \oplus - \ominus E_2^{n_1}H_2 \ldots E_2^{n_j}H_0E_1^{n_{j+1}}H_1 \ldots H_1E_1^{n_1} \oplus + + \tag{41}$$

The total number of spin–flips in (41) is lower by one unit than that in (40) and the difference of the total excitation energies of (41) and (40) is equal to

$$|J_1| - (2J_0 + 4h)$$

Note that the sequences (41) correspond to the diagonal probability

$(H\tilde{G}H)$. Therefore almost all of the possible boundaries in the region $|J_1| < 2J_0 + 4h$ become stable, except the simple cascade of phase transitions written below:

$$\langle H\rangle\text{–}\langle HE\rangle\text{–}\langle HE^2\rangle\text{–} \ldots \text{–}\langle HE^n\rangle\text{–} \ldots \text{–}\langle E\rangle$$

These results resemble those obtained in Section 6.2.

This analogy can be made complete after the boundary $\langle EH\rangle$–$\langle H\rangle$ stability is investigated. Instead of (39) we must consider the other sequence

$$\ldots H^{m_1}EH^{m_2}E \ldots H^{m_k}EH^{m_1} \ldots \tag{42}$$

For reference we quote all the relevant spin–flips in the corresponding regions. Two of the most significant ones at $|J_1| > 2J_0 + 4h$ are

$$\begin{aligned} &+ + \oplus - \ominus H_2^{m_1}E_2 \ldots H_2^{m_k}E_2H_2^{m_1} + \oplus - - \\ &- - \oplus + \ominus - H_1^{m_1}E_1 \ldots H_1^{m_k}E_1H_1^{m_1} \oplus + + \end{aligned} \tag{43}$$

The set of $(\sum_{j=1}^{k} m_j + m_1 - 1)$ different sequences of spin–flips arises owing to the energetically favored "impurity" H_0 at $|J_1| < 2J_0 + 4h$. One of these sequences is

$$+ + + - - H_2^{m_1}E_2 \ldots H_2^{m_i - m}H_0H_1^{m-1}E_1 \ldots E_1H_1^{m_1} + + + \tag{44}$$

There are off-diagonal probabilities of spin–flips (43) and a diagonal one of the set of spin–flips (44).

We can reformulate the conclusion of Section 6.2 slightly changing the symbols and inequalities, as follows

If $|J_1| < 2J_0 + 4h$ (region III in Fig. 8), there exists only one infinite cascade of phase transitions, namely

$$\langle H\rangle\text{–}\langle EH\rangle\text{–}\langle E^2H\rangle\text{–} \ldots \text{–}\langle E^nH\rangle\text{–} \ldots \text{–}\langle E\rangle$$

which splits in the region $|J_1| > 2J_0 + 4h$ generating a "complete devil's staircase" sequence of phase transitions with the phases $\langle E\rangle$ and $\langle H\rangle$ as limits. As before, the crossover region, $|J_1| - 2J_0 - 4h \sim T$, contains an infinite number of triple points.

7.3. The Boundary F–$\langle 2.\bar{2}\rangle$ for $J_0 < h$, $2J_0 < |J_1|$

In this section we employ the ideas of Sections 6.2 and 7.2 to establish the sequence of the phase transitions. We shall not refer again to the results and conclusions obtained in those sections.

If the coupling constants obey $|J_1| > 6J_0$, only the following elementary spin–flips within E and H particles are allowed:

$$E_1, \qquad E_2, \qquad E_5 = \oplus + \ominus\ominus$$

$$H_1, \qquad H_4 = + \oplus\oplus\ominus\ominus, \qquad H_3 = \oplus\oplus + \ominus\ominus$$

These are consistent with the vanishing of an in-chain excitation energy of a whole spin–flip sequence. The allowed transitions from one element to another are

$$\begin{pmatrix} E_1 \\ H_1 \end{pmatrix} \to \begin{pmatrix} E_1 \\ E_5 \end{pmatrix}, \qquad E_2 \to \begin{pmatrix} E_2 \\ H_4 \end{pmatrix}, \qquad \begin{bmatrix} E_5 \\ H_3 \\ H_4 \end{bmatrix} \to \begin{bmatrix} E_2 \\ H_1 \\ H_3 \\ H_4 \end{bmatrix} \tag{45}$$

Generally the boundary $\langle E \rangle$–$\langle EH \rangle$ may split creating a set of periodic structures, such as

$$\ldots E^{n_1} H E^{n_2} H \ldots E^{n_k} H E^{n_1} \ldots$$

or (46)

$$\langle G \rangle = \langle E^{n_1} H E^{n_2} H \ldots E^{n_k} H \rangle$$

For the sake of simplicity consider the possible boundary $\langle G \rangle$–$\langle EG \rangle$. In this case the most significant spin–flips with vanishing in-chain excitation energy look like

$$+ + \oplus - \ominus E_2^{n_1} H_4 E_2^{n_2} H_4 \ldots E_2^{n_k} H_4 E_2^{n_1} + \oplus - \ominus \tag{47}$$

To this one adds the inverted sequence. These spin–flips correspond to the off-diagonal probabilities $(H\tilde{G}E)$ and $(E\tilde{G}H)$, where $\tilde{G} = GE^{n_1}$. In the region $|J_1| > 6J_0$, marked IV in Fig. 8, there exists a "complete devil's staircase" with the phases $\langle E \rangle$ and $\langle EH \rangle$ as original ones.

Now consider the boundary $\langle EH \rangle$–$\langle H \rangle$ in the same region of coupling constants. The basic spin–flips along this original boundary are

$$+ + \oplus - \ominus H_4 + \oplus - \ominus$$

and

$$\ominus - \oplus + \ominus \ominus H_1 \oplus + +$$

They have the off-diagonal probabilities (HHE) and (EHH), hence the boundary $\langle EH \rangle$–$\langle H \rangle$ splits.

Two new boundaries appear, namely, $\langle EH \rangle$–$\langle EH^2 \rangle$ and $\langle EH^2 \rangle$–$\langle H \rangle$. The latter is stable. This can easily be checked, because here the most significant spin–flip has the form

$$+ + \oplus - \ominus H_4 H_1 \oplus + + \tag{48}$$

Its probability is diagonal, namely, (HH^2H).

Along the boundary $\langle EH \rangle$–$\langle EH^2 \rangle$ the basic spin–flips can be written using the transitional rules (45) as

$$+ + \oplus - \ominus H_4 E_2 H_4 + \oplus - \ominus \tag{49}$$

and

$$\ominus - \oplus + \ominus\ominus H_1 E_5 H_1 \oplus + +$$

Obviously the probabilities of (49) are both off-diagonal: (HGE) and (EGH), where $G = HEH$. The boundary splits and we must elucidate the further behavior of the new pair of boundaries: $\langle EH \rangle$–$\langle EHEH^2 \rangle$ and $\langle EH^2 \rangle$–$\langle EHEH^2 \rangle$. Along the latter the most significant spin–flips include two sequential H elements, as in the case (48), namely,

$$+ + \oplus - \ominus H_4 E_2 H_4 H_1 E_5 H_1 \oplus + + \tag{50}$$

The probability of this is diagonal, thus making the boundary stable.

There also exist spin–flips with dimerized sequences of E and H elements, as in the sequence (49) which is dissimilar to (50). These become the basic ones along the boundaries of a kind $\langle (EH)^n EH^2 \rangle$–$\langle (EH)^{n-1} EH^2 \rangle$ and have the form

$$+ + \oplus - \ominus H_4 E_2 H_4 E_2 H_4 \ldots H_4 E_2 H_4 + \oplus - \ominus$$

and (51)

$$\ominus - \oplus + \ominus\ominus H_1 E_5 H_1 E_5 H_1 \ldots H_1 E_5 H_1 \oplus + +$$

The probabilities of (51) are both off-diagonal. Hence the conclusion: the boundary $\langle EH \rangle$–$\langle H \rangle$ generates an infinite cascade of phase transitions:

$$\langle H \rangle\text{–}\langle EH^2 \rangle\text{–}\langle EH^2EH \rangle\text{–} \ldots \text{–}\langle EH^2(EH)^n \rangle\text{–} \ldots \text{–}\langle EH \rangle$$

In the region $|J_1| < 6J_0$ the boundary $\langle E \rangle$–$\langle H \rangle$ can be easily investigated by considering the indirect H_0-mediated transition

$$E_2 \to H_0 \to E_1$$

which leads, as in Section 7.2, to the following cascade of the phase transitions:

$$\langle H \rangle\text{–}\langle EH \rangle\text{–}\langle E^2H \rangle\text{–} \ldots \text{–}\langle E^nH \rangle\text{–} \ldots \text{–}\langle E \rangle$$

The region $|J_1| < 6J_0$ is marked V in Fig. 8. Finally, the crossover region $|J_1| - 6J_0 \sim T$ contains an infinite number of triple points, as depicted in Fig. 7.

8. CONCLUSION

The low-temperature expansion for free energy has been employed to construct the phase diagram of the ANNNI model in an external magnetic field. The phase diagram pattern found is much more complicated than that found in Refs. 8–9 for the case of zero magnetic field. The principal difference is in the existence of several "complete devil's staircase" sequences of phase transitions. This is, indeed, surprising for the conventional

ANNNI model. It is worth emphasizing that Smith and Yeomans[13] have investigated the boundary F–$\langle 2.\bar{2}\rangle$. They obtained the infinite cascade of phase transitions, coinciding in the regions III and V (Fig. 8) with that found in the present work. If coupling constants map into areas II and IV (Fig. 8) a more accurate consideration of excitations above the degenerate ground state must be taken into account (see Sections 7.1–7.3).

It would be interesting to find the low-temperature crossover regime, connecting the behavior of the ANNNI model in zero to that in the finite magnetic field. The previous analysis[11] of the crossover regime in the magnetic field refers only to the highly anisotropic ANNNI model with $J_0 \gg J_1, J_2$.

Another interesting problem, briefly considered in the present paper, is the temperature crossover, leading to sequences of bifurcations, as depicted qualitatively in Fig. 7.

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APPENDIX

Here we exhibit several in-chain spin sequences and configurations of flipped spins used throughout the paper:

$$A = ++-, \qquad B = +++-, \qquad C = +-$$
$$E = ++--, \qquad H = +++--$$
$$A_1 = \oplus + \ominus, \qquad A_2 = + \oplus \ominus, \qquad A_3 = \oplus\oplus\ominus, \qquad \bar{C} = \oplus\ominus$$
$$E_1 = \oplus + \ominus -, \qquad E_2 = + \oplus - \ominus, \qquad E_3 = + \oplus \ominus \ominus$$
$$E_4 = \oplus\oplus\ominus\ominus, \qquad E_5 = \oplus + \ominus\ominus$$
$$H_0 = + \oplus + \ominus -, \qquad H_1 = \oplus\oplus + \ominus -, \qquad H_2 = + \oplus \oplus - \ominus$$
$$H_3 = \oplus\oplus + \ominus\ominus, \qquad H_4 = + \oplus\oplus\ominus\ominus$$

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