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1998 J. Phys. A: Math. Gen. 31 4871

(http://iopscience.iop.org/0305-4470/31/21/006)

J. Phys. A: Math. Gen. **31** (1998) 4871–4883. Printed in the UK PII: S0305-4470(98)89287-1

Low-temperature phase diagram and critical behaviour of the four-state chiral clock model

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Received 17 November 1997

Abstract. The low-temperature behaviour of the four-state chiral clock model is re-examined using a systematic low-temperature series expansion of the free energy. Previously obtained results for the low-temperature phases are corrected and the low-temperature phase diagram is derived. In addition, the phase transition from the modulated region to the high-temperature paraphase is shown to belong to the universality class of the three-dimensional XY-model.

1. Introduction

Uniaxially modulated structures are observed in very different classes of magnetic and ferroelectric substances. In many cases they exhibit rather complex phase diagrams with large varieties of phases. Phase transitions from a high-temperature paramagnetic or paraelectric phase (paraphase) to commensurately and incommensurately modulated phases occur, as external control parameters such as temperature and elastic stresses are varied. Microscopic models are successfully used for the description of these modulated systems [1]. An interesting example is the p-state chiral clock model [2], whose Hamiltonian is

$$H = -J_0 \sum_{\alpha} \sum_{\langle ij \rangle} \cos\left[\frac{2\pi}{p}(n_{i,\alpha} - n_{j,\alpha})\right] - J \sum_{i} \sum_{\alpha} \cos\left[\frac{2\pi}{p}(n_{i,\alpha} - n_{i,\alpha+1} + \Delta)\right]. \qquad (1)$$

α labels the layers perpendicular to the direction of the modulation (chiral direction) and i, j the crystal units in these layers. $\langle ij \rangle$ runs over neighbouring pairs in the layers. The integer variables $n_{i,\alpha}$ describe the state of the unit (i, α). They assume one of the values from 0 to $p - 1$. Below they are called spins. The two terms in equation (1) describe couplings ($J_0 > 0$, $J > 0$) between nearest neighbours in the same and adjacent layers, respectively.

In the ground state every layer is ferromagnetically ordered. Depending on the value of Δ, various ordering patterns of the different layers are realized. For $0 \leqslant \Delta < \frac{1}{2}$ nearest neighbours in the chiral direction couple ferromagnetically (ferromagnetic bond), thus leading to a ferromagnetic ground state where all spins are equal. For $\frac{1}{2} < \Delta \leqslant 1$ the spin increases by one for successive layers (chiral bond), thus yielding the right-handed chiral pattern

$$\ldots 012 \ldots (p-1)01 \ldots$$

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$\Delta = \frac{1}{2}$ is a multiphase point at which an infinity of different phases are degenerate since ferromagnetic and chiral bonds have the same energy.

Whereas the three-state model ($p = 3$) [3, 4] has been very thoroughly investigated, few results are known for the general case $p \geqslant 4$. There are derivations of the low-temperature phase diagram of the general p-state model by an expansion of the free energy in the vicinity of the multiphase point [2] as well as by a low-temperature mean-field theory [5], in which it was claimed that, for the four-state model ($p = 4$), only the phases $\langle 12^k \rangle$, $\langle 12^k 12^{k+1} \rangle$, $\langle 2^k 3 \rangle$, $\langle 2^k 32^{k+1} 3 \rangle$, $\langle 4 \rangle$, and $\langle \infty \rangle$ ($k = 0, 1, 2, \ldots$) are stable at low-temperatures. $\langle u_1 \ldots u_r \rangle$ is a shorthand notation for the phase with a period consisting of r bands with $u_1, u_2, \ldots, u_r$ layers with spins n, $n+1, \ldots, n+r$ (all modulo p) respectively. The phase $\langle 12 \rangle$, for example, is given by the layer sequence

$$\ldots 011233011 \ldots$$

The ferromagnetic and chiral ground states are denoted by $\langle \infty \rangle$ and $\langle 1 \rangle$ respectively.

McCullough [6] investigated the phase diagram for $p = 3$, 4, and 5 using the mean-field transfer-matrix (MFTM) method. From the numerical extrapolation of the data it was concluded that the low-temperature phase diagrams for $p = 3$ and $p = 4$ were consistent with the results of the low-temperature series expansion [2, 7]. It is interesting, that, for $p = 5$, new phases not predicted by the low-temperature series expansion [2] were found to be stable at low temperatures.

Scholten and King [8] presented Monte Carlo simulations of the four- and six-state models. They investigated especially the transition from the modulated phases to the ferromagnetic phase (i.e. $\Delta < \frac{1}{2}$). As it was not possible to resolve particular phases, they determined the 'interface spacing' as the average number of layers in a band for a given phase. They claimed that, for $\Delta = 0.45$, the results were not inconsistent with the predictions in [2]. In the case $p = 4$ and $\Delta = 0.2$ new phases with an interface spacing larger than the interface spacings of the phases predicted in [2] were observed close to the transition to the ferromagnetic phase.

Recently the four-state chiral clock model was shown [9] to be a special case of the double Ising spin (DIS) model [10–12], which was introduced to describe uniaxially modulated ferroelectrics.

In the following new results for the four-state chiral clock model (CC_4) are presented. In sections 2 and 3 we will re-examine the low-temperature phase diagram and discuss discrepancies with previous results. In section 4 it is shown that the transition from the modulated phases to the paramagnetic phase belongs to the universality class of the three-dimensional XY-model, and in section 5 short conclusions are given.

2. The low-temperature series expansion

The present series expansion technique for the CC_4 model is similar to the method developed by Fisher and Selke [13] for the axial next-nearest-neighbour Ising (ANNNI) model. At low temperatures the reduced free energy per spin $f = \frac{F}{Nk_BT}$ (N is the total number of spins) may be expanded in the form [13]

$$f = \frac{E_0}{k_B T} - \frac{1}{N} \sum_{n \geqslant 1} \Delta Z_N^{(n)}. \tag{2}$$

$\Delta Z_N^{(n)}$ is the total contribution to the partition function from configurations in which n spins have flipped (in comparison with the ground state). E_0, the ground-state energy per spin,

can be expressed [2, 7] in terms of the structural variables [13] $l_k = L_k/L$ (L_k: number of k-layer bands; L: total number of layers):

$$E_0(\{l_k\}) = -\tfrac{1}{2}q_\perp J_0 - J_1 - J_1\delta\sum_{k\geqslant 1} l_k$$

with $J_1 = J\cos(\frac{\pi}{2}\delta)$ and $\delta = \tan(\frac{\pi}{2}\Delta) - 1$. The number of nearest neighbours in the layers is $q_\perp$; it is 4 for the primitive cubic lattice.

The contributions $\Delta Z_N^{(n)}$ are expressed in terms of the elementary Boltzmann factors

$$w = \exp(-K_0) \qquad x = \exp\left(-2K\cos\left(\frac{\pi}{2}\Delta\right)\right)$$

and

$$y = x^{1+\delta} = \exp\left(-2K\sin\left(\frac{\pi}{2}\Delta\right)\right)$$

with $K_0 = J_0/(k_BT)$ and $K = J/(k_BT)$. The reduced free energy per spin can be expanded in a convergent power series of w, provided that $x, y \gg w^{q_\perp}$. At low temperatures (i.e. $T \ll J$, which is assumed throughout this paper) this condition holds for $q_\perp J_0 > 2J\max[\cos(\frac{\pi}{2}\Delta), \sin(\frac{\pi}{2}\Delta)]$. This is clearly satisfied if $q_\perp J_0 > 2J$. The weight w results from changing an in-layer bond between spins with equal values to a bond between spins with values differing by 1. The lowest orders involved are $w^{q_\perp}$ (overturning one spin), $w^{2q_\perp-2}$ (overturning two neighbouring spins in one layer) and $w^{2q_\perp}$ (overturning two spins not being in-layer nearest neighbours).

There are three possible environments of a given spin (the numbers in parentheses are the values of the spins in three consecutive layers where the considered spin belongs to the middle layer): (a) spins with two ferromagnetic bonds in the chiral direction (e.g. $0\hat{0}0$), (b) spins with one ferromagnetic and one chiral bond (e.g. $0\hat{1}1$), and (c) spins with two chiral bonds (e.g. $0\hat{1}2$).

Let us discuss, as an example, the contribution to $\Delta Z_N^{(1)}$ (first-order term in equation (2)) for case (a). By overturning one spin, three different final states can be obtained (m being the initial state): $(m+1)$ mod 4, $(m+2)$ mod 4, and $(m+3)$ mod 4. This leads to the Boltzmann factor

$$\begin{aligned}\sum_{n=1}^{3}\exp[-(E_f(n) - E_i)/(k_BT)] &= \sum_{n=1}^{3}\left(\exp\left\{2K\cos\left(\frac{\pi}{2}\Delta\right)\left[\cos\left(\frac{\pi}{2}n\right) - 1\right]\right\}\right.\\ &\quad\left.\times\exp\left\{q_\perp K_0\left[\cos\left(\frac{\pi}{2}n\right) - 1\right]\right\}\right)\\ &= xw^{q_\perp} + x^2w^{2q_\perp} + xw^{q_\perp}. \qquad (3)\end{aligned}$$

It is obvious from equation (3) that the process $m \longrightarrow (m+2)$ mod 4 does not contribute to the lowest-order term in the expansion, as it has the same in-layer Boltzmann factor $w^{2q_\perp}$ as the higher-order process by which the values of two uncoupled spins change by 1. In fact, this process of the order $w^{2q_\perp}$ does not even contribute to the lowest-order correction term, which is of the order $w^{2q_\perp-2}$ (flipping of two neighbouring spins in one layer [13]).

In [2] the following contribution to $\Delta Z_N^{(1)}$ for case (a) is given:

$$\begin{aligned}\sum_{n=1}^{3}\exp[-(E_f(n) - E_i)/(k_BT)] &= \sum_{n=1}^{3}\exp\left\{2K\cos\left(\frac{\pi}{2}\Delta\right)\left[\cos\left(\frac{\pi}{2}n\right) - 1\right]\right\}\omega^{q_\perp}\\ &= (x + x^2 + x)\omega^{q_\perp} \qquad (4)\end{aligned}$$

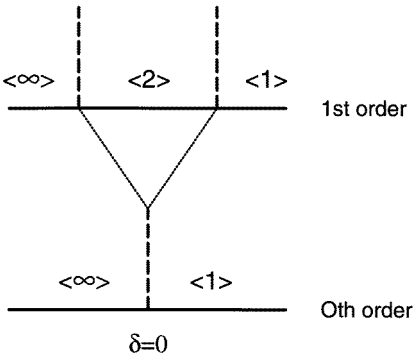

Figure 1. Schematic phase diagram showing, for given small value of T, the phase sequence on a line along which δ varies. Lower horizontal line: sequence in zeroth order of the expansion (exact for $T = 0$); upper line: first-order results. $\delta = 0$ is the multiphase point. Broken lines indicate boundary lines at which an infinity of phases are degenerate.

with

$$\omega = \sum_{n=1}^{3} \exp\left\{K_0\left[\cos\left(\frac{\pi}{2}n\right) - 1\right]\right\} = w + w^2 + w. \tag{5}$$

A comparison of equations (4) and (5) with equation (3) reveals that the treatment of the in-layer bonds is erroneous in [2]. The free energy is written in [2] as an expansion in terms of the (erroneous) Boltzmann factor ω. As a consequence, contributions from different orders of the expansion are treated in [2] as if they were of the same order. Thus, in our example, the term x^2, resulting from the process $m \longrightarrow (m+2)$ mod 4 and contributing to a higher-order correction in the polynominal expansion in w (see equation (3)), contributes to the lowest order in [2] (see equation (4)). This error is repeated for all considered spin configurations and for all considered p-state models ($p \geqslant 4$), thus leading to a wrong low-temperature phase diagram not only for the CC_4 model, but also for the generalized p-state chiral clock model with $p \geqslant 4$. One should emphasize that the treatment of the in-layer bonds is correct in the analyses of the CC_3 model [7].

With the correct contributions, the reduced free energy (2) in first order is given by

$$f = -\tfrac{1}{2}q_\perp K_0 - K_1 - \tfrac{1}{2}K_1\delta - (1+xy)w^{q_\perp} + a_1(\delta)l_1 + \sum_{k\geqslant 3} a_k(\delta)l_k + \mathrm{O}(w^{2q_\perp - 2}) \tag{6}$$

with

$$a_1(\delta) = -\tfrac{1}{2}K_1\delta - (2y - xy - 1)w^{q_\perp}$$

and

$$a_k(\delta) = (k-2)[\tfrac{1}{2}K_1\delta - (2x - xy - 1)w^{q_\perp}].$$

The set of structural variables l_k minimizing f for given values of δ and T determine the stable phases occurring in first order (see figure 1): the $\langle\infty\rangle$-, the $\langle 1\rangle$-, and the $\langle 2\rangle$-phase. Phases $\langle\infty\rangle$ and $\langle 2\rangle$ are, in this order of the expansion, separated by a boundary, at which all phases that are degenerate at the multiphase point and that do not contain 1-layer bands have the same free energy. Likewise, phases containing only 1- and 2-layer bands are still degenerate on the boundary between the $\langle 1\rangle$- and $\langle 2\rangle$-phase.

One could now proceed in considering processes involving two spins, then three spins and so on. This is very cumbersome and only feasible for processes involving few spins. In the next section the phases stable in general order in the series expansion will be determined using a transfer-matrix method.

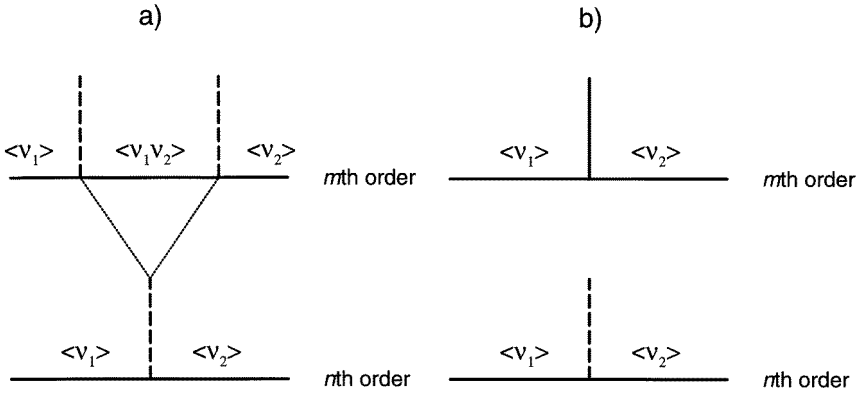

Figure 2. Schematic phase diagram in the vicinity of the boundary between two stable phases $\langle\nu_1\rangle$ and $\langle\nu_2\rangle$. The horizontal lines correspond, as in figure 1, to a given value of T and present results of nth and mth order. (*a*) The phase $\langle\nu_1\nu_2\rangle$ is stable at higher order leading to new boundary lines. (*b*) The phase $\langle\nu_1\nu_2\rangle$ is not stabilized leading to a true phase boundary between the two phases $\langle\nu_1\rangle$ and $\langle\nu_2\rangle$. The broken line indicates a boundary at which an infinity of phases are degenerate, the full line indicates a true phase boundary.

3. Transfer-matrix method

3.1. Introductory remarks

One should first note that the Hamiltonian (1) is left invariant by the transformation

$$\begin{aligned} &\Delta \longrightarrow \Delta' = 1 - \Delta \\ &n_{i,\alpha} \longrightarrow n'_{i,\alpha} = (-n_{i,\alpha} + \alpha) \bmod 4. \end{aligned} \tag{7}$$

Therefore, the phase diagram of the CC_4 model is invariant under a reflection in the line $\Delta = \frac{1}{2}$. In the following we will discuss the low-temperature phase diagram for the case $\Delta > \frac{1}{2}$, i.e. we will analyse in detail the stability of the boundary line between the $\langle 1\rangle$- and $\langle 2\rangle$-phase, the phase diagram for $\Delta < \frac{1}{2}$ being inferred by the transformation (7).

In the ground state and low-temperature expansion every phase $\langle\nu\rangle$ consists of a periodic arrangement of a sequence of $n(\nu)$ layers called ν-sequences ($n(\nu)$ is the period of the phase). Suppose now that in a certain order of the series expansion two stable phases, $\langle\nu_1\rangle$ and $\langle\nu_2\rangle$, are separated by a boundary at which the phases produced by ν_1- and ν_2-sequences are degenerate (see figure 2). In first order the boundary under consideration separates the phases $\langle 1\rangle$ and $\langle 2\rangle$. At higher order a new phase $\langle\nu\rangle = \langle\nu_1\nu_2\rangle$ consisting of a structure with alternating ν_1- and ν_2-sequences might be stable in the vicinity of the boundary. If

$$a_\nu = f_{\langle\nu\rangle} - \frac{n(\nu_1)}{n(\nu_1)+n(\nu_2)} f_{\langle\nu_1\rangle} - \frac{n(\nu_2)}{n(\nu_1)+n(\nu_2)} f_{\langle\nu_2\rangle} \tag{8}$$

is negative, the new phase has a lower free energy than the phases $\langle\nu_1\rangle$ and $\langle\nu_2\rangle$ [13, 14] and it will be stabilized in the vicinity of the $\langle\nu_1\rangle : \langle\nu_2\rangle$ boundary (see figure 2(*a*)). The stability of the boundaries between the phases $\langle\nu_1\rangle$ and $\langle\nu_1\nu_2\rangle$ and the phases $\langle\nu_1\nu_2\rangle$ and $\langle\nu_2\rangle$ must then be examined at higher orders. If, on the other hand, a_ν is positive, the phase $\langle\nu_1\nu_2\rangle$ (and therefore every phase consisting of ν_1- and ν_2-sequences) has a higher free energy than $\langle\nu_1\rangle$ or $\langle\nu_2\rangle$. The boundary is a true phase boundary which remains stable in all orders of the low-temperature series expansion (see figure 2(*b*)).

The reader is referred to [13, 7] for details concerning the construction of the series expansion to general order.

3.2. Formulation in terms of transfer matrices and vectors

The sign of a_v, and therefore the stability of the phase $\langle v \rangle$, is determined by the leading term in its expansion in terms of w. This term is obtained by considering all flipping processes involving a spin chain of $n(v) - 1$ spins in $n(v) - 1$ different layers [7]. Besides the linear configuration with all $n(v) - 1$ spins connected, the various decompositions of this configuration into $2, 3, \ldots, n(v) - 1$ different parts must be taken into account. The contributions from these processes can be written as a product of transfer matrices and vectors. The matrices describe a bond between two flipping spins, the vectors an initial or a final bond preceding the first or following the last flipped spin respectively. Every spin can flip to three different values and hence 3×3 matrices occur. As we are only interested in the sign of the a_v we can restrict ourselves to the two processes contributing in lowest order, thus excluding the process $m \longrightarrow (m + 2) \bmod 4$ only relevant for the correction term. Of course, if one considers all possible processes (i.e. 3×3 matrices), the leading term is identical to the term obtained by the 2×2 matrices. This has already been noticed in the low-temperature analyses of a six-state clock model with competing axial nearest- and next-nearest neighbour couplings [14], where the corresponding 2×2 matrices have been considered instead of the general 5×5 matrices.

As two axial next-nearest-neighbours are either coupled by a ferromagnetic or by a chiral bond, only two different matrices are to be constructed. For a ferromagnetic or a chiral bond between two spins in the layers α and $\alpha + 1$ one obtains, respectively, the transfer matrices

$$\mathbf{F}_{\alpha,\alpha+1} = \begin{pmatrix} 1-x & x(1-y) \\ x(1-y^{-1}) & 1-x \end{pmatrix} w^{q_\perp} \tag{9}$$

and

$$\mathbf{C}_{\alpha,\alpha+1} = \begin{pmatrix} 1-y & y(1-x^{-1}) \\ y(1-x) & 1-y \end{pmatrix} w^{q_\perp}. \tag{10}$$

The matrix elements are the Boltzmann factors for a simultaneous change of the values of the two spins. The first (second) row corresponds to a change $\Delta n_{i,\alpha} = +1(-1)$ and the first (second) column to $\Delta n_{i,\alpha+1} = +1(-1)$. Every element of the matrices $\mathbf{F}$ and $\mathbf{C}$ is a sum of two terms, the first term resulting from changing the values of two axially coupled spins. As already mentioned, disconnected pairs of spins (i.e. two spins that are not neighbours to each other but neighbours to an unchanged spin) also contribute to the partition sum. Since every disconnected pair must be associated with a minus sign [13], the corresponding Boltzmann factors enter the different matrices with a negative sign.

The factor $w^{q_\perp}$ resulting from changing the in-layer bonds in layer $\alpha + 1$ is common to all elements of the matrices $\mathbf{F}$ and $\mathbf{C}$. This is a direct consequence of the fact that only flipping processes $m \longrightarrow (m \pm 1) \bmod 4$ are to be considered for obtaining the leading order in the expansion of a_v. For the full 3×3 matrices this is not the case as the flipping process $m \longrightarrow (m + 2) \bmod 4$ has the in-layer Boltzmann factor $w^{2q_\perp}$. In [2] the phase diagram has been determined to general order using 3×3 transfer matrices. Due to the erroneous treatment of the in-layer interactions (see section 2) the 'common term' $\omega^{q_\perp}$ has been factorized, thus leading, again, to the treatment of terms belonging to different orders as being of the same order.

Table 1. The leading orders b_ν in the expansion of the quantities a_ν (equation (8)) determining the stability of different families of phases consisting of 1- and 2-layer-bands.

ν	b_ν
1^k2	$-(\boldsymbol{a}_c^T-\boldsymbol{a}_f^T)\mathbf{C}^k(\boldsymbol{b}_f-\boldsymbol{b}_c)$
$1^k21^{k-1}2$	$-(\boldsymbol{a}_c^T-\boldsymbol{a}_f^T)\mathbf{C}^k\mathbf{F}\mathbf{C}^k(\boldsymbol{b}_f-\boldsymbol{b}_c)$
12^k	$-(\boldsymbol{a}_c^T-\boldsymbol{a}_f^T)(\mathbf{CF})^{k-1}\mathbf{C}(\boldsymbol{b}_f-\boldsymbol{b}_c)$
12^k12^{k+1}	$-(\boldsymbol{a}_c^T-\boldsymbol{a}_f^T)(\mathbf{CF})^k\mathbf{C}(\mathbf{CF})^k\mathbf{C}(\boldsymbol{b}_f-\boldsymbol{b}_c)$

A spin at the end of the spin chain is a neighbour of an unchanged spin. To determine the contributions of these spins, four different cases are to be distinguished: (a) the considered spin is the first spin of the chain and its bond to the left (i.e. to an unchanged spin) is a ferromagnetic or a chiral bond (subscripts f and c respectively) or (b) it is the last spin of the chain and its bond to the right is a ferromagnetic or a chiral bond. The Boltzmann factors for the flipping of these single spins are written as vectors:

$$\boldsymbol{a}_f=\begin{pmatrix}y^{-\frac{1}{2}}\\ y^{\frac{1}{2}}\end{pmatrix}x^{\frac{1}{2}}w^{q_\perp} \tag{11}$$

$$\boldsymbol{a}_c=\begin{pmatrix}x^{\frac{1}{2}}\\ x^{-\frac{1}{2}}\end{pmatrix}y^{\frac{1}{2}}w^{q_\perp} \tag{12}$$

$$\boldsymbol{b}_f=\begin{pmatrix}y^{\frac{1}{2}}\\ y^{-\frac{1}{2}}\end{pmatrix}x^{\frac{1}{2}} \tag{13}$$

$$\boldsymbol{b}_c=\begin{pmatrix}x^{-\frac{1}{2}}\\ x^{\frac{1}{2}}\end{pmatrix}y^{\frac{1}{2}}. \tag{14}$$

Vectors (13) and (14) do not include the Boltzmann factor resulting from the change of the in-layer bonds. This factor has already been included in the matrix describing the overturning of the two last spins in the spin chain.

3.3. Derivation of the low-temperature phase diagram

With matrices (9) and (10) and vectors (11)–(14) it is now possible to compute the leading-order term b_ν of the quantities a_ν (and, thus, to determine the sign of a_ν) for all phases degenerate at the multiphase point and containing only 1- and 2-layer bands. All considered phases can be viewed as periodic arrangements of spin sequences with a 1-layer band as the first and a 2-layer band as the last band in the sequence [7]. The sequence $\tilde{\nu}$ obtained by stripping the original sequence ν by its last and first band is called core. All sequences based on the same core $\tilde{\nu}$ enter in the computation of the b_ν: Sequences $1\tilde{\nu}2$ and $2\tilde{\nu}1$ contribute negatively, sequences $1\tilde{\nu}1$ and $2\tilde{\nu}2$ contribute positively [13]. The expressions b_ν for different families of phases are summarized in table 1.

3.3.1. Stability of some series of phases. For the series of phases $\langle 12^k\rangle$ the expression (see table 1)

$$b_{12^k}=-(\boldsymbol{a}_c^T-\boldsymbol{a}_f^T)(\mathbf{CF})^{k-1}\mathbf{C}(\boldsymbol{b}_f-\boldsymbol{b}_c) \tag{15}$$

gives the leading contribution to a_{12^k}. The four different sequences based on the core $\tilde{\nu}=2^{k-1}$ yield the four different contributions to b_{12^k}.

The eigenvalues $\exp(-\Gamma_\pm)$ of the matrix **CF** are real and positive. Expression (15) can be written in the form

$$b_{12^k} = A_+ \exp\left(-\frac{k}{2}\Gamma_+\right) + A_- \exp\left(-\frac{k}{2}\Gamma_-\right)$$

with $\Gamma_+ < \Gamma_-$. A close examination reveals that for finite temperatures $A_+ < 0$, $A_- > 0$ and $A_+ + A_- < 0$. Thus, b_{12^k} is negative for all k, i.e. all phases of the form $\langle 12^k\rangle$ spring from the multiphase point and have a finite stability range at temperatures above zero.

The leading-order contribution for the phases $\langle 1^k 2\rangle$ is

$$b_{1^k 2} = -(\boldsymbol{a}_c^T - \boldsymbol{a}_f^T)\mathbf{C}^k(\boldsymbol{b}_f - \boldsymbol{b}_c). \tag{16}$$

The eigenvalues of the matrix **C** are complex conjugate. They are written in the form

$$\xi_1 \pm \mathrm{i}\xi_2 = \exp(-\Gamma_0 \pm \mathrm{i}\Omega) \tag{17}$$

with $\Gamma_0 = -\frac{1}{2}\ln(\xi_1^2 + \xi_2^2) > 0$, $\Omega = \arctan(\frac{\xi_2}{\xi_1})$ and $\xi_1 = 1 - x^{1+\delta}$, $\xi_2 = (1-x)x^{\frac{1}{2}+\delta}$. We then obtain the expression

$$b_{1^k 2} = -(A_1 + \mathrm{i}A_2)(\xi_1 + \mathrm{i}\xi_2) + (A_1 - \mathrm{i}A_2)(\xi_1 - \mathrm{i}\xi_2) = -|\Delta|\exp(-k\Gamma_0)\cos(k\Omega + \phi)$$

with $|\Delta|\exp \mathrm{i}\phi = A_1 + \mathrm{i}A_2$. The temperature-dependent quantities $|\Delta|$, ϕ, Γ_0, and Ω do not depend on k.

$b_{1^k 2}$ is negative for small values of k. If k exceeds the value $k_{\max} = \frac{1}{\Omega}(\frac{\pi}{2} - \phi)$, then $b_{1^k 2}$ becomes positive and, thus, all phases with $k > k_{\max}$ are unstable at the considered point of the phase diagram. Since $k_{\max} \longrightarrow \infty$ for $T \longrightarrow 0$, there is, for every k, a temperature below which the phase $\langle 1^k 2\rangle$ is stable. Thus, all phases $\langle 1^k 2\rangle$ spring from the multiphase point, but the higher commensurate phases disappear at non-zero temperatures T_k, which decrease with increasing k. Such a cut-off of the high commensurate phases at finite temperatures is also observed in the ANNNI model [15].

Following the general line we also examined the series of phases $\langle 12^k 12^{k+1}\rangle$ and $\langle 1^k 21^{k-1}2\rangle$. For the case $\langle 12^k 12^{k+1}\rangle$ we find that all these phases are stable at finite temperatures in the vicinity of the multiphase point with no cut-off for the phases with a large value of k, i.e. the results for the series $\langle 12^k 12^{k+1}\rangle$ resemble the results for the series $\langle 12^k\rangle$. Analysing the leading contribution for the phases $\langle 1^k 21^{k-1}2\rangle$ we find a behaviour similar to the behaviour of the phases $\langle 1^k 2\rangle$, i.e. all phases with $k < k_{\max}$ (the value of $k_{\max}$ being series dependent) are stable and $k_{\max} \longrightarrow \infty$ as $T \longrightarrow 0$.

It should be emphasized that the low-temperature expansion yields a convergent power series only if $x \gg w^{q_\perp}$. For the CC_3 model [16] an analysis in Bethe approximation showed no cut-off of the low-commensurate phases $\langle 1^k 2\rangle$ with $k \leqslant 10$ for the case $J_0 = J$, in contrast to the corresponding low-temperature expansion [7]. In fact, for these low-commensurate phases the cut-off temperature is of the order of J. Results of the series expansion for such high temperatures are only reliable if $q_\perp J_0 \gg J$ is assumed. Therefore, the results of the expansion in [7] do not contain the case $J_0 = J$ considered in [16].

3.3.2. Phases containing general sequences of 1- and 2-layer bands. In the following we will show that all phases consisting only of 1- and 2-layer bands and obeying the rules of the structure combination spring from the multiphase point, the higher commensurate phases of some series becoming unstable at higher temperatures. The leading contribution to a_ν for all these phases is of the form (see table 1)

$$b_\nu = -(\boldsymbol{a}_c^T - \boldsymbol{a}_f^T)\mathbf{DC}(\boldsymbol{b}_f - \boldsymbol{b}_c) \tag{18}$$

where **D** is a product of powers of matrices **C** and (**CF**). The contributions of the first and last band are given by $(\boldsymbol{a}_c^T - \boldsymbol{a}_f^T)$ and $\mathbf{C}(\boldsymbol{b}_f - \boldsymbol{b}_c)$ respectively. A 1-layer band in the core contributes a matrix **C**, whereas a 2-layer band yields the matrix product (**CF**). The product over all bands in the core yields the matrix **D** (see equation (18)).

The diagonal elements of the matrix

$$\mathbf{CF} = \begin{pmatrix} 2(1-y)(1-x) & x(1-y)^2 - x^{-1}y(1-x)^2 \\ y(1-x)^2 - xy^{-1}(1-y)^2 & (1+xy)(1-y)(1-x) \end{pmatrix} w^{2q_\perp} \tag{19}$$

are positive whereas the non-diagonal elements are negative, since $y = x^{1+\delta}$ with $x \ll 1$. We now follow [14] and introduce the unitary matrix

$$\mathbf{U} = \begin{pmatrix} -1 & 0 \\ 0 & 1 \end{pmatrix} = \mathbf{U}^{-1}.$$

All elements of $\mathbf{U}(\mathbf{CF})\mathbf{U}$, and therefore of $\mathbf{U}(\mathbf{CF})^k\mathbf{U}$, are positive. This is also the case for the two vectors $(\boldsymbol{a}_c^T - \boldsymbol{a}_f^T)\mathbf{U}$ and $\mathbf{UC}(\boldsymbol{b}_f - \boldsymbol{b}_c)$, thus (see equation (15) and table 1)

$$(\boldsymbol{a}_c^T - \boldsymbol{a}_f^T)(\mathbf{CF})^{k-1}\mathbf{C}(\boldsymbol{b}_f - \boldsymbol{b}_c) = (\boldsymbol{a}_c^T - \boldsymbol{a}_f^T)\mathbf{UU}(\mathbf{CF})^{k-1}\mathbf{UUC}(\boldsymbol{b}_f - \boldsymbol{b}_c)$$

is positive, i.e. $b_{12^k} < 0$, in agreement with the aforementioned calculations.

Phases of the series $\langle 12^k 12^{k+1}\rangle$ contain a single 1-layer-band in the core yielding the matrix product $(\mathbf{CF})\mathbf{C}(\mathbf{CF})$ with positive diagonal and negative non-diagonal elements for small x. Hence, the product (see table 1)

$$\begin{aligned}(\boldsymbol{a}_c^T - \boldsymbol{a}_f^T)&(\mathbf{CF})^k\mathbf{C}(\mathbf{CF})^k\mathbf{C}(\boldsymbol{b}_f - \boldsymbol{b}_c) \\ &= (\boldsymbol{a}_c^T - \boldsymbol{a}_f^T)\mathbf{UU}(\mathbf{CF})^{k-1}\mathbf{UU}(\mathbf{CF})\mathbf{C}(\mathbf{CF})\mathbf{UU}(\mathbf{CF})^{k-1}\mathbf{UUC}(\boldsymbol{b}_f - \boldsymbol{b}_c)\end{aligned}$$

is positive, showing the stability of the phases $\langle 12^k 12^{k+1}\rangle$. Following this line of thought one easily shows that all phases appearing between the phases $\langle 2\rangle$ and $\langle 12\rangle$ (i.e. phases with only isolated 1-layer-bands in the core) are stable in the vicinity of the multiphase point. Indeed, as no new matrix products show up in the computation of the different b_ν, all these expressions can be written, using the matrix **U**, as a product of vectors and matrices having only positive elements.

For phases containing consecutive 1-layer-bands in the core the following additional vectors and matrices may contribute to the b_ν as can be seen from table 1: $\mathbf{UC}^k(\boldsymbol{b}_f - \boldsymbol{b}_c)$, $(\boldsymbol{a}_c^T - \boldsymbol{a}_f^T)\mathbf{C}^k\mathbf{U}$, and $\mathbf{U}(\mathbf{CF})\mathbf{C}^k\mathbf{U}$ with $k \geqslant 2$. Introducing the eigenvalues of the matrix **C** (see equation (17)), we obtain

$$\begin{aligned} \mathrm{v}_1 &= \exp(-k\Gamma_0)[x^{\frac{\delta}{2}}(1-x)\cos k\Omega + x^{-\frac{1+\delta}{2}}(1-x^{1+\delta})\sin k\Omega]w^{kq_\perp} \\ \mathrm{v}_2 &= \exp(-k\Gamma_0)[x^{-\frac{\delta}{2}}(1-x^{1+\delta})\cos k\Omega - x^{1+\delta}(1-x)\sin k\Omega]w^{kq_\perp} \end{aligned}$$

for the components of the vector $\boldsymbol{v} = \mathbf{UC}^k(\boldsymbol{b}_f - \boldsymbol{b}_c)$. Whereas v_1 is always positive, $v_2 > 0$ only if the inequality

$$\tan k\Omega < \frac{x^{-\frac{\delta}{2}}(1-x^{1+\delta})}{x^{1+\delta}(1-x)}$$

holds. This is the case for temperatures smaller than an upper limit which depends on δ and k. In a similar way one shows that for temperatures smaller than some k-dependent temperature all the components of the vector $(\boldsymbol{a}_c^T - \boldsymbol{a}_f^T)\mathbf{C}^k\mathbf{U}$ and of the matrix $\mathbf{U}(\mathbf{CF})\mathbf{C}^k\mathbf{U}$ are positive. The free energy differences a_ν for all phases containing consecutive 1-layer-bands in the core are therefore negative below a certain temperature, i.e. these phases possess a stability region below this temperature.

3.3.3. Conclusion. The results obtained so far can be summarized as follows. All phases consisting only of 1- and 2-layer-bands, that can be formed by means of the aforementioned structure combination rules, spring from the multiphase point, where they are degenerate. The higher commensurate phases of some series, i.e. those phases formed in higher orders of the combination process, disappear again at temperatures individually depending on the series under consideration.

From these results the complete low-temperature phase diagram of the CC_4 model is deduced by applying the transformation (7). At non-zero temperatures all phases appearing between the phases $\langle 12 \rangle$ and $\langle 3 \rangle$ are stable since the transformation (7) transforms phase $\langle 12 \rangle$ into $\langle 3 \rangle$ and leaves the phase $\langle 2 \rangle$ invariant. Some of the long commensurate phases appearing between the phases $\langle 1 \rangle$ and $\langle 12 \rangle$ for $\Delta > \frac{1}{2}$ and between the phases $\langle \infty \rangle$ and $\langle 3 \rangle$ for $\Delta < \frac{1}{2}$ are unstable at a given temperature. Upon reducing the temperature, increasingly more of these phases become stable, and in the limit $T \longrightarrow 0$ all phases obeying the rules of the structure combination are stable. Therefore, the CC_4 model exhibits a complete devil's staircase in the low-temperature limit.

3.4. Comparison with other work

The low-temperature behaviour of the general p-state chiral clock model was analysed in [2] using a series expansion technique similar to the one presented here. Due to the incorrect expansion (see section 2) only some specific families of phases were shown to possess a finite stability region at small temperatures. Particularly, it was claimed that the phases $\langle 1^k 2 \rangle$ with $k > 2$ are not stable at low temperatures, implying that, due to transformation (7), for $\Delta < \frac{1}{2}$ a direct transition from the ferromagnetic phase to the $\langle 4 \rangle$-phase exists. In order to corroborate these calculations a low-temperature mean-field analyses of the CC_p model was presented in [5] were it was claimed that in the vicinity of the multiphase point the mean-field approximation yields the same stable phases as [2]. In that work the model in mean-field approximation was mapped onto an one-dimensional array of interacting domain walls. This mapping was derived under the approximation that the mean-field average spin $(\langle \cos \frac{\pi}{2} n_{i,\alpha} \rangle_{\mathrm{MF}}, \langle \sin \frac{\pi}{2} n_{i,\alpha} \rangle_{\mathrm{MF}})$ in each layer (layer spin) does only deviate from the $T = 0$ value in amplitude but not in phase. In a detailed analyses of the mean-field phase diagram of the CC_3 model, Siegert and Everts [17] showed that this approximation leads to a wrong phase diagram at low temperatures, thus concluding that the layer spin must also be allowed to deviate in phase from its ground-state value. This should not only be the case for the three-state but also for the general p-state model. The results of [5] for the mean-field low-temperature behaviour of the CC_p model must therefore be considered with care.

In the preceding sections we have shown that the results of the series expansion in [2] are erroneous due to wrong Boltzmann factors for the in-layer bonds. In fact, the four-state model exhibits in the low-temperature limit a complete devil's staircase. Furthermore, it results from our calculations that no direct transition from the ferromagnetic to the $\langle 4 \rangle$-phase exists as phases with longer periods are stable between these two phases.

In the Monte Carlo simulation of the CC_4 model [8] long-period spin patterns were observed when going from the ferromagnetic phase to the modulated phases at rather high temperatures. In view of this work one must interpret these patterns as reflecting the existence of phases springing from the multiphase point and intercalating between the ferromagnetic and the $\langle 4 \rangle$-phases.

4. The critical behaviour

The critical behaviour of the general p-state chiral clock model at the transition to the paraphase is an interesting topic since for $p = 2$ the chiral clock model reduces to the anisotropic Ising model, for $p = \infty$ it corresponds to the classical three-dimensional XY-model. Siegert and Everts [18] showed that the CC_3 model belongs to the universality class of the three-dimensional XY-model. On the basis of his contradictory MFTM results, McCullough [6] speculated about a change in the universality class from three-dimensional Ising behaviour to three-dimensional XY-behaviour for p close to 5. In the following we will show that for $p = 4$ an effective Ginzburg–Landau–Wilson Hamiltonian can be derived which can be transformed to the effective Hamiltonian of the three-dimensional XY-model.

For the case $p = 4$ the Hamiltonian (1) can be rewritten in the form

$$H = -J \sum_i \sum_\alpha \boldsymbol{S}_{i,\alpha} \mathbf{R}(\Delta) \boldsymbol{S}_{i,\alpha+1} - J_0 \sum_\alpha \sum_{\langle ij \rangle} \boldsymbol{S}_{i,\alpha} \boldsymbol{S}_{j,\alpha}$$

where we introduced the spin vector $\boldsymbol{S}_{i,\alpha} = (\cos \frac{\pi}{2} n_{i,\alpha}, \sin \frac{\pi}{2} n_{i,\alpha})$ and the rotation matrix

$$\mathbf{R}(\Delta) = \begin{pmatrix} \cos \frac{\pi}{2}\Delta & \sin \frac{\pi}{2}\Delta \\ -\sin \frac{\pi}{2}\Delta & \cos \frac{\pi}{2}\Delta \end{pmatrix}.$$

Rotating all spins in layer α by the angle $\frac{\pi}{2}\alpha\Delta$, i.e. introducing new vectors $\boldsymbol{\sigma}_{i,\alpha} = \mathbf{R}(\alpha\Delta)\mathbf{S}_{i,\alpha}$, leads to the expression

$$Z = \sum_{\{\boldsymbol{\sigma}\}} \exp\left[-\frac{1}{2} \sum_{ij} \sum_{\alpha\beta} \sum_{\kappa=1}^{2} \sigma_{i,\alpha}^{\kappa} K_{i\alpha,j\beta} \sigma_{j,\beta}^{\kappa} \right] \tag{20}$$

for the partition function, κ labelling the two spin components. The elements $K_{i\alpha,j\beta}$ of the coupling matrix are zero unless the lattice sites (i, α) and (j, β) are nearest neighbours.

Expression (20) may be transformed [19, 20] into

$$Z = C \sum_{\{\boldsymbol{\sigma}\}} \left(\prod_{\rho=1}^{2} \prod_{k\gamma} \int \mathrm{d}h_{k,\gamma}^{\rho} \right) \exp\left[-\tfrac{1}{2} \sum_{ij} \sum_{\alpha\beta} \sum_{\kappa} h_{i,\alpha}^{\kappa} L_{i\alpha,j\beta}^{-1} h_{j,\beta}^{\kappa} + \sum_i \sum_\alpha \sum_\kappa h_{i,\alpha}^{\kappa} \sigma_{i,\alpha}^{\kappa} \right].$$

Here C is a numerical constant, N is the total number of lattice sites and $\mathbf{I}$ is the $N \times N$ identity matrix. The matrix $\mathbf{L}$ is given by $\mathbf{L} = \mu\mathbf{I} - \mathbf{K}$ where the positive number μ is chosen large enough to ensure that all the eigenvalues of $\mathbf{L}$ are positive.

The sum over all states can be easily computed:

$$\sum_{\{\boldsymbol{\sigma}\}} \exp\left(\sum_i \sum_\alpha \sum_\kappa h_{i,\alpha}^{\kappa} \sigma_{i,\alpha}^{\kappa} \right) = 2^N \prod_{i\alpha} [\cosh(h_{i,\alpha}^1 c_\alpha - h_{i,\alpha}^2 s_\alpha) + \cosh(h_{i,\alpha}^1 s_\alpha + h_{i,\alpha}^2 c_\alpha)] \tag{21}$$

with $c_\alpha = \cos(\frac{\pi}{2}\alpha\Delta)$ and $s_\alpha = \sin(\frac{\pi}{2}\alpha\Delta)$. Using the expansion

$$\ln\cosh x = -\sum_n (-1)^n \frac{2^{2n-1}(2^{2n}-1)B_n}{n(2n)!} x^{2n}$$

(B_n: Bernoulli number) the expression on the right-hand side of (21) can be written as the exponential of a sum of powers of $h_{i,\alpha}^1$ and $h_{i,\alpha}^2$:

$$C_1 \exp\left[\sum_n \sum_{k=0}^{2n} \sum_{l=0}^{k} \sum_{l'=0}^{2n-k} c(n,k,l,l') \sum_i \sum_\alpha c_\alpha^{l+l'} s_\alpha^{2n-l-l'} (h_{i,\alpha}^1)^k (h_{i,\alpha}^2)^{2n-k} \right] \tag{22}$$

where $c(n, k, l, l')$ is a number depending on n, k, l and l'.

Introducing new variables $\phi^{\kappa}_{i,\alpha} = L^{-1}_{i\alpha,j\beta} h^{\kappa}_{j,\beta}$, taking the continuum limit and turning to the wavenumber representation leads to the partition function

$$Z \propto \left(\prod_{\boldsymbol{q}} \int \mathrm{d}\boldsymbol{\tau}_{\boldsymbol{q}} \right) \exp[-\bar{H}]$$

with the effective Hamiltonian

$$\bar{H} = -\frac{1}{2} \int_{\mathrm{BZ}} \frac{\mathrm{d}^3 q}{(2\pi)^3} \left(r + q_{\perp}^2 + \frac{\Upsilon}{\Upsilon_0} q_{\parallel}^2 \right) [\boldsymbol{\tau}(\boldsymbol{q}) \boldsymbol{\tau}(-\boldsymbol{q})] - u \int_{\mathrm{BZ}} \frac{\mathrm{d}^3 q \, \mathrm{d}^3 q' \, \mathrm{d}^3 q''}{(2\pi)^9} [\boldsymbol{\tau}(\boldsymbol{q}) \boldsymbol{\tau}(\boldsymbol{q}')][\boldsymbol{\tau}(\boldsymbol{q}'') \boldsymbol{\tau}(-\boldsymbol{q} - \boldsymbol{q}' - \boldsymbol{q}'')] \tag{23}$$

with

$$\boldsymbol{\tau}_{\boldsymbol{q}} = (\tau^1_{\boldsymbol{q}}, \tau^2_{\boldsymbol{q}}) \qquad \text{and} \qquad \phi^{\kappa}(\boldsymbol{r}) = \int_{\mathrm{BZ}} \frac{\mathrm{d}^3 q}{(2\pi)^3} \exp(\mathrm{i}\boldsymbol{q} \cdot \boldsymbol{r}) \tau^{\kappa}_{\boldsymbol{q}}.$$

The integration is over the first Brillouin zone with $\boldsymbol{q} = (\boldsymbol{q}_{\perp}, \boldsymbol{q}_{\parallel})$, its components $\boldsymbol{q}_{\perp}$ and $\boldsymbol{q}_{\parallel}$ being perpendicular and parallel to the direction of the modulation respectively. $r = \frac{1}{\Upsilon_0}(1 - 2\Upsilon_0 - \Upsilon)$ with $\Upsilon = \frac{J}{k_B T}$ and $\Upsilon_0 = \frac{J_0}{k_B T}$ varies linearly with temperature.

In deriving equation (23) we neglected fourth and higher harmonics, i.e. fast oscillating terms containing $\exp(\mathrm{i}n\frac{\pi}{2}\alpha\Delta)$ with $n \geqslant 4$. Furthermore we did not include terms of higher than fourth order in τ. If we rescale $q_{\parallel}$ in the effective Hamiltonian [21] we end with the effective Ginzburg–Landau–Wilson Hamiltonian of the three-dimensional XY-model.

5. Conclusions

A low-temperature series expansion technique is suitable to obtain exact results on the low-temperature behaviour of the CC_4 model. All phases degenerate at the multiphase point ($T = 0, \Delta = \frac{1}{2}$) and obeying the structure combination rules spring from the multiphase point. Some of these phases disappear at higher temperatures. In the low-temperature limit the CC_4 model exhibits a complete devil's staircase. Differences in the low-temperature phase diagrams derived in the present and in a previous publication can be traced back to an inconsistency in the series expansion of the latter. Long-period spin patterns derived in this paper as stable phases between the ferromagnetic and the $\langle 4 \rangle$-phase and not occurring in the analyses presented in [2], were recently seen in Monte Carlo simulations just above the boundary of the ferromagnetic phase.

Furthermore, the critical behaviour at the boundary between the paraphase and the modulated structures follows from the derivation of an effective Ginzburg–Landau–Wilson Hamiltonian. It is shown that the latter can be transformed into the effective Hamiltonian of the three-dimensional XY-model. The four-state model thus belongs to the universality class of the XY-model.

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