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Critical behaviour of the two-dimensional biaxial next-nearest-neighbour Ising model: series expansions

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Abstract. The critical behaviour of an Ising model with competing first- and third-nearest-neighbour interactions ('biaxial next-nearest-neighbour Ising' or BNNNI model) on the square lattice is investigated by high- and low-temperature series.

1. Introduction

The study of Ising systems with further-neighbour interactions, particularly competing interactions, continues to be an active area of research (e.g. Selke 1984 and references therein). The full understanding of the occurrence and nature of spatially modulated phases, both commensurate and incommensurate with the lattice, in such simple models is a necessary first step to the understanding of such phenomena in real systems.

One model which has been extensively studied is the axial next-nearest-neighbour Ising (or ANNNI) model. This model does exhibit both commensurate and incommensurate modulated phases and an extremely rich phase diagram. In three dimensions the model has a finite-temperature Lifshitz point.

A related model, which has received far less attention, has competing nearest- and next-nearest-neighbour interactions along two of the lattice directions. We choose to call this the biaxial next-nearest-neighbour Ising model or BNNNI model. In two dimensions this is, of course, an isotropic version of the ANNNI model and there have been some previous studies of this case. The three-dimensional version, consisting of ferromagnetically coupled planes, is expected to exhibit a 'biaxial Lifshitz point' and is of interest for this reason. However in the present paper we concentrate on the two-dimensional case, leaving the three-dimensional version for future work.

The Hamiltonian of the model, which is illustrated in figure 1(a), is given by

$$\mathcal{H} = -J \sum_{\langle ij \rangle} s_i s_j - J' \sum_{[ij]} s_i s_j \tag{1}$$

where the first sum is over nearest-neighbour pairs and the second sum is over next-nearest-neighbour pairs in the axial directions (actually third neighbours). In this paper we consider only the zero-field case. Because the free energy is an even function of J we may, without loss of generality, take this interaction to be ferromagnetic ($J > 0$).

For $J' > -\frac{1}{2}J$ the ground state is ferromagnetic and the transition from the ordered phase to the disordered phase is expected to be of the universal 2D Ising type. At the point $J' = -\frac{1}{2}J$ the ground state is infinitely degenerate, with a non-zero entropy, and consequently the critical temperature will drop to zero at this point. For $J' < -\frac{1}{2}J$ the antiferromagnetic interactions are sufficiently strong to stabilise two types of ground state, the 'chessboard' and 'staircase' configurations, shown in figure 1(b). Both of these structures can be regarded as commensurate modulated structures with wavevector in the diagonal direction $q = (\frac{1}{4}, \frac{1}{4})$. These two states have the same energy but it can be argued that at low but non-zero temperature a 'chessboard-like' structure will be favoured because of higher entropy and hence lower free energy (Selke and Fisher 1980). The nature of the transition, or sequence of transitions, from this ordered phase to the high-temperature disordered phase is uncertain. Early Monte Carlo work (Hornreich *et al* 1979, Selke and Fisher 1980) indicated a transition from the commensurate phase to an incommensurate phase followed by a second transition to the disordered phase, this transition presumably being of Kosterlitz–Thouless type. However, more recent Monte Carlo studies by Landau and Binder (1985) show a single first-order transition directly from the commensurate ordered phase to the disordered phase, without the presence of an intermediate phase. The two possible forms of the phase diagram are shown in figure 2(a).

An alternative way of exhibiting the phase diagram of the model is via the lines of singularities of the free energy per spin $f(K, K')$ where $K = J/kT$ and $K' = J'/kT$. Since for $K' = 0$, or for $K = 0$, the model reduces to the nearest-neighbour square lattice problem there will be singularities of the Onsager type at the four symmetrically located points $(\pm K_0, 0)$, $(0, \pm K_0)$ with $K_0 = 0.440\,68\ldots$, as shown in figure 2(b). There will be a pair of critical lines passing through these points and asymptotically approaching the lines $K' = \pm\frac{1}{2}K$. There will be a branch, or possibly two branches, in the lower half-plane, as shown.

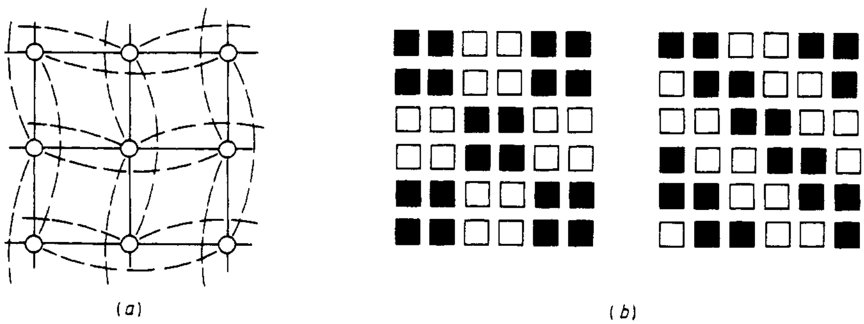

Figure 1. (a) The interactions of the two-dimensional BNNNI model. (b) The two types of ground state for $J' < -\frac{1}{2}J$.

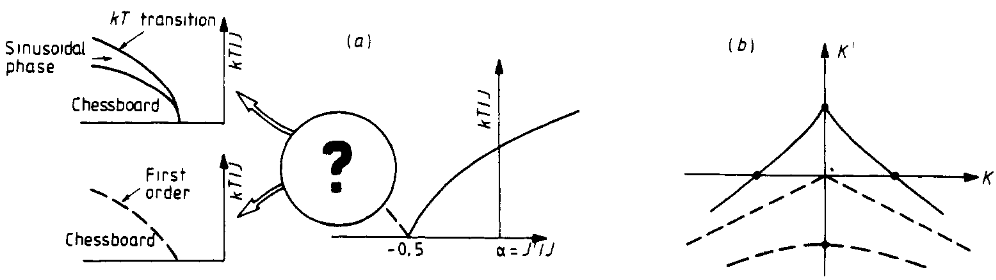

Figure 2. (a) The expected variation of transition temperature with $\alpha = J'/J$, showing the two possible pictures for $\alpha < -0.5$. (b) The lines of singularities of the free energy $f(K, K')$. The lower branch may be a single line of first-order transitions or two second-order lines.

In the present paper we investigate the properties of this system by the technique of exact series expansions. We first derive and analyse high-temperature series for the wavevector-dependent susceptibility $\chi(\boldsymbol{q})$. This leads to an accurate determination of the critical temperature over most of the ferromagnetic region. Over most of the modulated region, $K' < -\frac{1}{2}K$, we are able to locate a consistent singularity in a $\chi(\boldsymbol{q})$ with non-zero wavenumber and there is some evidence that the singularity is of the Kosterlitz–Thouless form. This then tends to support the picture of two transitions rather than a single first-order transition. To test this question further we derive both high- and low-temperature series for the free energies. By plotting both the high- and low-temperature free energies it is possible, in principle, to distinguish between first- and second-order transitions. In the present case we find no strong evidence for a first-order transition, although we cannot completely exclude it.

In the following paper (Oitmaa *et al* 1987) we study the same model using a completely different approach, based on transfer matrix calculations for finite width strips and a finite-size scaling analysis.

2. The susceptibility series

The high-temperature series for the wavevector-dependent susceptibility was derived using the connected multigraph expansion (Oitmaa 1981). The susceptibility is expressed in the general form

$$\chi(\boldsymbol{q}) = 1 + 2 \sum_{\{G\}} W_G X_G(\boldsymbol{q}, \{v_\alpha\}) \tag{2}$$

where the sum is over the set of connected graphs with single and multiple edges which have exactly two vertices of odd degree. Through eleventh order there are 3296 such graphs, and the graph weights W_G, which are the same for any Ising problem, are available from previous work. The factor X_G is a sum over all embeddings of G on the lattice, in which, for each embedding, two factors are included:

(i) a contribution from each edge of $\tanh \beta J_\alpha$, where J_α is the particular interaction parameter, and

(ii) a factor $\exp(2\pi i \boldsymbol{q} \cdot \boldsymbol{R})$ where $\boldsymbol{R}$ is the vector joining the two odd vertices.

In this way we obtain a two-variable series of the form

$$\chi(\boldsymbol{q}) = 1 + \sum_{n=1}^{\infty} \sum_{s=0}^{n} C_{ns}(\boldsymbol{q}) v^{n-s} w^s \tag{3}$$

where $v = \tanh K$, $w = \tanh K'$. The data are too extensive to publish but can be supplied on request. For $\boldsymbol{q} = 0$ the coefficients are given in table 1.

The analysis proceeds in the usual way. For any chosen value of $\alpha = J'/J$ we expand the hyperbolic tangent factors and obtain a series in the single variable $K = J/kT$. The resulting single variable series are then analysed by standard ratio and Padé approximant methods.

In the ferromagnetic region $\alpha > -0.5$ the $\boldsymbol{q} = 0$ susceptibility is expected to diverge at a critical point $K_c(\alpha)$ with a power law

$$\chi \sim C\left(1 - \frac{K}{K_c}\right)^{-\gamma} \tag{4}$$

with the exponent taking the universal value $\gamma = \frac{7}{4}$. For $\alpha > 0$ (all interactions ferromagnetic) the position of the singularity can be estimated with an uncertainty of less than

Table 1. Coefficients of the high-temperature susceptibility (3) for $q = 0$.

n	s = 0	1	2	3	4	5	6	7	8	9	10	11
1	4	4										
2	12	32	12									
3	36	148	160	36								
4	100	592	1 064	672	100							
5	276	2 040	5 520	5 900	2 528	276						
6	740	6 672	23 292	38 352	28 064	8 864	740					
7	1 972	20 688	89 520	194 928	221 920	120 460	29 536	1 972				
8	5 172	62 384	318 048	873 104	1 334 472	1 132 624	480 088	94 752	5 172			
9	13 492	182 792	1 078 192	3 530 912	6 896 448	7 927 824	5 271 792	1 808 004	295 008	13 492		
10	34 876	526 256	3 505 912	13 387 856	31 540 608	46 781 392	42 363 708	22 857 424	6 511 600	896 672	34 876	
11	89 764	1488 168	11 079 056	48 078 760	133 136 688	240 314 536	282 662 832	208 469 864	93 666 464	22 617 748	2671 904	89 764

1% from ratios and from Padé approximants to the logarithmic derivative series. Padé approximants to $\chi^{4/7}$ yield four figure accuracy. In the region $-0.5 < \alpha < 0$ the analysis is complicated by the presence of competing singularities closer to the origin than the physical singularity, as shown in figure 3. In such situations it is often possible to improve the analysis by using transformations to move the physical singularity closer to the origin. An Euler transformation of the form

$$K'' = \frac{K}{1 + aK}$$

has the effect of moving the antiferromagnetic singularity on the negative real axis further away, and gives much improved consistency in the Padé approximants. We have also tried a variety of transformations which expand the imaginary axis and contract the real axis, in conjunction with Euler transformations, but no significant improvement is obtained. In table 2 we present some results of our analysis for the case $\alpha = -0.3$. As α approaches -0.5 and the critical coupling K_c approaches infinity, the analysis becomes increasingly difficult and cannot be carried beyond about -0.35 or -0.4.

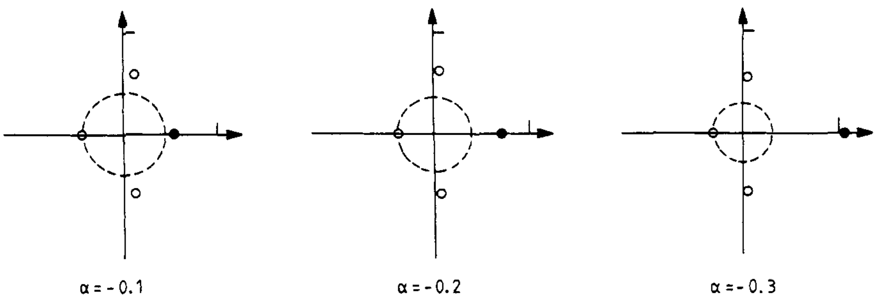

Figure 3. Singularities in the complex K plane for the ferromagnetic susceptibility. The full circle is the ferromagnetic critical point, which lies further and further outside the disc of convergence of the series as $\alpha \to -0.5$.

In the 'modulated' region $\alpha < -0.5$ it is necessary to carry out a systematic analysis of $\chi(\boldsymbol{q})$ series for the entire range of $\boldsymbol{q}$ values. (Throughout this paper, we restrict $\boldsymbol{q}$ to be in the diagonal direction, i.e. $\boldsymbol{q} = q(1, 1)$.) If there is a transition from the disordered phase to an incommensurate modulated phase then we would expect to find a critical wavenumber $q_c(\alpha)$ for which $\chi(\boldsymbol{q})$ shows a consistent singularity on the positive real axis. Such behaviour is seen in the two-dimensional ANNNI model (Oitmaa 1985). Since in this case we are seeking the location of the lower transition line (see figure 2(b)), which passes through the point $(0, -K_0)$, it is more convenient to write $K = \beta K'$ $(\beta = \alpha^{-1})$ and to obtain single variable series in $K' = J'/kT$. We then look for singularities on the negative real axis of the complex K' plane. There will, in all cases, be interference from a closer singularity on the positive real axis, which corresponds to the upper branch in figure 2(b). An Euler transformation is used to expand the positive axis and contract the negative axis to bring the physical singularity closest to the origin.

For $\beta = 0$ the series for $q = 0.25$ is the nearest-neighbour Ising staggered susceptibility and shows a power law singularity of the form (4) with $K'_c = --0.4407$ and $\gamma = 1.75$. As β becomes negative the series for $q = 0.25$ no longer show a consistent pole on the negative real axis but rather a complex conjugate pair. However it is possible to find

Table 2. Analysis of susceptibility series for the case $\alpha = -0.3$. (*a*) Coefficients of the series $\chi(K)$ and of the series $\chi(K'')$ with $K'' = K/(1+2.5\,K)$, (*b*) poles of Padé approximants to logarithmic derivative series, (*c*) poles of Padé approximants to $\chi^{4/7}$.

(*a*)	K series	K'' series
	1.0	1.0
	2.8	2.8
	3.48	10.48
	3.730 666 667 ...	38.630 666 67 ...
	−3.750 8	133.229 2
	11.193 637 33 ...	440.460 637 3 ...
	−3.762 563 2	1 437.774 570 ...
	55.106 632 78 ...	4 784.540 435 ...
	−131.045 013 6 ...	16 639.989 43 ...
	264.175 356 0 ...	60 857.360 21 ...
	−716.964 097 5 ...	230 808.285 3 ...
	2187.751 839 ...	887 100.474 7 ...

(*b*)	K series	K'' series
[4, 6]	−0.3205,	
	0.0291 ± 0.5761i	0.2669 ± 0.1786i
[5, 5]	1.5998, −0.3206,	0.2422 ± 0.0429i
	0.0304 ± 0.5781i	0.2554 ± 0.1985i
[6, 4]	−0.3205,	0.2849 ± 0.0483i
	0.0277 ± 0.5745i	0.2616 ± 0.1852i
[4, 5]	1.3549, −0.3204,	0.2424
	0.0354 ± 0.5748i	0.2461 ± 0.2086i
[5, 4]	0.9930, −0.3203,	0.2473
	0.0421 ± 0.5753i	0.2408 ± 0.2080i

(*c*)	[4, 7]	[5, 6]	[6, 5]	[7, 4]	[4, 6]	[5, 5]	[6, 4]
K series	1.0645	1.0453	1.0801	—	1.0262	1.0133	—
K'' series	0.2889	0.2916	0.2864	0.2898	0.2871	0.2868	0.2868
Estimate			$K''_c = 0.288 \pm 0.002$ gives $K_c = 1.03 \pm 0.03$				

a wavenumber for which $\chi(\boldsymbol{q})$ does show a consistent singularity on the axis. In table 3 we present a summary of the analysis for $\beta = -1.0$, a wavenumber $q = 0.22$ giving, in this case, the most consistent singularity. If this represents a transition to an incommensurate phase then there are arguments for expecting the asymptotic behaviour to be of the Kosterlitz-Thouless form $\exp[c(1-K/K_c)^{-\gamma}]$. Guttmann (1978) has suggested that, to distinguish between an essential singularity of this form and a conventional power law singularity of the form (4), one should look at the second logarithmic derivative. An algebraic singularity in the original series would give a simple pole with residue 1 whereas an essential singularity of the Kosterlitz-Thouless form would give a simple pole with residue $1+\gamma$. In the present case, for the $\beta = -1.0$ series, the residue is around 1.4–1.5, which is consistent with a Kosterlitz-Thouless transition, with $\gamma = \frac{1}{2}$.

The results of our analysis are summarised in figures 4 and 5. On the ferromagnetic side the locus of the critical temperature agrees very well with the Monte Carlo (MC) results of Landau and Binder (1985). However, on the modulated side our critical line deviates significantly from the MC first-order transition temperature. Moreover

Table 3. Analysis of susceptibility series for $\beta = -1.0$ with wavenumber $q = 0.22$. (a) Coefficients of series $\chi(K')$ and of the Euler transformed series $\chi(K'')$, with $K'' = K'/(1-5K')$, (b) position of pole on negative real axis and residue from Padé approximants to logarithmic derivative series, (c) position of pole on negative real axis and residue from Padé approximants to second logarithmic derivative series.

(a)	K' series	K'' series
	1.0	1.0
	−4.468 631 201 90 . . .	−4.468 631 201 90 . . .
	11.968 664 818 6 . . .	34.311 820 828 0 . . .
	−32.713 586 906 0 . . .	−264.116 015 139 . . .
	56.268 674 673 6 . . .	2 003.201 239 89 . . .
	−225.185 749 117 . . .	−15 034.824 189 0 . . .
	−2.186 304 493 91 . . .	111 953.159 788 . . .
	−2 375.518 431 62 . . .	−828 350.977 021 . . .
	−6 914.354 079 35 . . .	6 096 159.559 92 . . .
	−44 681.717 671 7 . . .	−44 652 729.763 1 . . .
	−201 308.393 489 . . .	325 688 842.249 . . .
	−1042 828.304 51 . . .	−2366 449 154.69 . . .

(b)	K' series	K'' series
[4, 6]	−0.5463 (2.025)	−0.1464 (2.025)
[5, 5]	−0.5463 (2.025)	−0.1464 (2.025)
[6, 4]	−0.4405 (0.459)	−0.1464 (2.024)
[4, 5]	−0.5475 (2.046)	−0.1466 (2.066)
[5, 4]	−0.6096 (3.474)	−0.1466 (2.081)
[4, 4]	−0.6660 (5.832)	−0.1449 (1.766)
Estimate	$K''_c = -0.146 \pm 0.001$	
	$K'_c = -0.541 \pm 0.014$	

(c)	K' series	K'' series
[3, 6]	−0.4684 (0.566)	−0.1508 (1.438)
[4, 5]	−0.4475 (0.458)	−0.1503 (1.394)
[5, 4]	−0.2313 (0.006)	−0.1501 (1.374)
[6, 3]	−0.2734 (0.022)	−0.1503 (1.392)
[3, 5]	−0.5089 (0.751)	−0.1511 (1.468)
[4, 4]	−1.1198 (9.561)	−0.1570 (2.318)
[5, 3]	—	−0.1511 (1.460)

our transition temperatures are higher than the MC values so that the discrepancy is not attributable to the series seeing a spinodal curve within the region of metastability. One possible explanation is that there are indeed two transitions and that the Monte Carlo work is picking up the lower one whereas the series are seeing the higher transition. This point is investigated further in the following section.

3. The free energy series

It is difficult, using series techniques, to unambiguously identify a first-order transition. The only practical approach is the technique of 'free energy matching', which uses

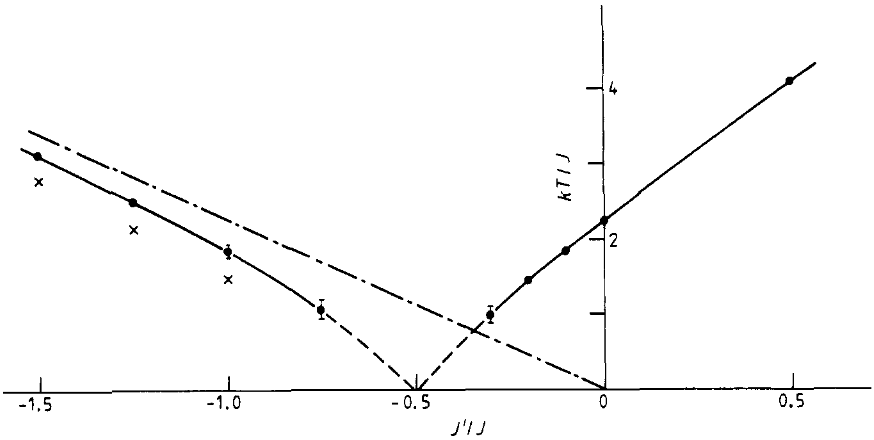

Figure 4. Variation of transition temperature with $\alpha = J'/J$, as determined from high-temperature series. The crosses are Monte Carlo estimates from Landau and Binder (1985). The chain curve is the asymptote corresponding to $J \to 0$, $kT/J' = -2.27$.

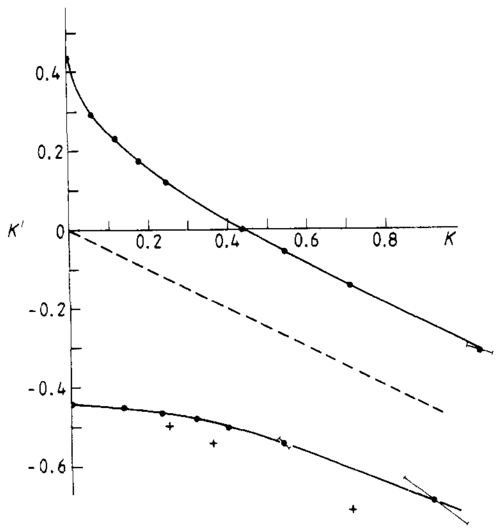

Figure 5. The singularities of $f(K, K')$ as determined from high-temperature series. The crosses are the Monte Carlo estimates for Landau and Binder (1985).

both high- and low-temperature expansions to evaluate the free energy as a function of temperature. If the two branches thus obtained meet smoothly, with no change in slope, then the transition is of second (or higher) order. If, however, there is a change in slope then the point of intersection is a first-order transition point. There are, of course, difficulties with this procedure since there is always numerical uncertainty in the values of the free energy thus obtained. However, the method has been successfully used in several recent studies (Velgakis and Ferer 1983, Styer 1985). We have therefore attempted to resolve the question of the nature of the transition to the modulated phase by this method.

A high-temperature expansion for the zero-field energy has been obtained by the same technique as used in the previous section for the susceptibility. The expansion has been computed through twelfth order, to which order there are 508 graphs. The expansion has the form

$$-\beta f = \ln 2 + 2 \ln \cosh K + 2 \ln \cosh K' + \sum_{n=1}^{\infty} \sum_{s=0}^{n} a_{ns} v^{n-s} w^{s} \qquad (5)$$

where $v = \tanh K$, $w = \tanh K'$ as before. The coefficients $\{a_{ns}\}$ are given in table 4. For any value of $\alpha = K'/K$ we then obtain a series in the single variable $K = J/kT$. By computing Padé approximants to this series and evaluating these at a sequence of K values we obtain an estimate of the high-temperature 'branch' of the free energy.

Table 4. Coefficients of the high-temperature free energy expansion (5).

	s												
n	0	1	2	3	4	5	6	7	8	9	10	11	12
3	0	2	0	0									
4	1	0	4	0	1								
5	0	12	0	14	0	0							
6	2	0	73	0	36	0	2						
7	0	32	0	356	0	94	0	0					
8	$4\frac{1}{2}$	0	316	0	1 540	0	236	0	$4\frac{1}{2}$				
9	0	92	0	$2\,450\frac{2}{3}$	0	6 180	0	618	0	0			
0	12	0	1178	0	15 964	0	23 353	0	1 556	0	12		
11	0	316	0	11 892	0	91 236	0	84 028	0	3978	0	0	
12	$37\frac{1}{3}$	0	4988	0	$101\,333\frac{1}{2}$	0	$472\,073\frac{1}{8}$	0	291 106	0	9956	0	$37\frac{1}{3}$

A low-temperature expansion for the free energy, valid in the modulated region, has been obtained by starting from the ordered chessboard state and enumerating configurations with a small number of overturned spins. Rather than use the partial generating function method (Sykes *et al* 1965), which becomes very complicated in the present case because of the large number of sublattices, we have used a more primitive approach. The configurations have been enumerated by hand and a computer program written to evaluate the lattice embedding constants. The series is given by

$$\begin{aligned}-\beta f = {} & 2|K'| + v^2 + 2v^3 + (u + 2\tfrac{1}{2} + u^{-1})v^4 + 6(u + u^{-1})v^5 \\ & + (2u^2 + 22u - 10\tfrac{2}{3} + 22u^{-1} + 2u^{-2})v^6 \\ & + (20u^2 + 62u - 34 + 62u^{-1} + 20u^{-2})v^7 + \dots \end{aligned} \qquad (6)$$

where $v = \exp(-4|K'|)$, $u = \exp(-4K)$. Padé approximants are then used to evaluate the low-temperature free energy.

Results are presented for the case $\alpha = -1$, which is typical. In table 5 we give numerical estimates for the high- and low-temperature free energies, obtained from the highest-order Padé approximants. These results are used as the basis of the plot shown in figure 6. As can be seen from this figure there is no apparent crossing of the two branches, and hence no indication of a first-order transition. At the positions of the transition obtained in the previous section ($K \simeq 0.54$) and that obtained from the diagram of Landau and Binder ($K = 0.71$) the high-temperature free energy lies clearly below the low-temperature curve. It may be that our error estimates for the high-temperature free energy for $K \geqslant 0.6$ are too optimistic. Indeed, if one were to disregard

Table 5. Estimates of $-\beta f$ from (a) high- and (b) low-temperature expansions.

(a)	K						
$[N, D]$	0.45	0.50	0.55	0.60	0.65	0.70	0.75
[5, 7]	1.007	1.075	1.150	1.232	1.321	1.417	1.519
[6, 6]	1.007	1.076	1.151	1.234	1.326	1.426	1.536
[7, 5]	1.006	1.074	1.148	1.229	1.316	1.409	1.509
[5, 6]	1.008	1.078	1.155	1.242	1.334	1.447	1.570
[6, 5]	1.007	1.075	1.149	1.230	1.318	1.413	1.515
[4, 6]	1.007	1.076	1.151	1.234	1.326	1.426	1.536
[5, 5]	1.006	1.074	1.148	1.229	1.316	1.409	1.509
[6, 4]	1.007	1.076	1.151	1.234	1.326	1.426	1.535
Estimate	1.007	1.076	1.151	1.234	1.325	1.425	1.53
	±0.001	±0.002	±0.003	±0.004	±0.01	±0.015	±0.02

(b)	K						
$[N, D]$	0.45	0.50	0.55	0.60	0.65	0.70	0.75
[2, 5]	0.994	1.036	1.120	1.2117	1.3072	1.4046	1.5029
[3, 4]	1.011	1.037	1.120	1.2117	1.3072	1.4046	1.5029
[4, 3]	1.004	1.037	1.120	1.2117	1.3072	1.4046	1.5029
[5, 2]	0.988	1.036	1.120	1.2117	1.3072	1.4046	1.5029
[2, 4]	0.949	1.032	1.119	1.2116	1.3072	1.4046	1.5029
[3, 3]	0.897	1.066	1.122	1.2122	1.3074	1.4046	1.5029
[4, 2]	0.960	1.033	1.119	1.2116	1.3072	1.4046	1.5029
Estimate	0.99	1.037	1.120	1.2117	1.3072	1.4046	1.5209
	±0.03	±0.003	±0.0005				

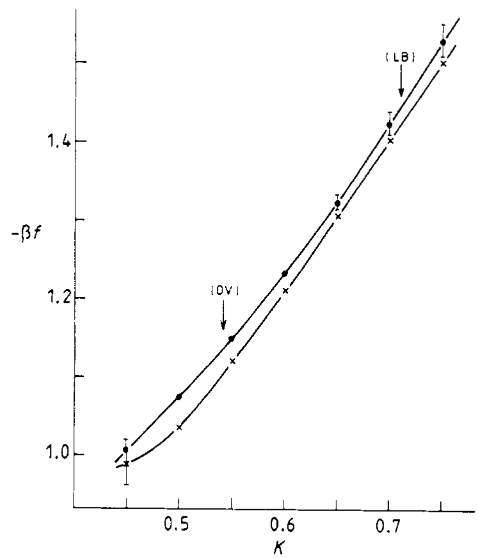

Figure 6. High- and low-temperature free energies for the case $\alpha = -1$. The arrows show the transition points obtained in this paper (OV) and by Landau and Binder (LB).

these points and extrapolate the high T curve from the clearly convergent values for $K \leqslant 0.5$, then it is conceivable that an intersection of the two curves, with a change in slope, might occur around the Landau–Binder esimate. However, it is also the case that if there exists an intermediate phase then one would not expect the high- and low-temperature series to converge to a single point of intersection and one might see the type of behaviour evident in figure 6.

4. Conclusions

The model we have studied in this paper is a very simple one, and yet has sufficiently subtle and rich behaviour as to remain only partially understood. In the regime where the third-neighbour interactions are ferromagnetic or weakly antiferromagnetic, the picture is clear and our results corroborate previous ones. When the third-neighbour interactions are sufficiently strongly antiferromagnetic, so that the ground state is the chessboard or staircase structure, our results are rather inconclusive. The series do show a transition, with some indication that it may be of Kosterlitz–Thouless form, at a temperature above the transition shown by the recent Monte Carlo work. This, and the fact that the free energy matching procedure shows no sign of a first-order transition, may be taken as favouring the picture of two separate transitions. However, the evidence is not clear cut and further studies of this model are clearly warranted.

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