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Phase equilibria and the scaling transformation

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Abstract. It is argued that the scaling transformation which is currently being used to obtain numerical estimates of critical exponents *cannot* safely be employed without prior knowledge that the points in question are second-order transition points. The transformation when defined on finite or semi-infinite systems will not distinguish between order–disorder critical points and any other point on the coexistence surface. This in turn means that the full phase diagram of the equilibrium surface can be found using the scaling transformation. The three-state Potts model is used to illustrate this.

Introduction

It has been demonstrated by several workers (Nightingale 1976, 1977, Sneddon 1978, Nightingale and Blöte 1980, Blöte *et al* 1981, Roomany *et al* 1980, Wood and Goldfinch 1980, and Goldfinch and Wood 1982) that the scaling transformation of Kadanoff (Kadanoff *et al* 1967) can be adapted to yield very accurate numerical estimates of any critical point parameter. Nightingale showed how the relative 'thickness' of semi-infinite systems could be intepreted as the rescaling factor; the calculations are very modest in scale and remain so even in quite complicated interaction spaces where even the lowest order calculations give surprisingly good results. In the calculations of Wood and Goldfinch (1980) and Goldfinch and Wood (1982) the method was applied to some two-dimensional models of lattice gases including the square well and hard square models. In each case the accuracy with which the correlation length exponent ν was obtained probably could not have been matched by any other method, and the results appear to show that both these models have Ising-like second-order transitions.

The purpose of the present paper is to show that this technique can be greatly extended into a method of obtaining the *whole phase equilibrium surface*. The applications of the method to date have mainly been restricted to second-order transitions, critical points, and exponents, although the scaling transformation has also been applied at known first-order transitions in the 2D zero-field Potts model by Blöte *et al* (1981), and Roomany and Wyld (1981). The claim of the present authors is that the method generally does *not* select only the critical points in the phase diagram, but all the points on the coexistence surface as well. Strictly the method cannot be safely applied to a critical point without prior knowledge that the point in question is a second-order transition since these points will only appear as end points or boundary surfaces to regions where first-order transitions can occur. An example of this is to be found in a recent publication by Roomany and Wyld (1981). These authors have used a finite lattice method in a Hamiltonian formalism to study the zero field q-state Potts model in $(1+1)$ dimensions, where a rescaling of the mass gap of the Hamiltonian is used to identify a phase transition and is very similar to the rescaling of the coherence length used in the present scaling transformation. Roomany and Wyld reported no sign of a first-order transition for $q \geqslant 5$ even though they obtained fixed points for all values of q which they examined; these they seem to have taken as false predictions of a second-order transition. The present work would imply that the results obtained by these authors are both correct and to be expected. Their finite system method should give positive results for both types of transition and fail to distinguish between them.

In § 2 we explain why in our view the scaling transformation should yield a convergent sequence of surfaces to the full phase equilibrium diagram. The ferromagnetic and antiferromagnetic models of the 2D Ising model are used to provide a simple illustration of this effect. In § 3 this claim is subjected to a more stringent test with an application to the 2D three-state Potts model in a field, which has been conjectured to have a fairly complicated phase equilibrium structure (for a review see Wu (1982)). We have found that all features of the coexistence surface are clearly evidenced even in a low order of the scaling transformation. Finally, aspects of the phase equilibrium surface are compared between the three-, four- and five-state Potts models.

2. Coexistence and long-range order

Consider a lattice model of N sites labelled i, which can assume any one of k states indexed by σ_i, and let the actual values taken by σ_i be $\alpha_1, \alpha_2, \ldots, \alpha_k$. Thus

$$n_p = \sum_i \delta_{\sigma_i, \alpha_p} \tag{1}$$

is the number of sites of species p in any configuration of the system, and $\langle n_p \rangle / N = \langle \delta_{\sigma_i, \alpha_p} \rangle$ is the mean fractional number of sites of species p. Thus in the Ising model $\alpha_1 = 1$, and $\alpha_2 = -1$, and the magnetisation is given by

$$m(T, h) = \langle n_1 \rangle - \langle n_2 \rangle, \tag{2}$$

where h is the ordering field of species 1 (and $-h$ the ordering field of species 2) by which we mean that $h > 0$ favours the occupancy of sites by species 1, giving rise to a 1-rich phase in which $\langle \delta_{\sigma_i, 1} \rangle > \frac{1}{2}$.

In a statistical mechanical treatment of the occupancies $\langle \delta_{\sigma_i, \alpha_p} \rangle$ it is necessary to specify the preparation of the sample, both with respect to the ordering fields present (h_l, $l = 1, 2, \ldots, k$) and the growth of the sample in the limit $N \to \infty$. The preparation with respect to the fields h_l clearly has a laboratory equivalent, but the thermodynamic limit does not. Thus in the Ising model case

$$m(T, h) = \lim_{N \to \infty} \lim_{h \to 0} [\langle \delta_{\sigma_i, 1} \rangle - \langle \delta_{\sigma_i, -1} \rangle] = 0 \tag{3}$$

at all temperatures, and

$$m^{\pm}(T, h) = \lim_{h \to 0^{\pm}} \lim_{N \to \infty} [\langle \delta_{\sigma_i, 1} \rangle - \langle \delta_{\sigma_i, -1} \rangle] \neq 0, \qquad T < T_c \tag{4}$$

where $m^{\pm}$ are the coexisting zero-field magnetisations. The second preparation has been done inside the 1-rich $(+)$ or 2-rich $(-)$ region of the phase diagram and since

$\pm h$ acts to suppress one or other phase in the growth $N \to \infty$, the sample has been forced into assuming just one of its two equilibrium states when an approach to a point on the coexistence surface Σ is made in this way. In (3) the sample has been prepared at a point on Σ and allowed to grow while being maintained at this point ($T < T_c, h = 0$); neither the 1-rich or 2-rich phase has at any stage been suppressed in the growth $N \to \infty$. In effect the sample has been prepared inside the two-phase region akin to a liquid in equilibrium with its vapour. Whereas in the laboratory we can force the assembly to adopt a mixed state in which the density lies between the coexisting vapour and liquid densities, we cannot do this theoretically since the statistical mechanics will assign equal proportions to each phase giving a density which is the average of the two coexisting states ($=\frac{1}{2}$ in the Ising model).

Similar considerations apply to assemblies with three or more species, differences between preparations 'on' or 'on an approach to' Σ can be made to appear in the thermodynamic limit. Thus with three species (see § 3) and ordering fields h_1, h_2, and h_3 the phase diagram will have j-rich regions, and somewhere in the space (T, h) we would expect to see a coexistence surface between pairs of species. If the assembly is prepared *at* a point on Σ bordering say the i-rich and j-rich regions, then (assuming no asymmetry in the preference for either species)

$$\langle \delta_{\sigma_i,\alpha_i} \rangle = \langle \delta_{\sigma_i,\alpha_j} \rangle \tag{5}$$

at coexistence, thus anywhere on that part of Σ marking coexisting phases i and j we would, theoretically, expect to see the result

$$\lim_{N\to\infty} \lim_{(T,h)\to\Sigma} [\langle \delta_{\sigma_i,\alpha_i} \rangle - \langle \delta_{\sigma_i,\alpha_j} \rangle] = 0. \tag{6}$$

If, however, the i-rich ordering field acts simultaneously to suppress the j phase then approaching Σ from within the i-rich region

$$\lim_{(T,h)\to\Sigma} \lim_{N\to\infty} [\langle \delta_{\sigma_i,\alpha_i} \rangle - \langle \delta_{\sigma_i,\alpha_j} \rangle] > 0. \tag{7}$$

On an approach to the boundary of Σ via the disordered phase (i.e. a critical point) two or more phases are nearly stable, thus droplets of each phase can grow into sizes where their free energy density $\sim kT$. In the statistical mechanics these configurations are the dominant terms in the total free energy, the so-called coherence length ξ is envisaged to be the mean size of such droplets measured over all configurations. At the critical point, each phase can grow indefinitely and in the thermodynamic limit ξ becomes infinitely large. This divergence is often referred to as infinitely divergent flctuations in the neighbourhood of a critical point, and is sometimes used to distinguish a second-order transition from a first-order transition, which is not associated with long-range fluctuations. The reason for this is again in the preparation; if the preparation follows (7) then only the large i-rich droplets can survive in the large-system limit (see Domb 1976) as Σ is approached and long-range fluctuations in $\delta_{\sigma_i,\alpha_i}$, and $\delta_{\sigma_i,\alpha_j}$ cannot be sustained. If, however, the preparation follows (6) then droplets of competing phases could grow equally and long-range fluctuations are possible. Thus so long as we remain on Σ and none of the competing species have been suppressed, then statistical mechanically, ξ should diverge. Hence everywhere on Σ the coherence length is divergent in this sense, and in relation to scaling theory, when assemblies are allowed to grow on their Σ-surfaces the coherence length should be invariant to a rescaling of length in the thermodynamic limit. This means that the basic equation which is used in the current applications of the scaling transformation (Nightingale 1976)

$$\xi' = \xi/L, \tag{8}$$

where all lengths in the prime system have been rescaled by a factor of L, should act as an approximation to the whole Σ-surface and *not* just the critical points.

The length ξ is commonly measured in terms of the characteristic decay length in the correlation of fluctuations, thus in terms of fluctuations (Kadanoff *et al* 1967) in the population distribution $\Gamma_{pp'}(\boldsymbol{R})$

$$\Gamma_{pp'}(\boldsymbol{R}) = \langle(\delta_{\sigma,\alpha_p}(0) - \langle\delta_{\sigma,\alpha_p}\rangle)(\delta_{\sigma,\alpha_{p'}}(\boldsymbol{R}) - \langle\delta_{\sigma,\alpha_{p'}}\rangle)\rangle \tag{9}$$

and is a measure of population fluctuations in species p and p' at distances R apart. If the regions which support a coherent fluctuation become infinitely large then

$$\lim_{\boldsymbol{R}\to\infty} |\Gamma_{pp'}(\boldsymbol{R})| > 0. \tag{10}$$

The characteristic decay length of the correlations is not thought to be dependent upon p and p' and is defined as

$$\xi^{-1} = -\lim_{\boldsymbol{R}\to\infty} (1/R) \ln|\Gamma_{pp'}(\boldsymbol{R})| \tag{11}$$

and our claim is that $\xi^{-1} = 0$ everywhere on Σ if the system is prepared in the manner of (6).

For classical lattice models, all of these correlation functions can be formulated in terms of the spectral properties of a real symmetric transfer matrix $\boldsymbol{T}$ (see Fisher and Burford 1967, Wood 1975 and Thompson 1972). In present applications of the scaling transformation we are concerned with $m \times \infty$ sections of the square lattice, where $\boldsymbol{T}$ can be defined in general terms with elements

$$T_{aa'} = \exp[-\beta(U(a)/2 + U(a')/2 + W(a, a'))] \tag{12}$$

where a and a' index the configurations of neighbouring m-site columns, $W(a, a')$ is the interaction energy between neighbouring columns, and $U(a)$ and $U(a')$ are the

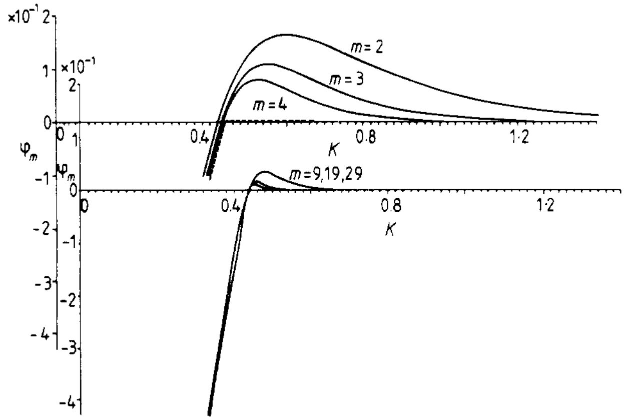

Figure 1. Plots of $\varphi_m(K, 0)$ obtained from the Onsager solution of $m \times \infty$ sections of the square lattice for $m = 2, 3, 4$, and 9, 19, 29. The intersections on the K axis are the scaling transformation approximations to the critical ponint ($K_c = 0.440687$).

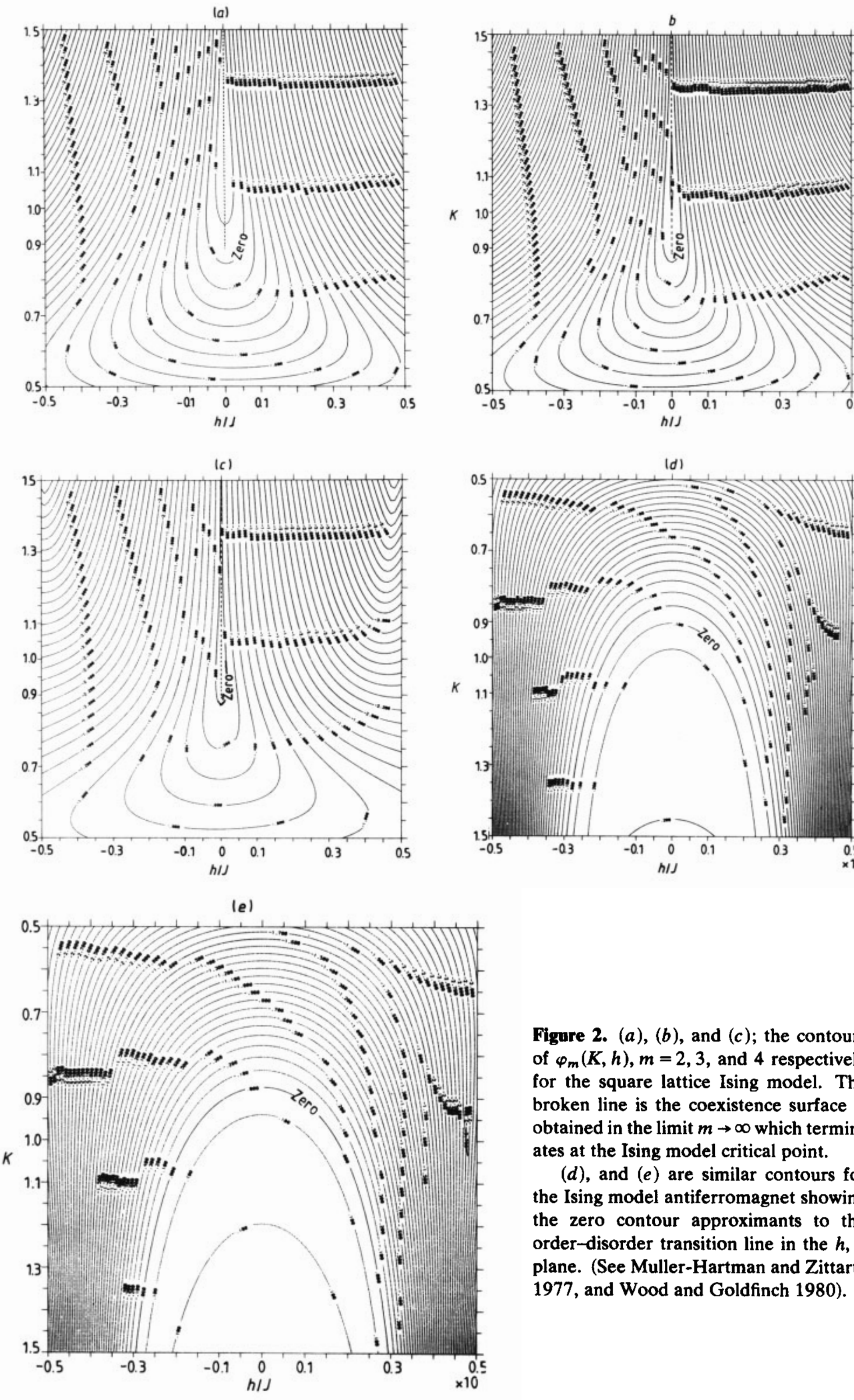

Figure 2. (a), (b), and (c); the contours of $\varphi_m(K, h)$, $m = 2, 3$, and 4 respectively for the square lattice Ising model. The broken line is the coexistence surface Σ obtained in the limit $m \to \infty$ which terminates at the Ising model critical point.

(d), and (e) are similar contours for the Ising model antiferromagnet showing the zero contour approximants to the order–disorder transition line in the h, T plane. (See Muller-Hartman and Zittartz 1977, and Wood and Goldfinch 1980).

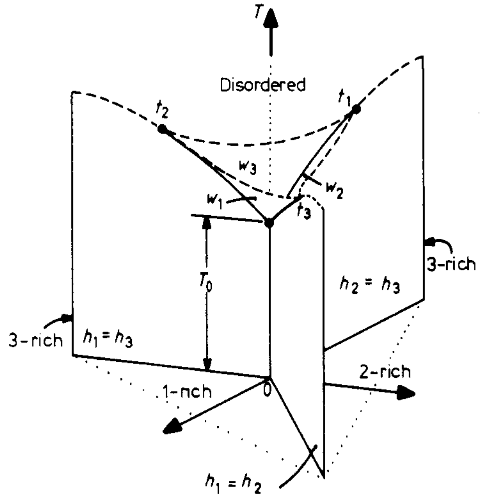

Figure 3. The form of the phase equilibrium diagram of the three-state Potts model as conjectured by Straley and Fisher (1973). The three coexistence planes $h_i = h_j$ define the coexisting states between the rich phases, and the three web extensions mark off coexistence between the rich and disordered phases.

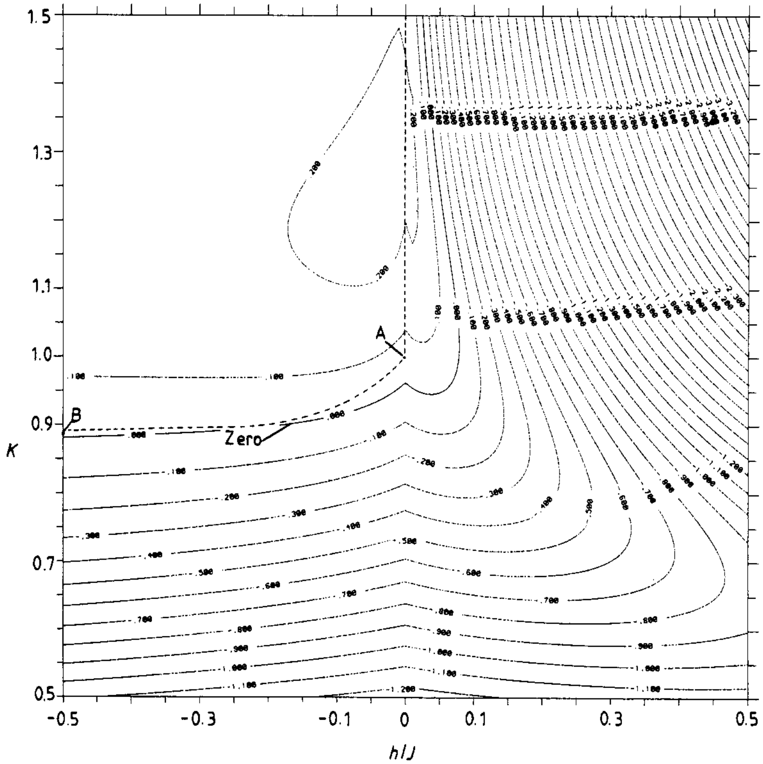

Figure 4. The contours of $\varphi_2(K, h)$ for the three-state Potts model Hamiltonian (18), the contours are those relating to one of the planes $h_i = h_j$ in figure 3. Here and in figure 5 the planar coexistence sheets are evidenced by the large plateau-like region in the domain $h < 0$. All the contours are cusped on the $h = 0$ line and the thumb-like loop of the zero contour inside $h > 0$ suggests the presence of the web-like extensions of figure 3. Points A and B are the exact zero-field transition points of the three- and two-state Potts models respectively, and the broken curve is a schematic representation of the limiting boundary line of Σ.

single column energies. For such sections of the square lattice the $N\to\infty$ limit above corresponds to $m\to\infty$; for m finite and R measured along a row we obtain

$$\Gamma_{pp'}(R)=\sum_{j=1}(\lambda_j/\lambda_0)^R(\varphi_0|\delta_{\sigma,\alpha_p}|\varphi_j)(\varphi_j|\delta_{\sigma,\alpha_{p'}}|\varphi_0) \tag{13}$$

where λ_0 is the maximum eigenvalue, and $\lambda_1>\lambda_2>\ldots$ are the other eigenvalues of $\boldsymbol{T}$ with corresponding eigenvectors φ_j. $\boldsymbol{T}$ is strictly non-degnerate at λ_0, hence (10) requires the asymptotic degeneracy of $\boldsymbol{T}$ in the limit $m\to\infty$, and this must be quite generally the case at all points on Σ. The only completely known case of this effect is seen in the Ising model solution of Onsager (1944) where λ_0 is asymptotically degenerate on the coexistence line $T\leqslant T_c, h=0$. Kac (1968) has argued that asymptotic degeneracy is a general mathematical mechanism for phase transitions and represents the appearance of two stable phases. Thus the scaling equation (8) could be alternatively viewed as an attempt to approximate a condition for degeneracy using finite systems in, say, the form

$$\xi_{m+1}(T,h)=(1+1/m)\xi_m(T,h). \tag{14}$$

over a surface in $(T,\boldsymbol{h})$ space. In relation to the scaling transformation, (14) is in effect an mth approximant to the whole of the phase equilibrium surface. If we define

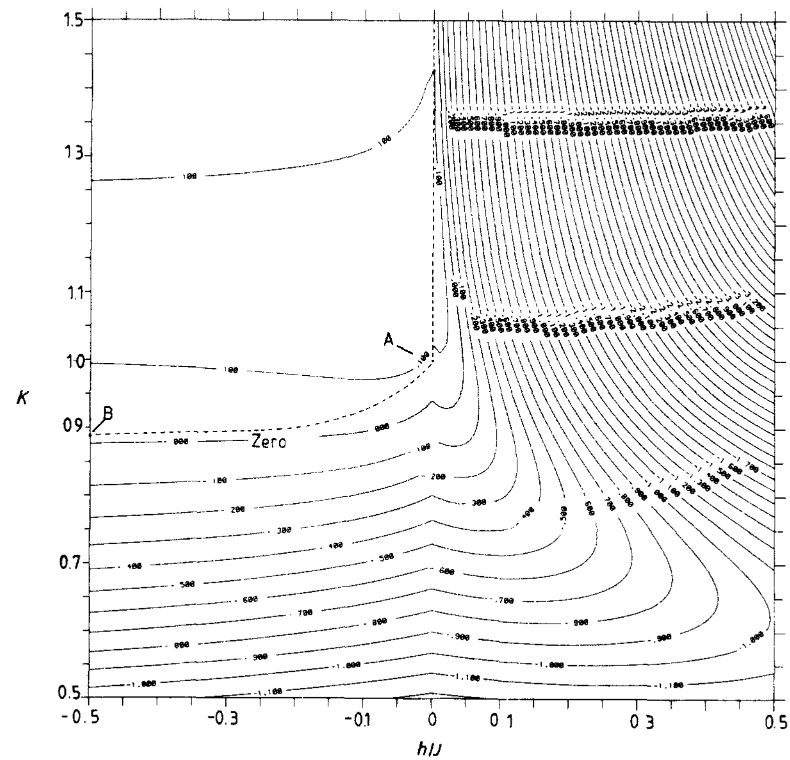

Figure 5. The contours of $\varphi_3(K,h)$ for the three-state Potts model. The zero-field contour has shifted closer to the limiting form of the boundary line of Σ (shown schematically by the broken curve) and the extent of the web-like extensions has diminished, see figures 9 and 10.

a sequence of functions φ_m in the form

$$\varphi_m(T, \boldsymbol{h}) = m\xi_m^{-1} - (m+1)\xi_{m+1}^{-1} \tag{15}$$

then the zero contour $\varphi_m = 0$ will approximate Σ, and the sequence of such contours should converge to Σ.

We can test this in relation to the Ising model where previous applications have only looked at the sequence of points $\varphi_m(T_c, 0) = 0$ (the critical point). Examples of the functions $\varphi_m(K, 0)(K = \beta J)$ are illustrated in figure 1, where the limiting line is shown schematically (broken line). To see how the phase diagram is approximated in the T, h plane, contour maps of φ_2, φ_3, and φ_4 are shown in figure 2. Here we can see that the critical point is not distinguished except as a probable end-point of the Σ line along $h = 0$. By contrast, the corresponding zero contours for the Ising model antiferromagnet† (see Muller-Hartmann and Zittartz 1977 and Goldfinch and Wood 1982) are also shown in figure 2, here the boundary of Σ is a line of order–disorder critical points and encloses a region which appears as a continuous sheet of coexistence

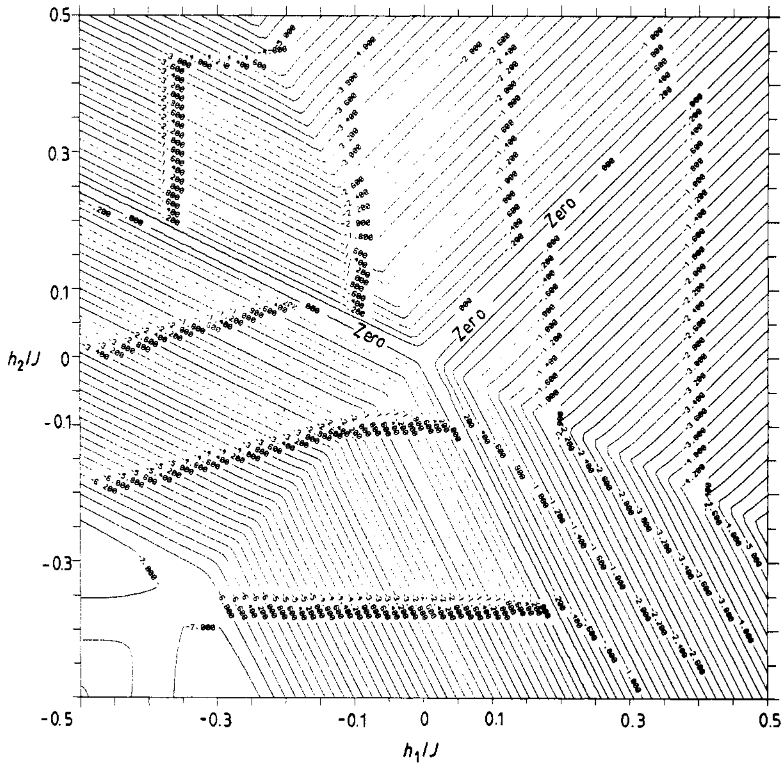

Figure 6. Contours of $\varphi_2(h_1, h_2)$ for the Hamiltonian of the three-state Potts model (19). The contours are taken in planes perpendicular to the temperature axis of figure 3, here the temperature has been fixed well below the transition point T_0, and the three coexistence sheets appear clearly as three lines originating form the origin.

† For the antiferromagnet, only even values of m can be used, and

$$\varphi_m(T, h) = m\xi_m^{-1} - (m+2)\xi_{m+2}^{-1}.$$

In figures 2(d) and (e) m is 2 and 4 respectively (Sneddon 1979).

points (similar sheets appear in the Potts model below). In the antiferromagnetic there are strictly two low-temperature phases which can coexist in a finite field, these are the domains in terms of the sequence 'up down up down . . .' being broken into the sequence 'down up down up . . .'.

3. Phase equilibrium in the three-state Potts model

To test the claims being made here for the scaling transformation we have applied it to the 2D three-state Potts model on the square lattice. The model Hamiltonian is

$$\mathcal{H} = -J \sum_{\mathrm{nn}} \delta_{\sigma_i \sigma_j} - \sum_i \sum_{l=1}^{3} h_l \delta_{\sigma_i l} \qquad \sigma_i \in (1, 2, 3) \tag{16}$$

where h_l is the ordering field of species l, and nn denotes nearest neighbour interactions. Following Straley and Fisher (1973) we impose the symmetry constraint

$$h_1 + h_2 + h_3 = 0 \tag{17}$$

and consider the case where two of the three fields are equal, hence with (17)

$$\mathcal{H} = -J \sum_{\mathrm{nn}} \delta_{\sigma_i \sigma_j} - h \sum_i (\delta_{\sigma_i 1} - \delta_{\sigma_i 2}/2 - \delta_{\sigma_i 3}/2). \tag{18}$$

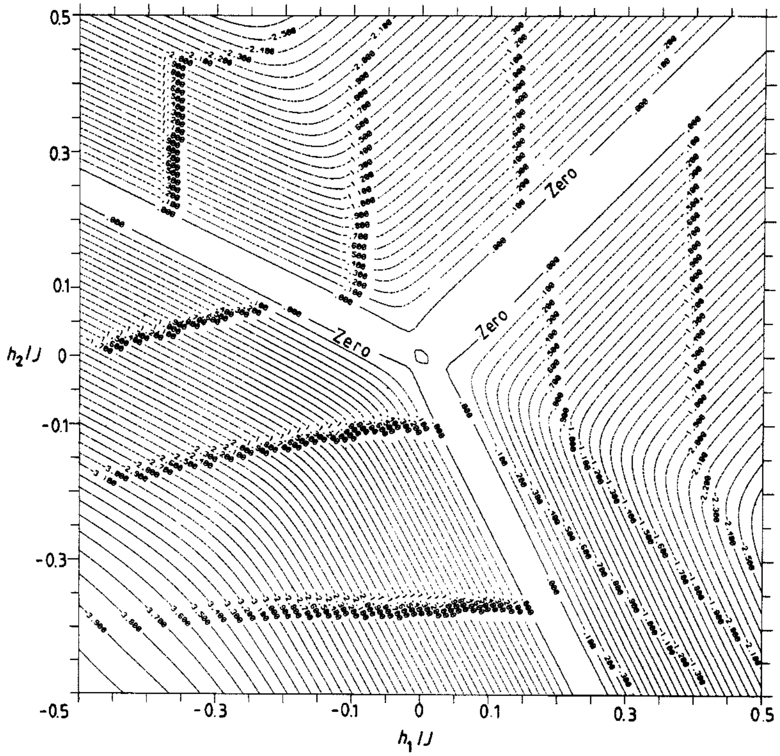

Figure 7. The contours as in figure 6 corresponding to a temperature close to the transition point T_0.

The outline form of the phase diagram for this model was originally conjectured by Straley and Fisher (1973) to be in the form shown in figure 3 (for a review see Wu (1982)). The three regions $h_i = h_j$ are the one-, two-, and three-rich phases in the domain $h_k > 0$; the negative field domains $h_k < 0$ are where we expect to find coexisting states. These are the three plane surfaces shown in figure 3, which are bounded by a line of critical points (shown dotted). The boundary lines should, in the limit $h \to -\infty$, approach the critical point of the two-state Potts model in zero field (the Ising model). The original conjecture that the plane surfaces were extended and connected by three web-like surfaces w_1, w_2, and w_3 is now thought to be true only in three dimensions. This view follows from the exact results obtained by Baxter (1973) in which the three- and four-state Potts models both have a second-order transition in zero field. Thus it is natural to think that the three boundary lines to the plane coexistence sheets meet at a point T_0 on the zero-field line. In this context the point T_0 is an anomalous tricritical point.

In figures 4 and 5 we show the contours of $\varphi_k(K, h)$ for $k = 2$ and 3 using the Hamiltonian (18) which confines the phase diagram to the $h_2 = h_3 = -h/2$ line (say). It appears to these authors quite remarkable that the large coexistence sheet in the domain $h < 0$ should be so sharply evidenced as a large plateau on the contour map

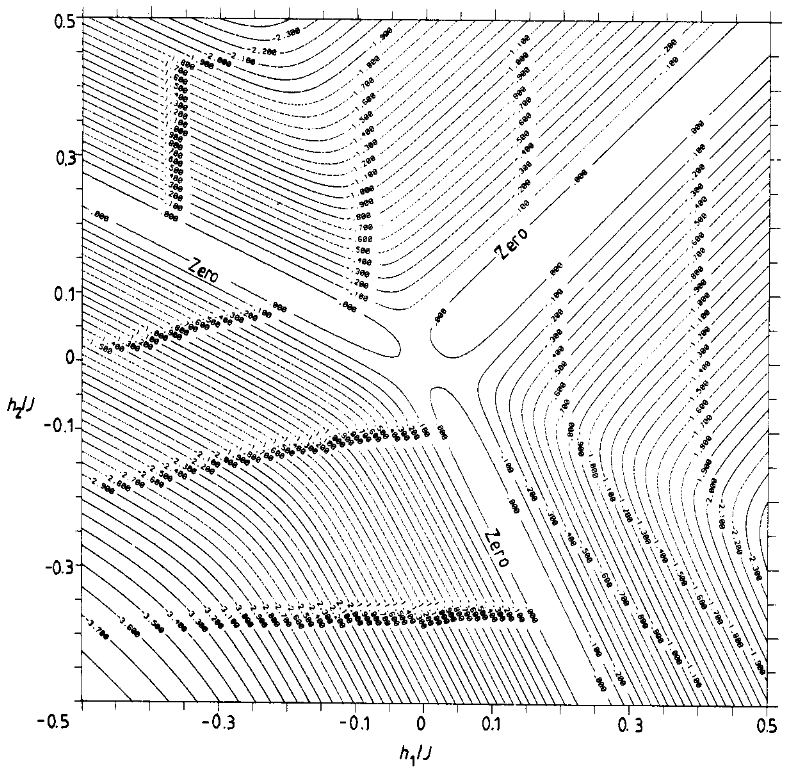

Figure 8. Contours as in figure 6 corresponding to a temperature between the two points marked A and B of figures 4 and 5. The hyperbolic enclosures of the origin in figures 6 and 7 have divided into three narrow loops on the zero contour. These clearly represent the sliced-off sections of the planar coexistence sheets above the temperatures t_1, t_2, and t_3 of figure 3 but below the limit points of the boundaries at $h \to -\infty$.

enclosed by the zero contour $\varphi_k = 0$. The small thumb-like loop just inside the domain $h > 0$ appears to represent the cross-section of the web w_1 along the h_1 axis, and the point T_0 is clearly evidenced by a cusp on Σ. Points which are marked on the figures are the exact location of T_0 ($K_0 = \ln(1+\sqrt{3}) = 1.00505\ldots$, Potts (1952), Baxter (1973)) and the critical point of the two-state Potts model which should be the limit points of the zero contours. We conjecture that the sequence of surfaces $\varphi_k = 0$ will converge rapidly to a good representation of the true coexistence surface (shown broken) in which the thumb-like loop has converged to the single point T_0.

We have also looked at this model in terms of two independent fields, for which the Hamiltonian can be put into the form

$$\mathscr{H} = -J \sum_{\mathrm{nn}} \delta_{\sigma_i \sigma_j} + h_1 \sum_i (\delta_{\sigma_i 1} - \delta_{\sigma_i 3}) + h_2 \sum_i (\delta_{\sigma_i 2} - \delta_{\sigma_i 3}) \qquad (19)$$

where by fixing K we can use the scaling transformation to view the intersection of the phase diagram in figure 3 with planes perpendicular to the temperature axis. Our results using φ_2 are shown in figures 6, 7, and 8 where the contours of $\varphi_2(h_1, h_2)$ are shown for temperatures below T_0, just above T_0, and just below the zero-field critical point of the two-state Potts model. Again the results seem to be quite remarkable for such a simple calculation. Figure 6 clearly shows the three coexistence planes $h_i = h_j$. In figure 8 we obtain exactly what is to be expected, namely three sections of the coexistence planes 'sliced' off before their common intersection at $h = 0$. In figure

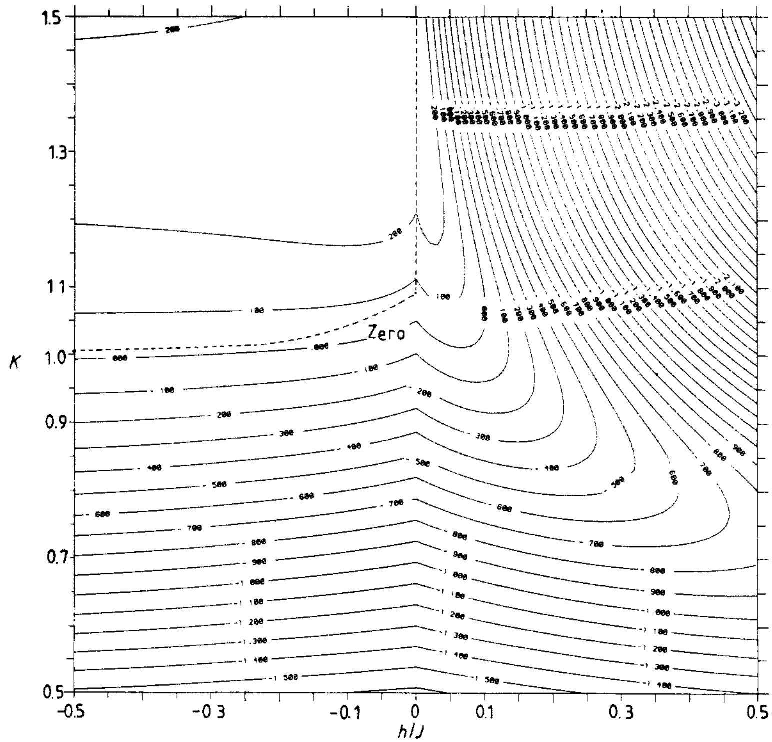

Figure 9. The contours of $\varphi_2(K, h)$ for the four-state Potts model, these contours correspond to those shown in figure 4 for the three-state Potts model.

7 the three disjoint zero contours of figure 8 have joined to form the three hyperbolic-like enclosures of the origin. This of course is consistent with the presence of the web-like extensions, but the resolution here is not sufficient to be certain of such a structure.

The generalisation of the Hamiltonian (18) to the q-state Potts model is

$$\mathcal{H} = -J \sum_{\mathrm{nn}} \delta_{\sigma_i \sigma_j} - h \sum_i \left[\delta_{\sigma_i 1} - \frac{1}{q-1} (\delta_{\sigma_i 2} + \delta_{\sigma_i 3} + \ldots + \delta_{\sigma_i q}) \right] \tag{20}$$

and we have used the scaling transformation to examine the phase equilibrium structure of the four- and five-state Potts models corresponding to the region shown in figures 4 and 5. The general q-state model in such a field has recently been looked at by Goldschmidt (1981) using high-q series expansions. Goldschmidt found that for $h > 0$ (and $q > 4$ (Baxter 1973)) in two dimensions there was evidence for a line of first-order transitions which terminated at a critical point in the h, T plane. This line appears to grow in length with increasing q. This of course is the cross-section of the web-like region as represented by the thumb loops above. The corresponding contours for the four- and five-state models are shown in figures 9 and 10. The thumb loops are are clearly present.

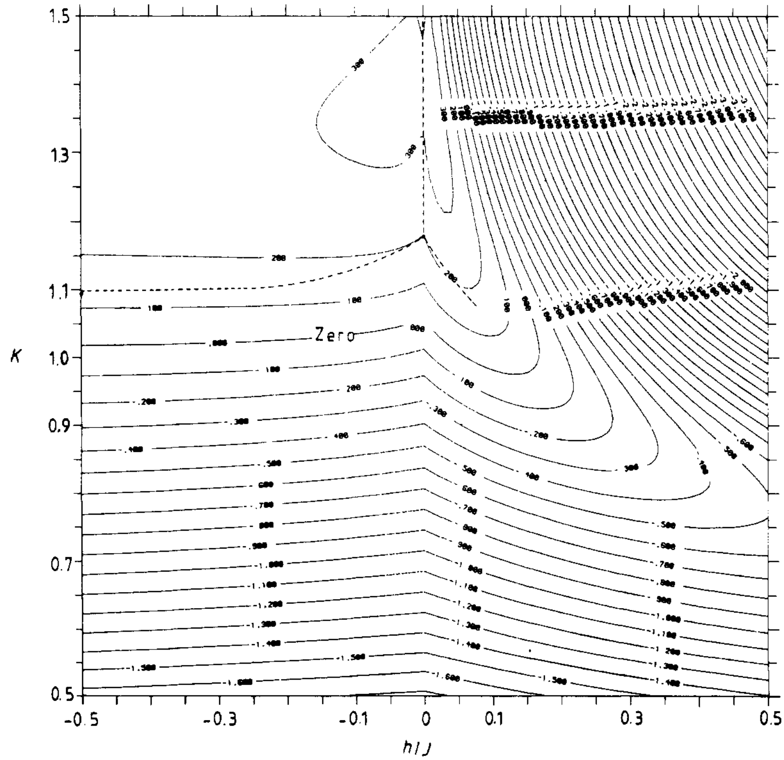

Figure 10. The contour $\varphi_2(K, h)$ of the five-state Potts model corresponding to those shown in figure 4 for the three-state model. Here as in the four-state model the planar and web-like structure of the Σ surface is clearly represented. The webs appear to be growing in size with q (see Goldschmidt 1981).

4. Summary and conclusions

We have argued that the scaling transformation (Nightingale 1976, Sneddon 1978, Wood and Goldfinch 1980) cannot be used to form estimates of critical parameters at critical points without prior knowledge that the point is a second-order transition point. The method does not distinguish order–disorder points from any other point on the coexistence or phase equilibrium surface. In effect the method is far more powerful than was originally envisaged, and is capable of providing a very good approximation to the whole phase equilibrium diagram. These claims have been illustrated by an application to the full phase equilibrium diagram of the three-state Potts model.

Acknowledgments

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