Droplet theory in low dimensions: Ising systems in an ordering field

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Abstract. We extend the microscopic droplet theory of Ising systems, developed recently, to incorporate the effects of an ordering field. The theory yields a free energy which is the solution of a renormalisation group equation describing droplet nesting, and which has a full scaling form. The behaviour near the coexistence curve is investigated and found to display the essential singularity suggested by primitive droplet models, but with parameters renormalised in a physically intelligible way. The droplet population function implied by the theory is parametrised in a simple way, in space dimension $d = 2$, to yield predictions for various thermodynamic properties, including the equation of state, which are found to be in fair accord with series-based results. The structure of the field-dependent droplet number distributions is investigated and contrasted with the forms assumed in phenomenological droplet theories. The domain of validity of the theory is assessed. It is concluded that the theory fails to describe (sufficiently) large droplets in the presence of a finite field, and is thus in principle trustworthy only in a region close to the coexistence curve.

1. Introduction

In a recent paper (Bruce and Wallace (1983), hereafter referred to as I) it was shown how one can construct an explicit and analytically tractable theory of the universal configurations underlying critical point phenomena in low-dimensional Ising systems. The theory represents the natural extension to space dimensions $d = 1 + \varepsilon$ of kink-based theories of the $d = 1$ Ising universality class (Bishop *et al* 1980). In $d = 1 + \varepsilon$ the basic configurational building block is a 'droplet' of one phase (Fisher 1967). The typical droplet is not spherical: deviations from the spherical state give the droplet surface area the character of a 'fractal' (Mandelbrot 1983) whose dimension prescribes the critical point thermal eigenvalue (the critical exponent $1/\nu$). The typical droplet is not homogeneously ordered either: deviations from the homogeneous state, realised as 'nested' droplets (Kadanoff 1976), give the droplet volume a fractal character, with a dimension which identifies the ordering field eigenvalue (effectively the ratio β/ν). Both the fluctuations from the spherical state, and the fluctuations from the homogeneous state, are controlled by the statistical mechanics of surface tension, which can be handled in a controlled way by renormalisation group methods in $1 + \varepsilon$ dimensions.

  The work reported in I was confined to the line of two-phase coexistence (the 'zero-field' limit, in the language of magnetism which we shall employ here). In the present paper we extend the analysis to include the effects of an ordering field. The motivation is fourfold.

Firstly, the extended analysis allows us to establish in a unified and systematic fashion the forms of the various field-derivatives of the free energy (specifically the order parameter and its susceptibility) whose critical behaviour on the coexistence curve was obtained in I with the aid of a novel, but perhaps less than transparent, 'decoration' argument.

Secondly, the analysis developed here allows us to establish the analytic structure of the free energy in the vicinity of the coexistence curve and critical point: we are thus able to determine the effects of critical fluctuations on the essential singularity (as a function of the ordering field) found in thermodynamic quantities in the subcritical region (Günther *et al* 1980, Lowe and Wallace 1980) and believed to control nucleation phenomena (Langer 1967).

The third motivation for the present study is to shed some light on the domain of validity of the droplet-based theory. As noted in I, while the theory remains controllable for any $T < T_c$, it undoubtedly breaks down for $T > T_c$ in zero field. It is thus natural to enquire in what region of the field–temperature plane the theory can be trusted.

Finally the incorporation of an ordering field is a prerequisite for dealing with a number of potentially interesting extensions of the theory, most notably the problem of percolation, which will be addressed in a separate paper (Schmittmann and Bruce 1985). The layout of the present paper is as follows.

In § 2 we determine the configurational weight (partition function) for a single droplet of one phase embedded in the opposite phase, incorporating into the analysis a field which couples to the droplet volume. In § 3 we show how this single-droplet partition function may be utilised to determine the full partition function and free energy of a many-droplet assembly. In contrast to the zero-field case treated in I, where the free energy emerges in a simple and explicit fashion, in the presence of a field we find that the free energy is prescribed implicitly as the solution of an integral equation. In § 4 we establish analytically the thermodynaic properties implied by the form of the free energy, demonstrating its scaling form, confirming the zero-field results of I for the field derivatives, and examining the effects of fluctuation renormalisation on the essential singularity. We then proceed to an explicit numerical study of the thermodynamic behaviour (notably the equation of state) in the particuar case $d = 2$. In § 5 we determine the form of the droplet number distribution, examining its implications for the validity of phenomenological droplet theories (Binder 1976). Finally, in § 6, we assess the theory we have developed, giving particular attention to its domain of validity in the field–temperature plane.

2. The single droplet partition function in finite field

As in I, we study the continuum limit of the fixed-length-spin Ising model. This model realised as a field theory is analytically more tractable than its lattice counterparts and we expect it to display the same (universal) configurational features as those of the lattice models. In the continuum limit a droplet is a connected region of space with an intrinsically sharp closed boundary separating the two ordered phases. We assume that the statistically dominant droplets are almost hyperspherical: it was proved in I that this assumption is at least self-consistent near dimension $d = 1$ and it turns out that the presence of an ordering field does not affect its validity.

We can thus characterise the size and shape of a droplet by the single valued function $f(\eta)$ which specifies the deviation of the droplet surface from a reference

hypersphere of radius R (I, figure 1). Here η specifies a direction from the centre of the reference droplet. The essential geometrical features of a droplet are its volume and surface area. The volume is given by (equation (I2.3))

$$\mathcal{V} = \mathcal{V}(f) = d^{-1} \int d\Omega \, (R+f)^d \tag{2.1}$$

where $d\Omega$ is the element of solid angle in d dimensions. The surface area is given by (equation (I2.7))

$$\mathcal{A} = \mathcal{A}(f) = \int d\Omega \, (R+f)^{d-1} [1 + \tfrac{1}{2}(R+f)^{-2}(L_{ij}f)^2]^{1/2} \tag{2.2}$$

where the L_{ij} are the angular momentum operators in d dimensions. The field f has a decomposition in the d-dimensional spherical harmonics $Y_{l,m}(\eta)$, which are the eigenfunctions of L^2:

$$f(\eta) = \sum_{l,m} a_{l,m} Y_{l,m}(\eta). \tag{2.3}$$

Within the continuum framework the dimensionless configurational energy, $\mathcal{H}$, of a sigle droplet relative to the zero-droplet ground state is uniquely prescribed by the droplet area and volume:

$$\mathcal{H} = T_0^{-1}\mathcal{A} + (-1)^\alpha 2h\mathcal{V}. \tag{2.4}$$

The first term represents the surface ('broken bond') energy; T_0^{-1} is a measure of the surface tension. The second term represents the field energy: $\alpha = 1$ or 2 according to the phase of the droplet; phase 1 is favoured by a positive external field, $h > 0$.

The single-droplet partition function (SDPF) is written as a functional integral over all fields f:

$$Z_{1,\alpha}(h) = \int Df \, e^{-\mathcal{H}(f)} = \int \left(\prod_{l,m} da_{l,m} \right) e^{-\mathcal{H}}. \tag{2.5}$$

The arguments supporting the choice of measure in (2.5) are given in appendix 1 of I where it was shown that the implied normalisation of Z_1 is consistent with a zero-droplet partition function that is unity.

The evaluation of the SDPF in zero field was described in detail in I. Here we shall summarise the results and indicate how they are modified by the presence of the ordering field. In zero field we found for a droplet of phase α embedded in phase $\bar{\alpha}$ (equation (I4.1))

$$Z_{1,\alpha}(h=0) = V_{\bar{\alpha}} \int dR \, v^{-1}(R)\psi(R,\xi). \tag{2.6}$$

Here $V_{\bar{\alpha}}$ is the volume of the embedding phase, $v(R) = S_d R^d / d$ is the volume of a hypersphere of radius R and ξ is the correlation length, with a critical behaviour prescribed by equation (I3.28b). The function $\psi(R, \xi)$ has the properties

$$\psi(R, \xi) = R^{-1} \tilde{\psi}(R/\xi) \tag{2.7a}$$

with

$$\tilde{\psi}(z) \approx \psi_0, \qquad\qquad\qquad z \ll 1, \tag{2.7b}$$

$$\approx \psi^{(\infty)} z^{\varepsilon(2+\varepsilon)/2} \exp(-S_d z^\varepsilon / c_0), \qquad z \gg 1. \tag{2.7c}$$

The parameter ψ_0 is a universal constant, exponentially small in the parameter $\varepsilon = d - 1$ (equation (I4.4d)). The parameter c_0 is a dimensionless constant which effectively prescribes the universal relationship between the dimensionless surface tension, $c_0^{-1}\xi^{-\varepsilon}$, and the correlation length ξ (Bruce 1984).

It is straightforward to extend these results to incorporate the field term in the configurational energy (2.4). One must first recall from I that the integrations with respect to the $l = 0$ and $l = 1$ coordinates in equation (2.5) have to be treated by collective coordinate methods: the integration with respect to the $l = 0$ mode yields an integration with respect to R, with a trivial Jacobian; the integration with respect to the d $l = 1$ modes becomes an integration with respect to the centre of the droplet (yielding the embedding volume) modulo a Jacobian which is in general non-trivial (i.e. f dependent) but which turns out to be effectively a constant at both one- and two-loop levels (Schmittmann 1984). One then finds that the finite-field partition function may be written in the form

$$Z_{1,\alpha}(h) = V_{\bar{\alpha}} \int dR\, v^{-1}(R)\psi(R, \xi) \left\langle \exp\left(-(-1)^\alpha 2hd^{-1} \int d\Omega\, [R + f(\eta)]^d \right) \right\rangle'_0 \qquad (2.8a)$$

where

$$\langle 0 \rangle'_0 \equiv \int \left(\prod_{l,m}' da_{l,m} \right) \exp[-\mathcal{H}(h=0)]0 \Big/ \int \left(\prod_{l,m}' da_{l,m} \right) \exp[-\mathcal{H}(h=0)]. \qquad (2.8b)$$

The prime denotes omission of the $l = 0$ and $l = 1$ spherical harmonic contributions to $f(\eta)$; the subscript zero indicates that the average is to be evaluated in the zero-field ensemble. Now

$$d^{-1} \int d\Omega\, [R + f(\eta)]^d = d^{-1}S_d R^d + \int d\Omega\, R^d \left(\frac{f(\eta)}{R} + \frac{\varepsilon f^2(\eta)}{R^2} + O(f^3) \right)$$

$$= v(R) + \varepsilon R^{d-2} \int d\Omega\, f^2(\eta) + O(f^3) \qquad (2.9)$$

where we have used the fact that the integral whose integrand is linear in f vanishes in the absence of the $l = 0$ component of the field. Writing

$$\left\langle \exp\left(-(-1)^\alpha 2h\varepsilon \int d\Omega\, R^{d-2}f^2(\eta) \right) \right\rangle'_0 = \exp[-(-1)^\alpha 2h\varepsilon S_d R^{d-2}\langle f^2(\eta) \rangle'_0 + O(f^4)]$$

$$\qquad (2.10)$$

and utilising equation (I4.18) for the variance $\langle f^2(\eta) \rangle$, we find that

$$\left\langle \exp\left(-(-1)^\alpha 2hd^{-1} \int d\Omega\, [R + f(\eta)]^{d-1} \right) \right\rangle'_0 = \exp\{[-(-1)^\alpha 2hv(R)[1 + O(\varepsilon^3)]\} \qquad (2.11)$$

implying that the fluctuation corrections to the 'classical' field-dependent weighting factor are negligible in low order. The SDPF in finite field thus assumes the simple form

$$Z_{1,\alpha}(h) = V_{\bar{\alpha}} \int dR\, v^{-1}(R)\psi(R, \xi) \exp[-(-1)^\alpha 2hv(R)]. \qquad (2.12)$$

3. The multi-droplet partition function

We now proceed to use the results of § 2 to determine the thermodynamic properties of the full many-droplet assembly, taking account of droplet nesting, and droplet interaction, to the extent that the latter is expressed in excluded volume effects. The key simplification in the analysis is the dilute droplet boundary approximation which was explained and justified in I and which facilitates a simple renormalisation group type of argument yielding an equation for the free energy of the multi-droplet ensemble. The domain of validity of the argument in the presence of a field requires further consideration which we defer to § 6.

We consider an Ising system which consists of a hyperspherical volume $V = d^{-1} S_d L^d$ containing droplets of both phases and of all scale sizes intermediate between L (in practice ξ) and some arbitrary minimum L_0. The partition function for this system is written as a sum over the set of all distinct configurations, $\{c(L, L_0)\}$, of all possible droplets of all possible shapes:

$$Z(h, L, L_0, \xi) = \sum_{\{c(L,L_0)\}} \exp\{-\mathcal{H}[c(L, L_0)]\} \tag{3.1}$$

where $\mathcal{H}[c(L, L_0)]$ is the dimensionless energy associated with the configuration $c(L, L_0)$. We proceed, as in I, by separating out the contribution made to this sum by the smallest droplets in the assembly—those with scale sizes between $L_0 + dL_0$ and L_0:

$$Z(h, L, L_0, \xi) = \sum_{\{c(L,L_0+dL)\}} \sum_{\{c(L_0+dL_0,L_0)\}^*}$$
$$\times \exp\{-\mathcal{H}[c(L, L_0+dL_0)] - \mathcal{H}[c(L_0+dL_0, L_0)]\} \tag{3.2}$$

where $\{c(L_0+dL_0, L_0)\}^*$ means the set of configurations of droplets with scale sizes between L_0+dL_0 and L_0 which are consistent with the configuration $c(L, L_0+dL_0)$ of larger droplets. We now invoke the dilute droplet boundary approximation: we assume that the volume, $V_\alpha[c(L, L_0+dL_0)]$, occupied by droplets of phase α in the configuration $c(L, L_0+dL_0)$ is such that there is a high probability that a droplet of scale size $\sim L_0$ embedded in this volume is located far from a droplet boundary. We may thus neglect the constraints on the configurational sum over the L_0-sized droplets originating in the finite and ramified character of the volumes $V_\alpha[c(L, L_0+dL_0)]$ which they occupy. The problem posed by this configurational sum is then essentially that addressed in § 2. Specifically, with the aid of equation (2.12), we find

$$\sum_{\{c(L_0+dL_0, L_0)\}^*} \exp\{-\mathcal{H}[c(L_0+dL_0, L_0)]\}$$
$$= 1 + V_2[c(L, L_0+dL_0)]v^{-1}(L_0) \exp[+2hv(L_0)]\psi(L_0, \xi) \, dL_0$$
$$+ V_1[c(L, L_0+dL_0)]v^{-1}(L_0) \exp[-2hv(L_0)]\psi(L_0, \xi) \, dL_0 + O(dL_0)^2. \tag{3.3}$$

The first term is the contribution from configurations $c(L_0+dL_0, L_0)$ containing no droplets, the second is from configurations containing one droplet of phase 1 only and the third is from configurations containing only one phase-2 droplet. Substituting (3.3) into (3.2) gives

$$Z(h, L, L_0, \xi)$$
$$= \sum_{\{c(L,L_0+dL_0)\}} \exp\{-\mathcal{H}[c(L, L_0+dL_0)]\}$$
$$\times \{1 + V_1[c(L, L_0+dL_0)]v^{-1}(L_0) \exp[-2hv(L_0)]\psi(L_0, \xi) \, dL_0$$
$$+ V_2[c(L, L_0+dL_0)]v^{-1}(L_0) \exp[+2hv(L_0)]\psi(L_0, \xi) \, dL_0\} \tag{3.4a}$$

implying

$$\frac{\partial \ln Z(h, L, L_0, \xi)}{\partial L_0} = -v^{-1}(L_0)\psi(L_0, \xi)\langle V_1 \exp[-2hv(L_0)] + V_2 \exp[2hv(L_0)]\rangle_{L_0} \quad (3.4b)$$

where $\langle \ \rangle_{L_0}$ denotes averaging over the canonical ensemble with infrared cut-off (minimum droplet size) L_0.

Noting the identities

$$\exp\{-\mathcal{H}[c(L, R)]\}\{V_1[c(L, R)] - V_2[c(L, R)]\} = (\partial/\partial h) \exp\{-\mathcal{H}[c(L, R)]\} \quad (3.5a)$$

and

$$V_1[c(L, R)] + V_2[c(L, R)] = V \quad (3.5b)$$

we find that equation (3.4) may be recast in the form

$$\frac{\partial \ln Z(h, L, L_0, \xi)}{\partial L_0}$$

$$= -Vv^{-1}(L_0)\psi(L_0, \xi)\left(\cosh[2hv(L_0)] \right.$$

$$\left. -\sinh[2hv(L_0)]V^{-1} \frac{\partial \ln Z(h, L, L_0, \xi)}{\partial h} \right). \quad (3.6)$$

Integrating this equation with the boundary condition

$$Z(h, L, L, \xi) = e^{hV} \quad (3.7)$$

we find that the dimensionless free energy

$$F(h, L, L_0, \xi) \equiv \ln Z(h, L, L_0, \xi) \quad (3.8)$$

obeys the integrodifferential equation

$$F(h, L, L_0, \xi) = V\left[h + \int_{L_0}^{L} dR\, v^{-1}(R)\psi(R, \xi) \right.$$

$$\left. \times \left(\cosh[2hv(R)] - \sinh[2hv(R)]\frac{1}{V}\frac{\partial F(h, L, R, \xi)}{\partial h} \right) \right]. \quad (3.9)$$

Finally, taking the thermodynamic $(L/\xi \to \infty)$ limit we obtain for the free energy density $f = V^{-1}F$ the representation

$$f(h, L_0, \xi) = h + \int_{L_0}^{\infty} dR\, v^{-1}(R)\psi(R, \xi)\{\cosh[2hv(R)] - Q(h, R, \xi)\sinh[2hv(R)]\}$$

$$(3.10a)$$

where (cf equation (3.5a))

$$Q(h, R, \xi) \equiv \frac{\partial f(h, R, \xi)}{\partial h} = \lim_{L\to\infty} V^{-1}\langle V_1 - V_2 \rangle_R \quad (3.10b)$$

represents the order parameter for an assembly with minimum droplet size R.

These equations constitute the basic results of this paper. In the limit of zero field they simplify to give the fully explicit representation of the free energy (equation (15.8))

$$f(0, L_0, \xi) = \int_{L_0}^{\infty} dR \, v^{-1}(R) \psi(R, \xi). \tag{3.11}$$

This simplification may be traced to the fact that, in zero field, the two phase volumes V_1 and V_2 appearing in (3.3) carry identical weightings: in view of the sum rule (3.5b) the RHS of (3.3) is thus *independent of the specific configuration* $c(L, L_0 + dL_0)$ of the larger droplets. In the presence of a field, however, these two terms can no longer be combined in this fashion: the result is the implicit equation (3.10), whose properties we now turn to examine.

4. Thermodynamic properties

We have established that the free energy of the many-droplet assembly obeys an implicit integral equation, (3.10). In this section we investigate the properties of the free energy implied by this equation recast in differential form by taking the derivative with respect to the minimum droplet scale size, L_0:

$$\frac{\partial f(h, L_0, \xi)}{\partial L_0} = -v^{-1}(L_0) \psi(L_0, \xi) \left(\cosh[2hv(L_0)] - \frac{\partial f(h, L_0, \xi)}{\partial h} \sinh[2hv(L_0)] \right). \tag{4.1}$$

Specifically, in § 4.1, we shall look for a solution to this equation in the form of a power series in the external field. In the process we shall establish that the free energy displays a properly scaling form in the critical region. From the low-order terms in the series solution we shall determine the zero-field forms of the free energy, the order parameter and the susceptibility; these results corroborate the predictions made in I on the basis of rather different arguments. In the form of the higher-order terms we shall find the signature of an essential singularity on the coexistence curve; these results substantiate the predictions of a subcritical droplet model calculation by Günther *et al* (1980) and refine them by the incorporation or critical fluctuations (droplet nesting) whose effect, it transpires, is simply to introduce a physically intelligible ξ-dependence into the basic parameters characterising the singularity.

In § 4.2 we shall turn our attention to the particular case of two dimensions. We shall establish, numerically, the implications of equation (4.1) for the equation of state and other characteristics of the $d = 2$ Ising universality class, comparing the predictions with the results of series expansion studies.

4.1. Series solution for the free energy

We consider the low-field expansion for the free energy,

$$f(h, L_0, \xi) = \sum_{n=0}^{\infty} f_n(L_0, \xi) \frac{h^n}{n!}. \tag{4.2}$$

In the critical region we expect the free energy to assume a scaling form, so we make the ansatz

$$f_n(L_0, \xi) = \xi^{-d} [\xi^d \tilde{Q}(L_0/\xi)]^n \tilde{f}_n(L_0/\xi) \tag{4.3a}$$

with

$$\tilde{Q}(L_0/\xi) \equiv \exp\left(-2\int_{L_0/\xi}^\infty z^{-1}\tilde{\psi}(z)\,\mathrm{d}z\right). \tag{4.3b}$$

Substitution of (4.2) and (4.3) into the differential equation (4.1) shows that the proposed scaling form is correct and that the functions $\tilde{f}_n(L_0/\xi)$ satisfy the differential equations

$$\tilde{f}'_n(z) = \tilde{\psi}(z)z^{-1}v^{-1}(z)\left(-[2v(z)]^n\delta_{n,e}\tilde{Q}(z)^{-n}\right.$$

$$\left. +n!\sum_{0<l<n/2}\frac{\tilde{Q}^{-2l}(z)\tilde{f}_{n-2l}(z)[2v(z)]^{2l+1}}{(n-2l-1)!\,(2l+1)!}\right) \tag{4.4a}$$

where

$$\delta_{n,e} = \begin{cases} 1, & n \text{ even,} \\ 0, & n \text{ odd.} \end{cases}$$

These differential equations have boundary conditions which are determined by the form of the free energy *outside* the critical region, where $L_0 \gg \xi$. From (3.10) one sees that, to within exponentially small corrections,

$$f(h, L_0, \xi) = h, \qquad L_0 \gg \xi. \tag{4.4b}$$

Equation (4.4a) then yields for the $n = 0$ term (the zero-field free energy) the result

$$f(h=0, L_0, \xi) \equiv f_0(L_0, \xi) = \xi^{-d}\tilde{f}_0\left(\frac{L_0}{\xi}\right) = \xi^{-d}\int_z^\infty \mathrm{d}z\, z^{-1}v^{-1}(z)\tilde{\psi}(z). \tag{4.5a}$$

This equation recovers the zero-field free energy determined in I (equation (I5.8)) where it was shown that the implied critical behaviour is generally of the form (cf however, the $d = 2$ case discussed in § 4.2.1)

$$f(h=0, L_0, \xi) \approx \xi^{-d}\tilde{f}_S + f_{NS} \tag{4.5b}$$

where $\tilde{f}_S$ is a constant and f_{NS} contains contributions which are analytic in the reduced temperature.

Setting $n = 1$ we find from equation (4.4a) that $f'_1(z) = 0$ whence we conclude from (4.2), (4.3) and (4.4b) that the function $\tilde{Q}(L_0/\xi)$ defines the zero-field order parameter

$$Q(h=0, L_0, \xi) \equiv \partial f(h, L_0, \xi)/\partial h|_{h=0} = \tilde{Q}(L_0/\xi) \tag{4.6a}$$

again recovering a result of I. The implied critical behaviour follows from the definition (4.3b) and the properties (2.7) of the scaling function $\tilde{\psi}$:

$$Q(h=0, L_0, \xi) \sim \xi^{-\beta/\nu}, \qquad \beta/\nu = 2\psi_0. \tag{4.6b}$$

Next we observe that the $n = 2$ term in the expansion (4.2) defines the zero-field susceptibility

$$\chi(h=0, L_0, \xi) \equiv \frac{\partial^2 f(h, L_0, \xi)}{\partial h^2}\bigg|_{h=0} \equiv f_2(L_0, \xi) = \xi^d\tilde{Q}^2(z)\tilde{f}_2(z). \tag{4.7a}$$

In the small-z regime $\tilde{f}_2(z)$ tends to a finite limiting value which follows from (4.4a)

and (4.4*b*):

$$\tilde{f}_2(0) = 4 \int_0^\infty dz\, z^{-1} v(z) \tilde{\psi}(z) \exp\left(4 \int_z^\infty \frac{\tilde{\psi}(z')\, dz'}{z'}\right). \tag{4.7b}$$

Thus

$$\chi(h = 0, L_0, \xi) \approx \xi^d Q^2(h = 0, L_0, \xi) \tilde{f}_2(0)$$

$$\sim \xi^{\gamma/\nu}, \qquad \gamma/\nu = d + 4\psi_0, \tag{4.7c}$$

again recovering a result of I, where χ was obtained by considering fluctuations in the ordering coordinates. The general structure of the series solution (4.2), in the scaling regime, is now apparent. Utilising (4.5*b*) and exploiting the face that, quite generally, the functions $\tilde{f}_n(z)$ have finite $z = 0$ limits (for $n > 1$), we see that

$$f(h, L_0, \xi) \approx \xi^{-d} \tilde{f}_0 + \xi^{-d} \sum_{n=1}^\infty \frac{1}{n!} [h \xi^d \tilde{Q}(z)]^n \tilde{f}_n(0) \tag{4.8a}$$

which displays the anticipated scaling form

$$f(h, L_0, \xi) \approx \xi^{-d} \tilde{f}(h \xi^{d-\beta/\nu}) \tag{4.8b}$$

to within corrections which are analytic in the reduced temperature.

Returning now to the explicit structure of the coefficients in the expansion (4.2) we note that, for $n > 2$, the differential equations (4.4*a*) become progressively more complicated: in general the equation for the nth-order coefficient involves the behaviour of all the $(n - 2l)$th coefficients. We may, however, establish an approximate solution by noting that, for $n > 1$, the functions $\tilde{f}_n$ are all formally of the order of the small parameter ψ_0: we then systematically discard those terms in (4.4*a*) which are of order ψ_0^2. We have checked that this approximation does not invalidate the expressions for the asymptotic (large-n) behaviour of the expansion for the free energy, whose form we now proceed to establish.

Within the stated approximation the differential equations (4.4*a*) assume closed decoupled forms which can be trivially integrated to give

$$\tilde{f}_n(0) \approx (-2)^n \int_0^\infty dz\, \tilde{\psi}(z) z^{-1} v^{n-1}(z). \tag{4.9}$$

Now for large n the dominant contributions to the remaining integral come from a region where z is sufficiently large that $\tilde{\psi}(z)$ may be approximated by its asymptotic form (2.7*c*). Recalling (4.8*a*), one then readily finds that the coefficient of h^n in the expansion of the free energy is

$$\frac{f_n(L_0, \xi)}{n!} = \xi^{-d} \frac{[Q(h = 0, L_0, \xi)\xi^d]^n}{n!} \tilde{f}_n(0)$$

$$= \xi^{-d} \frac{d\psi^{(\infty)}}{(d-1)S_d} \left(\frac{c_0 n d}{S_d(d-1)}\right)^{(d^2 - 2d - 1)/2(d-1)}$$

$$\times \left[-2 S_d d^{-1} \xi^d \left(\frac{c_0}{S_d}\right)^{d/(d-1)} Q(h = 0, L_0, \xi)\right]^n$$

$$\times \frac{\Gamma[nd/(d-1)]}{\Gamma(n+1)} \left[1 + O\left(\frac{1}{n}\right)\right] \tag{4.10}$$

where we have used the asymptotic properties of the Γ function. The discarded terms include contributions originating in the corrections to the asymptotic behaviour of the function $\tilde{\psi}(z)$. This result may be usefully compared with that yielded by analysis of a Landau–Ginzburg droplet model in the subcritical region (Günther *et al* 1980, Lowe and Wallace 1980), according to which the coefficient of the nth-order term has the form

$$\frac{f_n(L_0, \xi)}{n!} = (-1)^n \frac{BA^{b/a}}{\pi a} A^{-n/a} \Gamma\left(\frac{n-b}{a}\right)[1 + O(n^{(a-2)/a})] \qquad (4.11a)$$

with

$$a \equiv d - 1, \qquad b \equiv (3-d)d/2, \qquad (4.11b)$$

while A and B are unknown constants. In the 'dilute' (subcritical) theory the coefficients (4.11a) are obtained, through a dispersion relation, from the imaginary part of the analytic continuation of the free energy (across the coexistence curve), which has the asymptotic form for small $|h|$

$$\mathrm{Im}\, f(h, \arg h = +\pi) = -B|h|^b \exp\{-A|h|^{-a}[1 + O(h^2)]\} \qquad (4.12)$$

Comparison of (4.10) with (4.11a) reveals consistency *both* as regards the leading n-dependence of the coefficients themselves *and* as regards the next to leading, $O(1/n)$, term in the ratio of successive coefficients. Matching the expressions prescribes for A and B the explicit forms

$$A = \frac{S_d}{d} \frac{(d-1)^{d-1}}{2^{d-1}} [Q(h=0, L_0, \xi)]^{1-d}(c_0\xi^{d-1})^{-d} \sim [\xi^d Q(h=0, L_0, \xi)]^{-a}, \qquad (4.13a)$$

$$B = \psi^{(\infty)}\left(\frac{\pi d^2 c_0^{d+1}}{2(d-1)S_d^3}\right)\left(\frac{2}{d-1}\right)^{(3-d)d/2} [Q(h=0, L_0, \xi)]^{(3-d)d/2}(c_0\xi^{d-1})^{d(2-d)/2}$$

$$\sim \xi^{-d}[\xi^d Q(h=0, L_0, \xi)]^b. \qquad (4.13b)$$

These results merit some comment. Firstly we note that, though A and B are non-universal, their non-universality resides simply in the two usual non-universal scale factors (implicit in the definitions of the order parameter and the correlation length): accordingly the coefficients of the powers of ξ and Q appearing in (4.13a, b) should be universal. Secondly we observe that the net ξ dependence which the coefficients display in the critical region is, of course, just such as to ensure that (4.12) exhibits the expected scaling behaviour. Thirdly, to expose the physical significance of the structure displayed by equation (4.13a) we recall that, within the framework of the dilute droplet theory leading to equation (4.12), the parameter A is found to be such that the argument of the exponential in (4.12) is simply the (dimensionless) freed energy of the critical droplet for nucleation (Günther *et al* 1980); this critical droplet free energy is defined as that which maximises the sum of the interfacial free energy of a single classical droplet, and the free energy associated with its interactions with a field favouring its presence. Equation (4.13a) shows that this interpretation carries over into the critical region with two simple caveats. Specifically, one must take account of the renormalisation of the interfacial tension: the factor $c_0^{-1}\xi^{1-d}$ defines the critical surface tension in the present framework (cf equation (2.7c) *et seq*). One must also allow for a renormalisation of the field energy: the factor of Q^{1-d} ensures that the free energy cost (per unit critical droplet volume) has magnitude hQ, rather than simply h. Equation (4.13a) is thus quite transparent. Indeed such a result has recently been

conjectured by Binder (1984) in a study of nucleation barriers, and by Harris (1984) who has shown that with its aid one may understand aspects of series expansion studies of the $d = 2$ lattice Ising model, effectively determining the parameter A (Baker and Kim 1980). The critical behaviour of the amplitude A is found to be in accord with equation (4.13a) (using *exact* results for ξ and Q), both as regards the index of the implied power law *and* as regards the amplitude (universal in the sense indicated above). In fact Harris has found numerical evidence to suggest that equation (4.13a) actually holds even outside the critical region, provided it is generalised to allow for an anisotropic surface tension (which is no longer related to the correlation length in the simple power-law fashion implicit in (4.13a)).

The structure of equation (4.13b) is rather less transparent and, indeed, is not prescribed by the phenomenological arguments of Binder (1984) and Harris (1984) which do not incorporate the effects of the droplet zero modes (radial distortions and translations) which control its form. We shall examine the numerical value of the coefficient B implied by this result in the course of the explicit numerical studies of the $d = 2$ case, to which we now turn.

4.2. Numerical studies in $d = 2$

Although the droplet theory presented here is formally justifiable only in $d = 1 + \varepsilon$, it is, nevertheless, useful to consider its application to $d = 2$ where its reliability may be assessed by comparison with established data for the two-dimensional Ising model. This is the task we now address.

The key to this programme is the droplet population function $\tilde{\psi}$ whose limiting characteristics are defined in equations (2.7b, c) but which must now be fully prescribed. Although a full two-loop calculation of this function now exists (Schmittmann 1984), we have chosen to utilise the one-loop form (prescribed by equations (I4.3), (3.28a)) which is particularly tractable when extrapolated to $d = 2$. Explicitly we find

$$\psi(z) = \psi_0 (1 + T_c z / c_0)^{3/2} \exp(-2\pi z / c_0). \tag{4.14}$$

In obtaining this form we have made the assigments $\nu = 1$ (and $\varepsilon = 1$) in equation (I3.28a). In the same spirit the parameter ψ_0 is assigned the value implied by the identification (4.6b), namely $\psi_0 = \beta / 2\nu = \frac{1}{16}$. The parameter c_0 is fixed by a rather general argument which relates the surface tension and the (exponential fall-off) correlation length in $d = 2$ (Bruce 1984) and which yields $c_0 = 2$. The remaining parameter in equation (4.14), T_c, has no direct counterpart in $d = 2$: it cannot be identified with any unambiguously defined measure of the critical temperature. In assigning its value we thus have two choices. Firstly we can, of course, resort to the $d - 1$ expansion which yields $T_c = \varepsilon + O(\varepsilon^2)$. Alternatively we can choose to assign a value so that the droplet theory correctly reproduces some other universal characteristic of the $d = 2$ Ising model. We have adopted the latter strategy: the specific universal observable to which we have chosen to match the droplet theory, in this way, is the first of those we now consider.

4.2.1. Two scale factor universality amplitude. Consider first the behaviour of the zero-field free energy density. According to equation (4.5a) (in $d = 2$)

$$f(h = 0, L_0, \xi) = \pi^{-1} \xi^{-2} \int_{L_0/\xi}^{\infty} \mathrm{d}z \, z^{-3} \tilde{\psi}(z). \tag{4.15}$$

Now equation (4.14) shows that $\tilde{\psi}(z)$ has a simple Taylor series expansion around $z = 0$; this is a specific corollary of the fact that $\nu = 1$ in $d = 2$ (cf the form of the general expansion of $\tilde{\psi}$, noted in equation (I4.4c)). Thus writing

$$\tilde{\psi}(z) = \sum_n \psi_n z^n \tag{4.16}$$

we find immediately from (4.15) that the zero-field free energy density can be written as a sum of two terms, one of which is analytic, and the other non-analytic in ξ (and thus the reduced temperature t). Explicitly the non-analytic piece has the form

$$f_s = \psi_2 \pi^{-1} \xi^{-2} \ln \xi. \tag{4.17}$$

Noting that, in veiw of the defining relation (3.8), this result is expressed in units of $-k_B T_c$, we see that the singular part of the specific heat capacity (in units of k_B) is

$$C_s = \partial^2 f_s / \partial t^2 \approx -2\psi_2 \ln t / [\pi(\xi t)^2]. \tag{4.18}$$

We may thus identify the dimensionless parameter

$$X = -(t\xi)^2 C_s / \ln t \tag{4.19}$$

as

$$X = \frac{2\psi_2}{\pi} = \frac{2\psi_0}{\pi} \frac{1}{4}\left(\frac{3 T_c^2}{8} - 3\pi T_c + 2\pi^2\right). \tag{4.20}$$

The universality of this parameter, implied by the universality claimed for the droplet population function $\tilde{\psi}$, manifests the universal link between thermodynamic and correlation length scales, originally advanced by Stauffer *et al* (1972). For the $d = 2$ Ising model the exact result, noted by these authors, can be written in the form

$$X = 1/8\pi = 2\psi_0/\pi \tag{4.21}$$

where we have again made the identification (4.6b). We have also (in contrast to Stauffer *et al*) adopted the convention (consistent with the assignment $c_0 = 2$, implicit in (4.20)) that the correlation length ξ appearing in (4.19) be taken as the exponential fall-off correlation length (rather than the second moment correlation length).

The value of X implied by equation (4.20) depends sensitively upon the value assigned to the parameter T_c. Accord with the exact result (4.21) is obtained with $T_c = 1.8$. This is the value we shall adopt throughout the remaining numerical studies reported below.

4.2.2. The essential singularity amplitude. We now return briefly to consider the implications of equation (4.13b), in the light of the parametrisation (4.14). The data of Baker and Kim (1980) do not allow us to check fully the scaling properties of the coefficient B prescribed by this result. By utilising their data (at the higher of the two temperatures which they study) one may however infer from (4.13b) a value for the parameter $\psi^{(\infty)}$: this matching procedure yields $\psi^{(\infty)} \sim 0.12$. This result is subject to substantial (but not readily quantifiable) uncertainties since the magnitude of the prefactor B will certainly be influenced by departures from spherical symmetry in the critical droplet; such effects are, of course, absent from equation (4.13b) which describes asymptotic (isotropic) behaviour. In any event, this inferred value of $\psi^{(\infty)}$ may be compared with that implied by the parametrisation (4.14). Utilising equation (2.7c) we find that $\psi^{(\infty)} = \psi_0 (T_c/c_0)^{3/2} \sim 0.053$, where we have made the T_c assignment discussed in § 4.2.1.

In view of the considerable uncertainties inherent in the two estimates we feel that the comparison is not discouraging.

4.2.3. The equation of state. Finally we consider the equation of state which is prescribed by the differential equation following from equations (3.10*b*) and (4.1):

$$\frac{\partial Q(h, L_0, \xi)}{\partial L_0} = -2L_0^{-1}\tilde{\psi}\left(\frac{L_0}{\xi}\right)\left(\sinh[2hv(L_0)] - \cosh[2hv(L_0)]Q(h, L_0, \xi)\right.$$

$$\left. - [2hv(L_0)]^{-1}\sinh[2hv(L_0)]\frac{\partial Q(h, L_0, \xi)}{\partial L_0}\right). \tag{4.22a}$$

The boundary condition for this equation is

$$Q(h, L_0, \xi) = 1, \qquad L_0 = L, \tag{4.22b}$$

where L is a length scale large compared with ξ. With $\tilde{\psi}(z)$, and the parameters it contains, prescribed as we have indicated, we have integrated equation (4.22*a*) at fixed ξ and for a range of $h(>0)$ from $L_0 = L \gg \xi$ (in practice $L/\xi \sim 17$), into the critical region characterised by values of L_0 small compared with ξ. The integration procedure is straightforward except for the use of one approximation which merits explicit comment since it is a point to which we shall subsequently have occasion to return. The approximation in question is motivated by the form of the solution to equation (4.22*a*) yielded by iteration in powers of $\tilde{\psi}$. To leading non-trivial order we find

$$\frac{\partial Q(h, L_0, \xi)}{\partial L_0} = -2L_0^{-1}\tilde{\psi}\left(\frac{L_0}{\xi}\right)\exp[-2hv(L_0)][1 + O(\tilde{\psi})]. \tag{4.23}$$

The form of this result suggests that we impose on (4.22*a*) the additional boundary condition

$$\frac{\partial Q(h, L_0, \xi)}{\partial L_0} = 0, \qquad h > h_c, \tag{4.24}$$

where $h_c \equiv h_c(L_0, \xi)$ is such that

$$\tilde{\psi}(L_0/\xi)\exp[-2h_c v(L_0)] = 10^{-k}. \tag{4.25}$$

The parameter k (in effect the cut-off field h_c) has to be chosen to be sufficiently *large* that the errors introduced by the truncation (4.24) are small (on the scale of the desired accuracy) and yet sufficiently *small* that we avoid overflow in the hyperbolic function (in 4.22*a*) at large fields. In practice we found it adequate to take $k = 4$. (See, however, the further discussion of this choice in § 6.)

Implementing this procedure yields confirmation that the equation of state does have the asymptotic $(L_0 \gg \xi)$ scaling form implied by equation (4.8*b*), namely

$$Q(h, L_0, \xi) = (\xi/L_0)^{-\beta/\nu}\tilde{Q}(h\xi^{d-\beta/\nu}L_0^{\beta/\nu}) \tag{4.26}$$

where the dependence upon the (arbitrary) inner length scale L_0 is displayed explicitly. This length in effect determines the *single* non-universal scale characteristic of a scaling theory in which the temperature dependence is expressed through the correlation length. The function $\tilde{Q}(y)$ itself is universal: its form, as prescribed by the droplet theory, is displayed in figure 1.

To test this prediction we have utilised the results of series expansion studies of the square lattice Ising model (Gaunt and Domb 1970) which exist as Padé
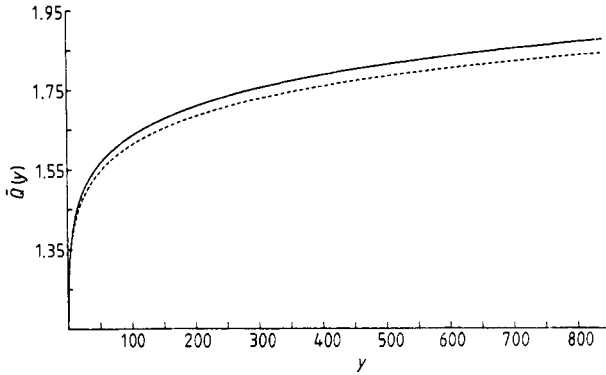
Figure 1. Plots of the function $\tilde{Q}(y)$ as determined by the droplet model (broken curve) and derived from series expansion studies of the square lattice Ising model (full curve).

approximants to the function $g(x)$ appearing in the equation of state written in the form

$$h = Q_{\mathrm{I}}^{\delta} g(t Q_{\mathrm{I}}^{-1/\beta}) \tag{4.27}$$

where Q_{I} is the Ising order parameter and t is the reduced temperature. This equation can be inverted numerically to yield $Q_{\mathrm{I}} = Q_{\mathrm{I}}(h, \xi)$. The result may be written in the form

$$Q_{\mathrm{I}}(h, \xi) = (\xi/L_0)^{-\beta/\nu} \tilde{Q}_{\mathrm{I}}(h\xi^{d-\beta/\nu} L_0^{\beta/\nu}). \tag{4.28}$$

We remark that, to satisfy the conventions implicit in equation (4.26), one requires that the field h and the order parameter Q_{I} in (4.28) be conjugate in the sense of equation (3.10b) and that the units of the correlation length ξ be those implicit in the free energy *density*: thus with Q_{I} the magnetisation per spin, h is the reduced field and ξ the correlation length in units of the lattice spacing.

The function $\tilde{Q}_{\mathrm{I}}$ in equation (4.28) is uniquely defined when the arbitrary scale parameter L_0 is fixed. We choose to prescribe this parameter by the requirement that

$$\tilde{Q}_{\mathrm{I}}(0) = \tilde{Q}(0) \tag{4.29a}$$

which is satisfied by

$$L_0 = 0.4010. \tag{4.29b}$$

The function $\tilde{Q}_{\mathrm{I}}(y)$ thus defined is also displayed in figure 1. Over the range shown it differs from our droplet calculation by no more than 2%. This level of agreement seems encouraging. Two caveats are, however, in order.

Firstly it should be recalled that, in practice, our theory is not parameter free: the principal indices β and ν, and the constant T_c (all, in principle, prescribed within the $d = 1 + \varepsilon$ expansion) have been tuned to known behaviour in $d = 2$. Nevertheless, since we have not directly fitted our theory to characteristics of the equation of state itself (cf the purely phenomenological droplet theory of Reatto and Rastelli (1972)), the accord displayed in figure 1 does appear significant.

Secondly, and more seriously, we believe that our mode of numerical solution of equation (4.22a) (specifically, our use of the auxiliary boundary condition (4.24)) actually camouflages a breakdown of the theory at large values of the scaling variable $h\xi^{d-\beta/\nu}$. We shall return to discuss this point in § 6, where we assess the general trustworthiness of the theory.

5. The droplet number distribution function

We now examine the function describing the distribution of droplets over scale sizes. The zero-field form of this function was considered in I. Like the free energy (to which, in zero field, the distribution function is simply related) the distribution is considerably more complex in the presence of an ordering field. Its form may, however, be prescribed by a straightforward extension of the renormalisation group arguments developed in § 3.

We denote by $n_\alpha(R, h, L_0, \xi)$ the mean number density of droplets of phase α with scale sizes in the range $R \to R + dR$. Then formally

$$n_\alpha(R, h, L_0, \xi) = V^{-1} Z^{-1}(h, L, L_0, \xi) \sum_{\{c(L, L_0)\}} \exp\{-\mathcal{H}[c(L, L_0)]\} \Delta_\alpha[R; c(L, L_0)]$$

$$= V^{-1} \langle \Delta_\alpha(R) \rangle_{L_0} \tag{5.1}$$

where $\Delta_\alpha[R; c(L, L_0)] = (dR)^{-1}$ (or 0) according to whether the configuration $c(L, L_0)$ contains 1 (or 0) droplet(s) of phase α, with scale size $R \to R + dR$. Separating out the contribution to the configurational sum (5.1) made by the small droplets (as in equation (3.2)) we write

$$n_\alpha(R, h, L_0, \xi) = V^{-1} Z^{-1}(h, L, L_0, \xi) \sum_{\{c(L, L_0+dL_0)\}} \sum_{\{c(L_0+dL_0, L_0)\}^*}$$

$$\times \exp\{-\mathcal{H}[c(L, L_0+dL_0)] - \mathcal{H}[c(L_0+dL_0, L_0)]\} \Delta_\alpha[R; c(L, L_0)]. \tag{5.2}$$

Now suppose first of all that the droplet scale size of interest, R, lies above the range $L_0 \to L_0 + dL_0$. Then we may make the replacement

$$\Delta_\alpha[R, c(L, L_0)] = \Delta_\alpha[R; c(L, L_0+dL_0)] \tag{5.3}$$

in the configurational sum. The partial trace over the smaller droplets may then be performed to yield (cf equation (3.4a))

$$n_\alpha(R, h, L_0, \xi) = V^{-1} Z^{-1}(h, L, L_0, \xi) \sum_{\{c(L, L_0+dL_0)\}} \exp\{-\mathcal{H}[c(L, L_0+dL_0)]\}$$

$$\times \Delta_\alpha[R; c(L, L_0+dL_0)]\{1 + V_1[c(L, L_0+dL_0)] v^{-1}(L_0)$$

$$\times \exp[-2hv(L_0)] \psi(L_0, \xi) \, dL_0$$

$$+ V_2[c(L, L_0+dL_0)] v^{-1}(L_0) \exp[2hv(L_0)] \psi(L_0, \xi) \, dL_0\} \tag{5.4a}$$

or (cf equations (3.4a) and (3.4b))

$$\partial n_\alpha(R, h, L_0, \xi)/\partial L_0 = -V^{-1} v^{-1}(L_0) \psi(L_0, \xi)\{[\langle \Delta_\alpha(R) V_1 \rangle_{L_0} - \langle \Delta_\alpha(R) \rangle_{L_0}] \exp[-2hv(L_0)]$$

$$+ [\langle \Delta_\alpha(R) V_2 \rangle_{L_0} - \langle \Delta_\alpha(R) \rangle_{L_0} \langle V_2 \rangle_{L_0} \langle V_1 \rangle_{L_0}] \exp[2hv(L_0)]\}. \tag{5.4b}$$

Utilising the trivial identity (3.5b) and recognising that (in view of (3.5a))

$$V^{-1}[\langle \Delta_\alpha(R)(V_1 - V_2) \rangle_{L_0} - \langle \Delta_\alpha(R) \rangle_{L_0} \langle V_1 - V_2 \rangle_{L_0}] = (\partial/\partial h) n_\alpha(R, h, L_0, \xi) \tag{5.5}$$

we find that (5.4b) may be recast in the simple form (cf equation (3.6))

$$\frac{\partial n_\alpha(R, h, L_0, \xi)}{\partial L_0} = v^{-1}(L_0) \psi(L_0, \xi) \sinh[2hv(L_0)] \frac{\partial n_\alpha(R, h, L_0, \xi)}{\partial h}. \tag{5.6}$$

Now consider the limiting case where the droplet scale size of interest (more precisely, the range $R \to R + dR$) coincides with the infrared cut-off (strictly, the range $L_0 \to L_0 +$

dL_0). Then the replacement (5.3) is no longer valid; instead the number operator $\Delta_\alpha(R)$ must be incorporated into the partial trace over the 'smallest' droplets so that equation (5.4a) is replaced by

$$n_\alpha(R, h, R, \xi) = V^{-1} Z^{-1}(h, L, R, \xi) \sum_{\{c(L, R+dR)\}} \exp\{-\mathcal{H}[c(L, R+dR)]\}$$

$$\times v^{-1}(R)\psi(R, \xi) V_{\bar{\alpha}}[c(L, R+dR)] \exp[-(-1)^\alpha 2hv(R)]$$

$$\equiv \langle V_{\bar{\alpha}}/V\rangle_R v^{-1}(R)\psi(R, \xi) \exp[-(-1)^\alpha 2hv(R)] \tag{5.7}$$

where $V_{\bar{\alpha}} \equiv V - V_\alpha$ is the volume available for droplets of phase $\bar{\alpha}$. Recalling equation (3.10b) we then obtain

$$n_\alpha(R, h, R, \xi) = \tfrac{1}{2}[1 + (-1)^\alpha Q(h, R, \xi)]v^{-1}(R)\psi(R, \xi) \exp[-(-1)^\alpha 2hv(R)] \tag{5.8}$$

which then prescribes the boundary conditions for the differential equation (5.6). The latter equation is very similar to that satisfied by the free energy density (equation (4.1)) and may be treated in a similar fashion. Specifically, we write the expansion

$$n_\alpha(R, h, L_0, \xi) = \sum_{k=0}^\infty n_{\alpha,k}(R, L_0, \xi) h^k/k! \tag{5.9}$$

and make the scaling ansatz (cf equation (4.3a))

$$n_{\alpha,k}(R, L_0, \xi) = \xi^{-(d+1)}[\xi^d \tilde{Q}(L_0/\xi)]^k \tilde{n}_{\alpha,k}(R/\xi, L_0/\xi) \tag{5.10}$$

which we find is vindicated by the form of equations (5.6) and (5.8), the coefficients $\tilde{n}_{\alpha,k}$ satisfying the equation (cf equation (4.4a))

$$\frac{\partial \tilde{n}_{\alpha,k}(y, z)}{\partial z} = 2\tilde{\psi}(z)z^{-1}k! \sum_{0 < l < k/2} \frac{\tilde{Q}^{-2l}(z)\tilde{n}_{\alpha,k-2l}(y, z)[2v(z)]^{2l}}{\Gamma(k-2l)\Gamma(2l)}. \tag{5.11}$$

The functions $\tilde{n}_{\alpha,k}(y, z)$ prescribed by this equation (together with the boundary conditions on $\tilde{n}_{\alpha,k}(y, y)$ following from equation (5.8)) are finite in the $z \to 0$ (scaling) limit. Thus (cf equation (4.8a))

$$n_\alpha(R, h, L_0, \xi) \approx \xi^{-(d+1)} \sum_{k=0}^\infty \frac{1}{k!} \left[h\xi^d \tilde{Q}\left(\frac{L_0}{\xi}\right) \right]^k \tilde{n}_{\alpha,k}\left(\frac{R}{\xi}, 0\right) \tag{5.12a}$$

which implies the scaling form

$$n_\alpha(R, h, L_0, \xi) \approx R^{-(d+1)} \tilde{n}_\alpha(hR^{d-\beta/\nu}L_0^{\beta/\nu}, R/\xi) \tag{5.12b}$$

where we have made use of equations (4.6a, b).

This result prompts a number of comments. Firstly, it is interesting to contrast the role played by the inner length scale (minimum droplet size) L_0 in the zero-field and finite-field distributions. In zero field the number distributions depend on L_0 only in the trivial sense that $n_\alpha(R, h = 0, L_0, \xi)$ is non-zero only for $L_0 < R$. In finite field the dependence upon L_0 is more subtle: the decoration of a droplet of scale size R with smaller droplets renormalises the field energy associated with its presence, and thus affects the probability with which it will occur. In effect, then, the field couples to the net magnetic moment of the decorated droplet which (to within an L_0-independent scaling function) varies as $R^d(R/L_0)^{-\beta/\nu}$.

Secondly we observe that, as a corollary of this effect, the presence of a field destroys the simple relationship that exists, in zero field, between the free energy density and the droplet distribution functions. For $h = 0$ we may recall from I or see immediately from equations (3.10*a*), (5.6) and (5.8) that the (dimensionless) free energy density is simply prescribed by the total number of droplets:

$$f(h = 0, L_0, \xi) = \sum_{\alpha=1}^{2} \int_{L_0}^{\infty} \mathrm{d}R \, n_\alpha(R, h = 0, L_0, \xi). \tag{5.13a}$$

On the other hand, in the finite field one discovers that the analogous relationship must be written in the form (cf equations (3.10*a*) and (5.8))

$$f(h, L_0, \xi) - h = \sum_{\alpha=1}^{2} \int_{L_0}^{\infty} \mathrm{d}R \, n_\alpha(R, h, R, \xi). \tag{5.13b}$$

The right-hand side is *not* the integral over the true droplet number distributions but, rather, the distributions which would be appropriate if the dressing of the field energy by droplet nesting were suppressed: replacing the true inner length scale L_0 by the running scale R has precisely this effect. (Alternatively one may employ the distributions with the true inner length scale L_0 but with the field h conjugate to droplets of scale size R replaced by a renormalised field $h(R) \equiv h\tilde{Q}(R/\xi)/\tilde{Q}(L_0/\xi)$; such a representation is correct to within terms $O(h^3\psi_0^2)$.) The absence of a simple relationship between the free energy and the droplet distributions, in finite field, is actually less surprising than the existence of such a relationship in zero field. A relationship of this kind is a hallmark of a theory of *non-interacting* droplets. In fact our theory actually includes droplet interactions to the extent that it accounts for excluded volume effects. As discussed in I (§ 5.2) the effects of excluded volume in the calculation of the free energy density are precisely offset by the effects of droplet nesting so that the result (5.13*a*) characteristic of the non-interacting limit is recovered. However, in the presence of a field this cancellation does not survive, and neither does the naive analogue of (5.13*a*).

Finally, returning briefly to clarify a point discussed in I (§ 6) we may usefully set the scaling form (5.12*b*) alongside the result of purely phenomenological arguments. According to Binder (1976, equation (2.5))

$$n(l, h, \xi) = e^{-(2+y/\delta)} \tilde{n}(hl^y, l^{y\nu/\beta\delta}/\xi) \tag{5.14}$$

gives the number density of droplets ('clusters') of the phase ('black', say, in the colour metaphor of I) opposite to that which is thermodynamically favoured ('white') and having volume ('number of spins') l. The relationship can be brought into correspondence with equation (5.12*b*) if one sets the index y to unity and identifies, to within a scaling function, $l = R^{d-\beta/\nu}$. However, thus identified, the variable l does not prescribe the true volume associated with a black droplet of scale size R (which actually scales as $R^{d-\beta/2\nu}$) but rather the total black volume within its boundaries *including* the contributions of nested black droplets. Accordingly the phenomenological order parameter sum rule (Binder 1976, equation (23*a*)) overcounts the contributions of black droplets; it appears to be consistent with the anticipated critical behaviour of the order parameter only because of a compensating failure to capture a subtle sum rule obeyed by the distribution function (cf I, § 6).

6. An assessment of the theory

In this paper we have considered a droplet theory of phase transitions originally formulated as a model of the configurational physics of Ising systems on the line of two-phase coexistence, and developed it to include the effects of an ordering field. We now turn to assess the reliability and utility of this extended theory.

Our principal task in this regard is to establish to what extent, and in what sense, the theory is trustworthy. In addressing this question we must note immediately that, in a strict sense, the domain of validity of the theory is certainly limited by the $d = 1 + \varepsilon$ expansion on which it is based: at present we have little confidence that such expansion methods can be developed to yield, in $d = 2$, predictions for universal parameters as successful as the $4 - d$ expansion results have proved in $d = 3$. Nevertheless, in the course of this paper (notably in § 4) we have seen indications that the structure of the theory may have a degree of validity (or, at least, utility) which transcends the formal small-ε limitation. We believe, however, that even though the latter limitations may be at least partially evaded by judicious reparametrisation of the theory, there remains inherent in the theory a further limitation which our arguments have thus far failed to expose. The limitation in question reflects the range of the parameters h and t (or, more naturally, h and ξ) in which the approximations implicit in the theory are even formally justifiable.

To explore this point let us recall the key elements of the picture underlying our calculations. The theory is constructed on the assumption that the coarse-grained configurations of Ising systems can be represented by patterns of droplets nested in such a way that the droplet boundaries are *dilute*. Loosely, the essential idea is that the *largest* droplet to be found housed within a droplet of some arbitrary scale size, R, will itself typically have a scale size *small* compared with R; this idea finds more precise expression in the claim that the *fractal* dimension of a droplet lies close to its *Euclidean* dimension. This assumption is embodied in the use of the *single*-droplet partition function (2.12) in the anslysis leading to the *multi*-droplet partition function (3.6). It is thus at the heart of the theory. On the coexistence curve the validity of the assumption is assured by the smallness of the droplet concentration function $\tilde{\psi}(z)$, as discussed fully in I, § 5.4. However, away from the coexistence curve $\tilde{\psi}(z)$ is no longer solely responsible for controlling the droplet population: in finite field the droplet concentration is enhanced or suppressed, according to phase, by a factor $\exp[2hv(R)]$. It is the balance between this volume factor and the surface tension factor $\psi(R/\xi)$ which determines the limits of validity of the theory. Consider the population of first-level droplets—those droplets of the unfavoured phase which are not nested within larger droplets: these droplets are the sole configurational ingredients of classical droplet theories. Since the population of these droplets is suppressed by the ordering field our treatment of them is correct (or at least as correct as it is in zero field). In contrast, however, the population of the droplets of the field-favoured phase nested immediately within the first-level droplets will be *enhanced* by the field to a degree which reflects both the magnitude of the field and the size of the nesting droplet, the latter being bounded by the size of the first-level droplet in which it is housed. The implication is clear: for large enough fields, or large enough first-level droplets, the population of nested droplets cannot be regarded as dilute. In effect we envisage that, as the field is increased, the favoured droplets (nested in any given first-level droplet) grow in number and ultimately percolate through the droplet in which they are housed. This picture is very similar to that advanced by Klein (1981) in a discussion of the

structure of the configurations responsible for spinodal decomposition. To assess its implications for the finite-field theory we must examine the extent of the contributions arising from the configurations which the theory treats incorrectly. There are two regimes to consider.

Consider first the regime where h is small. By 'small' we mean, in the first instance, that $h\xi^d \ll 1$. In this regime the droplets of scale sizes R small compared with ξ carry exponential field-weighting factors which are of order unity: for these the dilute approximation is still acceptable. On the other hand, for droplets with scale sizes R larger than ξ the field-weighting factors are no longer of order unity and the dilute approximation fails for droplets of the favoured phase. However, these droplets are housed within a suppressed population of larger first-level droplets such that the overall field-weighting factor associated with a nested droplet configuration is *less* than unity. Thus the contributions of the non-dilute droplets to a thermodynamic average are of order $\tilde{\psi}^2$ and, for $R > \xi$, these contributions, and thus the errors which they introduce, are negligible.

Consider now, however, the regime where $h\xi^d$ is large. There will then exist a range of first-level droplets with scale sizes satisfying $h^{-1/d} < R < \xi$ which house nested droplets that are *not* dilute, and whose contributions are *not* suppressed by an exponentially small surface tension factor.

This analysis suggests that the theory can be trusted only in the regime $h\xi^d \ll 1$. In fact it is clear, both from the formal scaling structure of the theory and on physical grounds, that the effect of small droplets will be to produce the renormalised criterion $h\xi^{d-\beta/\nu}L_0^{\beta/\nu} \ll 1$. This condition presupposes a temperature *below* the critical point. *Above* the critical point there are no surface tension effects to control the population of large droplets (cf the flow of the renormalised temperature discussed in I, § 8) and there is no region of the phase diagram where the droplet theory is controllable. We conclude that the theory developed here is restricted in its applicability to the region of the field–temperature plane close to the coexistence curve.

We have not succeeded in putting these heuristic arguments on a secure analytic foundation. It is, however, possible both to detect the breakdown of the theory and to assess (reassuringly, it turns out) its severity by considering the form which the equation of state assumes precisely at the critical point. If, as suggested above, the theory breaks down at a finite value of the scaling variable $y = h\xi^{d-\beta/\nu}L_0^{\beta/\nu}$ its domain of validity in h should shrink to zero at the critical point. Indeed, there is a sense in which this is true. Specifically, in the $\xi \to \infty$ limit one may deduce from equation (4.26) that the order parameter Q assumes the form $Q = Q_c(x)$ with $x = hv(L_0)$. This form is consistent with (4.22a), from which one may readily obtain a differential equation satisfied by Q_c. One then finds that, although this equation is consistent with the anticipated small-x behaviour ($Q_c(x) \sim x^{1/\delta}$ with δ the exponent implied by equation (4.6b) and hyperscaling) the differential equation possesses a singularity at a finite value of $x \equiv x_s$ (of order ε^{-1}) which makes it impossible to implement the physically sensible boundary condition $\lim_{x \to \infty} Q_c(x) = 1$. In principle, then, the droplet theory form of the equation of state does indeed fail at the critical point. There is, however, some reason to believe that in practice this failure is (or may be made) less severe than it first appears. In particular, we have investigated the differential equation for Q_c in $d = 2$ when the singularity occurs at $x_s = 2.55 \ldots$. If we suppose that the mapping of the square lattice Ising model onto the droplet model prescribed by the assignment (4.29b) holds, at least in some approximate fashion, even outside the critical region we find that this value of x_s corresponds to an order parameter which is extremely

close to saturation. With this observation as motivation (if not justification), we have investigated the consequences of imposing on the differential equation for Q_c the boundary condition $Q_c(x_0) = 1$ where $x_0 < x_s$, thus evading the singularity at x_s. Integrating the differential equation numerically then yields, for small h, the result $Q_c \approx 1.18$ $(hL_0^2)^{1/\delta}$. The amplitude of the power law is only very weakly dependent on the cut-off value x_0: the stated value is appropriate in the range $1 < x_0 < 2.54$. This result is in remarkably close agreement with the known behaviour of the square lattice Ising model (Gaunt and Domb 1970) for which, with the identification (4.29b), $Q_1(h, \xi \to \infty) \approx 1.196(hL_0^2)^{1/\delta}$. It is also consistent with the limiting behaviour of $Q(h, L_0, \xi)$ implied by the large-y extrapolation of our numerical results for the function $\tilde{Q}(y)$ (figure 1), which yields $Q(h, L_0, \xi \ll L_0) \approx (1.19 \pm 0.02)(hL_0^2)^{1/\delta}$.

Two comments are appropriate here. Firstly, the accord between our two separate calculations of the asymptotic finite-field critical behaviour reflects the fact that both calculations effectively discard the contributions associated with the large droplets which the theory does not control. In the critical point calculation discussed above, these contributions are eliminated by our choice of the boundary value x_0 from which the differential equation for Q_c is integrated; in the calculation of $\tilde{Q}(y)$ discussed in § 4.2 a similar approximation is made in our imposition of the auxiliary boundary condition (4.25). Indeed, if the value of the cut-off parameter k (equation (4.25)) is increased much beyond that prescribed in the text, the numerical integration of (4.22a) rapidly breaks down. Secondly, it seems reasonable to infer from the success with which the two calculations do in fact capture the 'observed' (series expansion) behaviour in the $d = 2$ lattice Ising model that the discarded contributions arising from these troublesome droplets (specifically, those with scale sizes in the range $(h^{\nu/\beta}L_0)^{-1/\delta} < R < \xi$) are truly small. Presumably the contributions which such large-scale structures make to the order parameter and its derivatives is strongly suppressed by virtue of their highly ramified character (Klein 1981) which is realised (qualitatively but not controllably) within our framework in the high population of the field-favoured droplets which they will accommodate.

Finally we offer a rather subjective view of the status and utility of the framework we have developed. It is clear that our calculations by no means constitute a complete first principles theory. We have already remarked on the need to circumvent the numerical limitations of the $\varepsilon = d - 1$ expansion by somewhat *ad hoc* procedures. We should also recall that, as noted in I (see also Wallace and Zia (1979)), our calculations do not capture the effects of interfacial overhangs which would be realised within our framework in partial droplet overlap effects (exponentially small in $\varepsilon = d - 1$).

On the other hand we believe that the present work, together with that of I, constitutes considerably more than a pure phenomenology of the Ising universality class. It offers an explicit theory of surface tension and the manner in which its effective strength evolves with the scale size of the interface in question. It provides a framework within which the behaviour of thermodynamic observables is explictly linked to the characteristics of the effective surface tension, manifested in our droplet population function. It provides qualitative insights (cf our discussion of the structure of the coexistence curve essential singularity). Its quantitative successes as regards the thermodynamic behaviour in $d = 2$ (cf the equation of state) seem significant even in the light of the parametrisations we have had to make. Finally its predictive power extends considerably beyond the sphere of thermodynamic quantities: the form of the droplet population function has implications for, amongst other observables, the correlation function (for which exact results exist in $d = 2$: McCoy and Wu (1973)) and the

magnetisation distribution (which is accessible to computer simulation: Binder (1982)). These are the subject of continuing study.

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