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Renormalization group flow of the stiffness matrix–free-energy relation

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Abstract. In order to satisfy exact sum-rule requirements for correlation function structure at complete wetting, a two-field Hamiltonian $H_I^{(2)}[l_1, l_2]$, modelling the coupling of order-parameter fluctuations near the wall and unbinding interface, has been introduced. The model is characterized by a stiffness matrix, $\mathbf{\Sigma}(l_1, l_2)$, whose bare (unrenormalized) elements are related to the mean-field free energy. We extend previous renormalization group studies to include the position dependence of the matrix elements and derive an elegant operator relationship which shows that the flow of the cross-coupling term $\Sigma_{12}(l_1, l_2)$ parallels that of the free energy. This establishes the validity of a stiffness matrix–free-energy relation in the presence of fluctuation effects at the marginal dimension $d = 3$ for systems with short-ranged forces. We further show that an analogous relation exists for systems with long-ranged molecular interactions.

1. Introduction

Recently significant advances have been made towards solving problems related to the theory of wetting in three-dimensional systems. In particular, new effective Hamiltonian models and methods have been proposed which improve upon phenomenological (capillary-wave) approaches and clarify the connection with 'microscopic' models which have proven too difficult to analyse [1–6]. These have led to new predictions [7–11] for fluctuation effects at the marginal dimensionality $d = 3$ (for systems with short-ranged forces) which are in encouraging agreement with old and new Ising model simulation results [12, 13]. Here we further analyse the fluctuation properties of the 'two-field' model of the *complete wetting* transition which is based on a coupled Hamiltonian $H_I^{(2)}[l_1, l_2]$ characterized by a stiffness matrix $\mathbf{\Sigma}(l_1, l_2)$. We have shown elsewhere that the (bare) parameters in this model satisfy a so-called stiffness matrix–free-energy (SMFE) relation which guarantees that an exact statistical mechanical sum rule is satisfied at mean-field (MF) level [4, 5]. This requirement is not met by the capillary-wave (CW) and Fisher–Jin (FJ) [1, 2] models (even with the latter's position-dependent stiffness coefficient) which do not allow for coupling in order-parameter fluctuations near the wall and unbinding interface.

In this paper we extend the renormalization group (RG) analysis of $H_I^{(2)}$ (presented in [7]) to include the position dependence of the stiffness matrix elements, and show that the SMFE relation retains its form under RG flow. Indeed beyond MF the SMFE relation takes the form of an elegant operator identity which ensures that the RG flow of the leading-order matrix element $\Sigma_{12}(l_1, l_2)$ parallels that of the free energy. As a consequence the sum rule is obeyed even in the presence of fluctuation effects which alter the value of critical

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amplitudes at the upper critical dimension $d = 3$. We believe that this is a significant success of the two-field theory and underlines the importance of allowing for coupling effects in the development of a self-consistent thermodynamic theory of complete wetting. To begin we first recall the derivation of the bare SMFE relation and comment on its interpretation.

2. The mean-field SMFE relation

2.1. Microscopics

A suitable starting point for the study of phase equilibria in semi-infinite systems with short-ranged forces is the Landau–Ginzburg–Wilson (LGW) Hamiltonian [14]

$$H_{\mathrm{LGW}}[m] = \int \mathrm{d}\boldsymbol{y} \int_0^\infty \mathrm{d}z\ \mathcal{L}(m, \boldsymbol{\nabla} m) \tag{2.1}$$

with

$$\mathcal{L}(m, \boldsymbol{\nabla} m) = \frac{K}{2}(\nabla m)^2 + \phi(m(\boldsymbol{r})) + \delta(z)\phi_1(m(\boldsymbol{r})) \tag{2.2}$$

where z measures the distance normal to the (planar) wall and $\boldsymbol{y}$ is the displacement vector along it. Here $\phi(m)$ and $\phi_1(m)$ are bulk and surface free-energy densities respectively; $\phi(m)$ has (for subcritical temperatures $T < T_c$) a standard double-well form showing bulk two-phase coexistence between phases β (with $m_\beta > 0$) and α (with $m_\alpha < 0$) in zero bulk field, $h = 0$. We shall assume that ϕ_1 is given by the standard expression $\phi_1(m) = -(h_1 m + g m^2/2)$ with h_1 the surface field and g the enhancement. The MF phase diagram of (2.1) is well understood [15] and exhibits first-order, tricritical and complete wetting transitions. Although the model cannot be solved exactly it is believed that these features survive beyond the MF level. Here we concentrate on the case of the complete wetting transition whereby the wall–α interface is completely wet by the β-phase in the limit of bulk two-phase coexistence $h \to 0^-$ (say), i.e. we assume that we are above any wetting temperature T_W.

In MF approximation, minimization of (2.1) with respect to magnetization configurations determines the (MF) equilibrium profile $m(z)$ and leads to the well known expression for the excess free energy per unit area (or surface tension)

$$\Sigma = K \int_0^\infty \mathrm{d}z\ m'(z)^2 + \phi_1(m_1) \tag{2.3}$$

with m_1 the surface magnetization $m_1 \equiv m(z = 0)$ and where we use primes to denote differentiation with respect to the argument. To evaluate this we have to specify what the bulk phase is, i.e. whether we consider the wall–α or wall–β phase interface. In the approach to a wetting transition in which the β-phase wets the wall–α interface it is customary to write the surface tension of the wall–α phase interface as

$$\Sigma_{w\alpha} = \Sigma_{w\beta} + \Sigma_{\alpha\beta} + f_{\mathrm{sing}} \tag{2.4}$$

where the three terms on the right-hand side correspond to the wall–β-phase interfacial tension, free $\alpha\beta$-surface tension, and singular contribution to the excess free energy respectively. The singularity in the free energy at complete wetting, written as $f_{\mathrm{sing}} \sim |h|^{2-\alpha_s^{co}}$ determines the thickness of the wetting layer $l \sim |h|^{-\beta_s^{co}}$ and transverse correlation length $\xi_\parallel \sim |h|^{-\nu_\parallel}$ via the exponent relations $1 - \alpha_s^{co} = -\beta_s^{co}$ and $\alpha_s^{co} = 2\nu_\parallel^{co}$. In MF approximation, explicit calculation yields

$$f_{\mathrm{sing}} \sim h \ln |h| \tag{2.5}$$

corresponding to $\alpha_s^{co} = 1$ consistent with $\beta_s^{co} = \mathrm{O}(\ln)$ and $\nu_\parallel^{co} = 1/2$.

The order-parameter correlation function $\mathcal{G}(\boldsymbol{r}_1, \boldsymbol{r}_2) \equiv \langle m(\boldsymbol{r}_1)m(\boldsymbol{r}_2)\rangle_c$ can also be calculated in MF theory. For this it is convenient to introduce the transverse Fourier transform

$$\mathcal{G}(z_1, z_2; Q) = \int \mathrm{d}\boldsymbol{y}_{12}\ \mathrm{e}^{\mathrm{i}\boldsymbol{Q}\cdot\boldsymbol{y}_{12}}\mathcal{G}(\boldsymbol{r}_1, \boldsymbol{r}_2) \tag{2.6}$$

and its moment expansion

$$\mathcal{G}(z_1, z_2; Q) = \sum_{n=0}^{\infty} Q^{2n}\mathcal{G}_{2n}(z_1, z_2). \tag{2.7}$$

Two functional derivatives of (2.1) with respect to $m(\boldsymbol{r})$ define the direct correlation function $C(\boldsymbol{r}_1, \boldsymbol{r}_2)$ (in MF approximation) which may be substituted into the Ornstein–Zernike integral equation to yield

$$(-K\partial_{z_1}^2 + KQ^2 + \phi''(m(z_1)))\mathcal{G}(z_1, z_2; Q) = \delta(z_1 - z_2) \tag{2.8}$$

where we have set $k_BT = 1$ for simplicity. Solving this equation we find that the properties of the second moment at the wall are particularly interesting, and explicit calculation shows (see for example [16])

$$\mathcal{G}_2(0, 0) = \frac{-(\Sigma - \phi_1(m_1))}{(cm'(0) - m''(0))^2} \tag{2.9}$$

for both $w\alpha$- and $w\beta$-interfaces. The denominator is well behaved in the approach to the complete wetting transition, and attention focuses on the numerator which shows that the correlations at the wall for the wall–α interface 'know' about the full excess free energy. This is symptomatic of the coherence of asymptotically long-wavelength fluctuations in the wetting film, and establishes a crucial connection between correlation functions and thermodynamic singularities.

There are very good reasons for believing that the relationship (2.9) is exact beyond the MF level since a very similar equation is known to be precisely true for the case of 'drying' by a vapour phase (β) at a hard-wall–liquid ($w\alpha$-) interface (see section 4). Thus (2.9) may be considered to be an exact sum-rule requirement. The problem, then, is to construct an effective-Hamiltonian theory (based on a collective coordinate) for thermodynamic and correlation function properties valid at MF level and beyond which is consistent with this exact sum-rule requirement. This task is not as simple as it may appear because the process of integrating out degrees of freedom, essential in the derivation of effective (or low-energy) Hamiltonians, is seldom precise (or systematic). Furthermore, even if this problem is overcome, the reconstruction of order-parameter correlations from collective coordinate distributions still needs to be addressed.

2.2. Two-field theory

Traditional capillary-wave approaches (and their extensions incorporating a position-dependent stiffness coefficient), which are based on a single collective coordinate ($l(\boldsymbol{y})$ say) describing the position of the $\alpha\beta$-interface, do not satisfy the sum rule at MF level where they fail to capture the term $f_{\mathrm{sing}} \sim h\ln|h|$ appearing in the numerator of (2.9). However, progress can be made using a 'two-field' Hamiltonian

$$H_I^{(2)}[l_1, l_2] = \int \mathrm{d}\boldsymbol{y}\ \left[\frac{1}{2}\Sigma_{\mu\nu}(l_1, l_2)\boldsymbol{\nabla}l_\mu \cdot \boldsymbol{\nabla}l_\nu + W^{(2)}(l_1, l_2)\right] \tag{2.10}$$

where $l_2(\boldsymbol{y})$ ($l_1(\boldsymbol{y})$) represents a surface of fixed magnetization m_2^X (m_1^X) which unbinds from (remains bound to) the wall in the limit of complete wetting. Note that $l_1(\boldsymbol{y})$ is an interface-like collective coordinate which models order-parameter fluctuations at the wall via translations in the surface of fixed magnetization $m_1^X \approx m_1$. Such a description is appropriate for complete wetting occurring at temperatures $T \gg T_W$ where there is a significant homogeneity at the wall (for further details see [6]). The coefficients $\Sigma_{\mu\nu}(l_1, l_2)$ can be considered the elements of a stiffness matrix $\boldsymbol{\Sigma}(l_1, l_2)$ which plays an essential role in the theory. Explicit expressions for $\boldsymbol{\Sigma}$ and $W^{(2)}$ may be calculated in terms of the planar constrained profile $m_\pi^{(2)}(z; l_1, l_2)$ corresponding to the profile which minimizes (2.1) subject to the double crossing constraint $m_\pi^{(2)}(z = l_\mu; l_1, l_2) = m_\mu^X$. In particular, calculation yields

$$W^{(2)}(l_1, l_2) = \int \mathrm{d}z\ \mathcal{L}(m_\pi^{(2)}, \partial_z m_\pi^{(2)}) \tag{2.11}$$

and

$$\Sigma_{\mu\nu}(l_1, l_2) = K \int \frac{\partial m_\pi^{(2)}}{\partial l_\mu} \frac{\partial m_\pi^{(2)}}{\partial l_\nu}\, \mathrm{d}z \qquad \mu, \nu = 1, 2. \tag{2.12}$$

Minimization of the binding potential $W^{(2)}$ recovers the MF positions, z_μ say, of the surfaces of fixed magnetization m_μ^X.

Subtracting off a trivial bulk contribution to $W^{(2)}$ (i.e. terms which are independent of l_1, l_2) we can identify the MF free-energy singularity as

$$f_{\mathrm{sing}} = W^{(2)}(z_1, z_2). \tag{2.13}$$

Continuing with the formalism, an important advantage of the two-field approach is the ability to make precise connection with the MF correlation functions $\mathcal{G}(z_\mu, z_\nu; Q)$ for particle positions near the wall and unbinding interface. To see this, consider the matrix elements

$$S_{\mu\nu}(Q; z_1, z_2) \equiv \int \mathrm{d}\boldsymbol{y}\ \mathrm{e}^{\mathrm{i}\boldsymbol{Q}\cdot\boldsymbol{y}} \langle \delta l_\mu(Q)\, \delta l_\nu(Q) \rangle \tag{2.14}$$

where $\delta l_\mu = l_\mu(\boldsymbol{y}) - \langle l_\mu(\boldsymbol{y}) \rangle$, and recall that the m_μ^X are chosen such that $\langle l_\mu(\boldsymbol{y}) \rangle = z_\mu$, $\mu = 1, 2$. Hereafter we are generally considering the case where $z_1 = 0$ and z_2 corresponds to the position of the zero of the magnetization profile (so $\langle l_2 \rangle$ is a measure of the thickness of the wetting layer). The correlation functions are then determined by the relations

$$\mathcal{G}(z_\mu, z_\nu; Q) = m'(z_\mu) m'(z_\nu) S_{\mu\nu}(Q; z_1, z_2). \tag{2.15}$$

The matrix elements $S_{\mu\nu}$ may be calculated using the relationship [5]

$$S_{\mu\nu}^{-1} = \begin{pmatrix} \partial_{11}^2 & \partial_{12}^2 \\ \partial_{12}^2 & \partial_{22}^2 \end{pmatrix} W^{(2)} + \boldsymbol{\Sigma} Q^2 \tag{2.16}$$

where $\partial_{ij}^2 \equiv \partial^2 / \partial l_i\, \partial l_j$ and is evaluated at equilibrium, $l_\mu = z_\mu$. Note that in writing (2.16) we have neglected terms $\mathrm{O}(Q^4)$ associated with the rigidity matrix—these play little part in our description of long-wavelength fluctuation effects at wetting.

Calculations are made easier on noting the separable property of the binding potential

$$W^{(2)}(l_1, l_2) = U(l_1) + W(l_2 - l_1) \tag{2.17}$$

where $U(l_1)$ serves to bind the lower surface of fixed magnetization $m_1^X \approx m_1$ to the wall (with $z_1 \approx 0$). The relative term $W(l_2 - l_1)$ is rather similar to the expression found in older capillary-wave approaches:

$$W(l) = \bar{h} l + a(T, h_1, \ldots) \mathrm{e}^{-\kappa l} + \cdots \tag{2.18}$$

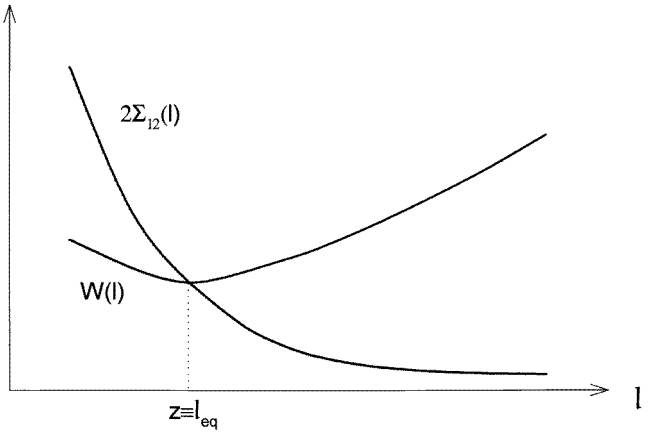

Figure 1. Diagrammatic representation of the bare SMFE relation. The functions $2\Sigma_{12}(l)$ and $W(l)$ intersect at (or very close to) the minimum of $W(l)$. The value of the functions at this extremum is equal to the singular part of the free energy. As $\bar{h}$ is reduced the locus of the binding potential minimum follows the curve of $2\Sigma_{12}(l)$.

where $\bar{h}$ is proportional to the bulk ordering field $\bar{h} \propto -h$, the coefficient a is positive (for $T > T_W$) and κ is the inverse bulk correlation length of the wetting phase. To understand the sum-rule result (2.9) for the $w\alpha$-interface we first calculate $S_{11}(Q; z_1, z_2)$ using the stiffness matrix formalism, finding

$$S_{11}(Q; z_1, z_2) = \frac{1}{U'' + Q^2[\Sigma_{11} + (\Sigma_{22} + 2\Sigma_{12})/(1 + Q^2\xi_\parallel^2)]} \tag{2.19}$$

where the transverse correlation length is identified (in standard fashion) as $\xi_\parallel^2 = \Sigma_{22}/W''$ and all quantities are evaluated at the equilibrium positions of the collective coordinates. The non-Lorentzian form of this structure factor has been elucidated at length in [16]. The numerator in the sum rule may be related to the $Q = 0$ limit of the terms in square brackets in (2.19). From (2.12) we have

$$\sum_{\mu,\nu=1}^{2} \Sigma_{\mu\nu} = K \sum_{\mu,\nu=1}^{2} \int \mathrm{d}z \left(\frac{\partial m_\pi^{(2)}}{\partial l_\mu} \frac{\partial m_\pi^{(2)}}{\partial l_\nu} \right) = K \int \mathrm{d}z \left(\frac{\partial m_\pi^{(2)}}{\partial l_1} + \frac{\partial m_\pi^{(2)}}{\partial l_2} \right)^2. \tag{2.20}$$

At equilibrium the integrand is simply the square of the total derivative with respect to z, so

$$\sum_{\mu,\nu=1}^{2} \Sigma_{\mu\nu} = K \int \mathrm{d}z \left(\frac{\mathrm{d}m}{\mathrm{d}z} \right)^2 = \Sigma_{w\alpha} - \phi_1(m_1) \tag{2.21}$$

by virtue of (2.3). This is (one version of) the SMFE relation crucial for the thermodynamic consistency of the two-field formalism. Further insight follows on calculating the position dependence of the matrix elements $\Sigma_{\mu\nu}$. Interestingly the off-diagonal element Σ_{12}, associated with the $\boldsymbol{\nabla} l_1 \cdot \boldsymbol{\nabla} l_2$ coupling, provides the leading-order decay

$$\Sigma_{12} \equiv \Sigma_{12}(l_{21}) \sim \frac{\kappa a}{2} l_{21} \mathrm{e}^{-\kappa l_{21}} + \cdots \tag{2.22}$$

where $l_{21} \equiv l_2 - l_1$, and we may approximate

$$\Sigma_{11} \approx \Sigma_{w\beta} - \phi_1(m_1) \qquad \Sigma_{22} \approx \Sigma_{\alpha\beta}. \tag{2.23}$$

Combining these explicit results with (2.21) leads to the identification

$$2\Sigma_{12}(z_{21}) = W^{(2)}(z_1, z_2) = W(z_{21}) \tag{2.24}$$

where $z_{21} = z_2 - z_1$ and we have subtracted a bulk contribution by setting $U(z_1) = 0$. This relation shows that the locus of the binding potential minimum follows the curve $2\Sigma_{12}$ as h is varied (see figure 1). Hence we arrive at the elegant two-field identity

$$2\Sigma_{12}(z_{21}) = f_{\text{sing}} \tag{2.25}$$

which we will regard as an alternative expression for the SMFE relationship. Neither (2.21) or (2.25) have any counterpart in traditional effective-Hamiltonian approaches to wetting transitions which neglect coupling between order-parameter fluctuations at the wall and unbinding interface.

Before we begin our RG analysis which will focus on the flow of the second relation (2.25) we conclude this section by making some brief remarks on the meaning of the full SMFE relation (2.21). One interpretation is in terms of the coherence of asymptotically long-wavelength fluctuations. This can be seen as follows [4]. Integrate out the 'fast' degrees of freedom of the upper field $l_2(\boldsymbol{y})$ to derive an effective Hamiltonian for the 'slow' lower field $l_1(\boldsymbol{y})$:

$$\mathrm{e}^{-H_I^{(1)}[l_1]} = \int \mathcal{D}l_2 \ \mathrm{e}^{-H_I^{(2)}[l_1, l_2]}. \tag{2.26}$$

At MF level (which is our only concern here) this functional integral may be evaluated using the saddle-point approximation

$$H_I^{(1)}[l_1] = \min_{l_2} H_I^{(2)}[l_1, l_2] = H_I^{(2)}[l_1, \bar{l}_2] \tag{2.27}$$

where $\bar{l}_2(\boldsymbol{y})$ is the distribution which minimizes $H_I^{(2)}$ for a given configuration $l_1(\boldsymbol{y})$. For asymptotically long-wavelength deviations of the lower field from a planar configuration it is straightforward to show that

$$\boldsymbol{\nabla}\bar{l}_2 = \boldsymbol{\nabla} l_1 \tag{2.28}$$

so the effective stiffness, Σ_1, of the lower field appearing in the Hamiltonian $H_I^{(1)}[l_1]$ is

$$\Sigma_1 \equiv \sum \Sigma_{\mu\nu}. \tag{2.29}$$

This neatly demonstrates how the total stiffness enters the MF expression for $\mathcal{G}_2(0, 0)$. However, with this interpretation (based on a saddle-point approximation) it is not at all obvious whether the singular form of the SMFE relation (2.25) survives if we treat the functional integral better, i.e. incorporate the roughness of the $\alpha\beta$-interface in $d = 3$. To investigate this we turn to a RG study.

3. Renormalization group analysis

The task of this section is to show that the SMFE relation retains its form under RG flow for $d = 3$. Recall that this is the marginal, or upper critical dimension, for complete wetting transitions in systems with short-ranged forces. For $d < 3$ the critical exponents take the universal values $\beta_s^{co} = (3 - d)/(d + 1)$, $\nu_\parallel = (d - 1)/(d + 1)$ and $\alpha_s^{co} = 4/(d + 1)$. At the marginal dimension itself, the critical exponents retain their MF values but the critical amplitudes are non-universal (reflecting the presence of fluctuation effects). There has been some debate on this issue recently since the two-field theory predicts that coupling effects influence the value of observable critical amplitudes leading to different results to the CW

model. Importantly, these new predictions are in agreement with Ising model simulation studies although such issues will not be of concern to us here where we focus on further developing the formalism. To this end we extend the RG analysis developed in [7] to include the position dependence of the stiffness coefficients. We shall show that this does not affect the value of the critical amplitudes but does allow discussion of the status of the SMFE relationship (beyond MF theory) not possible in previous treatments.

First, note that since the fluctuations of the lower surface are small we may reliably expand $U(l_1)$ in a Gaussian fashion about its minimum value which we set to zero without loss of generality. Hence we can write $H_I^{(2)}[l_1, l_2]$ in the form

$$H_I^{(2)}[l_1, l_2] = \int \mathrm{d}\boldsymbol{y} \left[\frac{1}{2}\Sigma_{11}(l_1, l_2)(\nabla l_1)^2 + \Sigma_{12}(l_2 - l_1)(\boldsymbol{\nabla} l_1)\cdot(\boldsymbol{\nabla} l_2) + \frac{1}{2}\Sigma_{22}(l_1, l_2)(\nabla l_2)^2 + \frac{1}{2}v_0 l_1^2 + W(l_2 - l_1)\right] \tag{3.1}$$

where we have utilized the symmetry of the stiffness matrix $\Sigma_{21} \equiv \Sigma_{12}$. It is convenient to separate the position-dependent contributions to the stiffnesses $\Sigma_{\mu\nu}$ from the constant 'free' contributions

$$\Sigma_{\mu\nu}(l_1, l_2) = \Sigma_{\mu\nu}^{\infty} + \Delta\Sigma_{\mu\nu}(l_1, l_2) \qquad \mu, \nu = 1, 2 \tag{3.2}$$

where the $\Delta\Sigma_{\mu\nu}$ decay to zero in the limit $l_2 - l_1 \to \infty$.

We shall use an extension of the RG scheme introduced in [7] which has the benefit of treating the fluctuations of the lower surface arising through the $v_0 l_1^2/2$ term exactly, while providing a linear treatment of the l_2-fluctuations. The new feature in the present RG treatment is that we include the position dependence of all of the stiffness matrix elements $\Sigma_{\mu\nu}(l_1, l_2)$ and in particular the off-diagonal term Σ_{12}. To begin we separate out a 'Gaussian-type' contribution $H_0^{(2)}[l_1, l_2]$ from (3.1) such that

$$H_I^{(2)}[l_1, l_2] = H_0^{(2)}[l_1, l_2] + H_W^{(2)}[l_1, l_2] \tag{3.3}$$

where

$$H_0^{(2)}[l_1, l_2] = \int \mathrm{d}^2\boldsymbol{y} \left[\frac{1}{2}\Sigma_{11}^{\infty}(\nabla l_1)^2 + \Sigma_{12}^{\infty}(\boldsymbol{\nabla} l_1)\cdot(\boldsymbol{\nabla} l_2) + \frac{1}{2}\Sigma_{22}^{\infty}(\nabla l_2)^2 + \frac{1}{2}v_0 l_1^2\right] \tag{3.4}$$

and the interaction term due to the wall–interface coupling is

$$H_W^{(2)}[l_1, l_2] = \int \mathrm{d}^2\boldsymbol{y} \left[\frac{1}{2}\Delta\Sigma_{11}(l_1, l_2)(\nabla l_1)^2 + \Delta\Sigma_{12}(l_2 - l_1)(\boldsymbol{\nabla} l_1)\cdot(\boldsymbol{\nabla} l_2) + \frac{1}{2}\Delta\Sigma_{22}(l_1, l_2)(\nabla l_2)^2 + W(l_2 - l_1)\right]. \tag{3.5}$$

Implicit in the definition of these Hamiltonians is a momentum cut-off Λ (or equivalently a short-distance cut-off Λ^{-1}) which we assume to be the same for both fields l_1 and l_2. We observe that $H_0^{(2)}[l_1, l_2]$ is invariant under the RG transformation that integrates out all Fourier components of l_1 and l_2 with wavenumbers $\boldsymbol{k}$ in the range $\Lambda/b < |\boldsymbol{k}| < \Lambda$ and then rescales the system according to

$$\begin{aligned} &\boldsymbol{y} \to \boldsymbol{y}' = \boldsymbol{y}/b \\ &l_i(\boldsymbol{y}) \to l_i'(\boldsymbol{y}') = l_i(\boldsymbol{y}) \qquad i = 1, 2 \\ &v_0 \to v_0' = b^2 v_0 \end{aligned} \tag{3.6}$$

where b is some rescaling factor $b > 1$. The inclusion of the lower surface fluctuation term in $H_0^{(2)}$ leads directly to the coupling constant rescaling in (3.6) which is not found in

standard RG analyses of wetting [17]. The RG flow of $H_0^{(2)}[l_1, l_2]$ under the rescaling (3.6) is well understood and forms the basis for a perturbatory analysis of the $H_W^{(2)}$-term. In other words we treat the interaction term $H_W^{(2)}[l_1, l_2]$ in a purely linearized RG fashion perturbing around the flow of the decoupled Hamiltonian $H_0^{(2)}$.

The first step of the RG analysis is to integrate out the 'fast' modes (i.e. the short-wavelength fluctuations) in the fluctuating fields. To this end we divide $l_1(\boldsymbol{y})$ and $l_2(\boldsymbol{y})$ into long-wavelength and short-wavelength parts:

$$l_i(\boldsymbol{y}) = l_i^<(\boldsymbol{y}) + l_i^>(\boldsymbol{y}) \qquad i = 1, 2 \tag{3.7}$$

where $l_i^<(\boldsymbol{y})$ and $l_i^>(\boldsymbol{y})$ contain all Fourier components of l_i with wavenumbers in the ranges $|\boldsymbol{k}| < \Lambda/b$ and $\Lambda/b < |\boldsymbol{k}| < \Lambda$ respectively. This division means that $H_0^{(2)}$ also separates:

$$H_0^{(2)}[l_1^< + l_1^>, l_2^< + l_2^>] = H_0^{(2)}[l_1^<, l_2^<] + H_0^{(2)}[l_1^>, l_2^>] \tag{3.8}$$

since the short-wavelength and long-wavelength parts have no cross-support in momentum space. Next we define the intermediate renormalized, unrescaled Hamiltonian $H'[l_1^<, l_2^<]$ via a partial trace over the 'fast' modes:

$$\exp(-H'[l_1^<, l_2^<]) = \frac{1}{N_0}\int\int \mathcal{D}l_1^> \; \mathcal{D}l_2^> \;\; \exp(-H_I^{(2)}[l_1^< + l_1^>, l_2^< + l_2^>]) \tag{3.9}$$

where N_0 is the appropriate normalization factor. By writing $H_I^{(2)}$ in momentum space and working to linear order in $H_W^{(2)}$ it is straightforward to calculate these momentum shell integrations using standard Gaussian path integration techniques. To complete the RG transformation we must rescale the resulting equations as prescribed by (3.6).

In order to generate continuous renormalization group flow equations we consider the infinitesimal rescaling limit $b = \mathrm{e}^{\delta t}$, $\delta t \to 0$, which yields

$$2W + \mathcal{C}\left(\Sigma_{22}^{\infty}\Lambda^2\frac{\partial^2}{\partial l_1^2} + (\Sigma_{11}^{\infty}\Lambda^2 + v_0)\frac{\partial}{\partial l_2^2} - 2\Sigma_{12}^{\infty}\Lambda^2\frac{\partial^2}{\partial l_1\,\partial l_2}\right)W + \mathcal{C}(\Sigma_{22}^{\infty}\Lambda^4\,\Delta\Sigma_{11} + (\Sigma_{11}^{\infty}\Lambda^2 + v_0)\Lambda^2\,\Delta\Sigma_{22} - 2\Sigma_{12}^{\infty}\Lambda^4\,\Delta\Sigma_{12}) = \frac{\partial W}{\partial t} \tag{3.10}$$

and

$$\mathcal{C}\left(\Sigma_{22}^{\infty}\Lambda^2\frac{\partial^2}{\partial l_1^2} + (\Sigma_{11}^{\infty}\Lambda^2 + v_0)\frac{\partial}{\partial l_2^2} - 2\Sigma_{12}^{\infty}\Lambda^2\frac{\partial^2}{\partial l_1\,\partial l_2}\right)\Delta\Sigma_{\mu\nu} = \frac{\partial\Delta\Sigma_{\mu\nu}}{\partial t} \qquad \mu, \nu = 1, 2 \tag{3.11}$$

where, for brevity, we have written

$$\mathcal{C} \equiv \frac{\kappa^2}{4\pi[(\Sigma_{11}^{\infty}\Sigma_{22}^{\infty} - \Sigma_{12}^{\infty 2})\Lambda^2 + \Sigma_{22}^{\infty}v_0]}. \tag{3.12}$$

These flow equations can be shown to correctly rederive known limits. In particular, if $\Sigma_{12}(l_1, l_2) = 0$ then (3.10) and (3.11) reduce to the flow equations previously derived using this RG scheme when the cross-gradient coupling term was ignored [7]. Further, the limit $v_0 \to \infty$ corresponds to completely suppressing fluctuations of the lower surface so that the coupled flow equations of Jin and Fisher [1, 2] are appropriate—this is indeed what we find. We also comment that in the limit $v_0 \to 0$ the flow equations reduce to those which are found from regarding the whole binding potential $(U(l_1) + W(l_{21}))$ in a linear fashion. This is true because we have not made use of the dependence of W only on the relative distance l_{21}.

The flow equations (3.10) and (3.11) are very general and consequently difficult to work with. However, we can make some simplifications pertinent to our study which are sufficient to allow the completion of our analysis. Recall that we are considering the case where the upper interface (represented by l_2) unbinds from the wall while the lower one (represented by l_1) remains bound—that is, in the limit of complete wetting the two surfaces of fixed magnetization are infinitely far apart and consequently there is no position-independent (or free) contribution to Σ_{12}. In addition we know the leading-order position dependence of the stiffness matrix arises from the off-diagonal element Σ_{12}, so to leading order we ignore the position dependence of the diagonal elements Σ_{11} and Σ_{22}. Hence for simplicity it suffices to consider bare matrix elements

$$\Sigma_{12}^{\infty} = 0 \qquad \Delta\Sigma_{11} = 0 \qquad \Delta\Sigma_{22} = 0. \tag{3.13}$$

For this model the flow equations simplify considerably. In particular the RG equations for W and $\Delta\Sigma_{12}$ decouple and reduce to

$$\frac{\partial W}{\partial t} = 2W + \frac{\kappa^2}{4\pi\Sigma_{11}^{\infty}}\left(\frac{\Sigma_{11}^{\infty}\Lambda^2}{\Sigma_{11}^{\infty}\Lambda^2 + v_0}\right)\frac{\partial^2 W}{\partial l_1^2} + \frac{\kappa^2}{4\pi\Sigma_{22}^{\infty}}\frac{\partial^2 W}{\partial l_2^2} \tag{3.14}$$

and

$$\frac{\partial \Delta\Sigma_{12}}{\partial t} = \frac{\kappa^2}{4\pi\Sigma_{11}^{\infty}}\left(\frac{\Sigma_{11}^{\infty}\Lambda^2}{\Sigma_{11}^{\infty}\Lambda^2 + v_0}\right)\frac{\partial^2 \Delta\Sigma_{12}}{\partial l_1^2} + \frac{\kappa^2}{4\pi\Sigma_{22}^{\infty}}\frac{\partial^2 \Delta\Sigma_{12}}{\partial l_2^2} \tag{3.15}$$

while those for the $\Delta\Sigma_{ii}$ merely conserve $\Delta\Sigma_{ii} = 0$. These diffusion equations may be straightforwardly integrated. In general the initial (bare) binding potential $W^{(0)}(l_{21})$ and cross-gradient stiffness coefficient $\Delta\Sigma_{12}^{(0)}(l_{21})$ are renormalized according to

$$W^{(t)}(l_{21}) = \mathcal{R}_W^{(t)}[W^{(0)}(l_{21})] \qquad \Delta\Sigma_{12}^{(t)}(l_{21}) = \mathcal{R}_{\Delta\Sigma_{12}}^{(t)}[\Delta\Sigma_{12}^{(0)}(l_{21})] \tag{3.16}$$

where $\mathcal{R}_W^{(t)}$ and $\mathcal{R}_{\Delta\Sigma_{12}}^{(t)}$ are the appropriate recursion operators. Thus integrating (3.14) and (3.15) we find

$$\mathcal{R}_W^{(t)}[W^{(0)}] = \frac{\mathrm{e}^{2t}}{4\pi t\sqrt{\omega_1\omega_2}}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\mathrm{d}l_1'\ \mathrm{d}l_2'\ W^{(0)}(l_2' - l_1')\exp\left\{-\sum_{i=1}^{2}\frac{(l_i - l_i')^2}{4\omega_i t}\right\} \tag{3.17}$$

and

$$\mathcal{R}_{\Delta\Sigma_{12}}^{(t)}[\Delta\Sigma_{12}^{(0)}] = \frac{1}{4\pi t\sqrt{\omega_1\omega_2}}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\mathrm{d}l_1'\ \mathrm{d}l_2'\ \Delta\Sigma_{12}^{(0)}(l_2' - l_1')\exp\left\{-\sum_{i=1}^{2}\frac{(l_i - l_i')^2}{4\omega_i t}\right\} \tag{3.18}$$

where, in analogy with the capillary parameter ω found in single-field analyses of wetting, we define

$$\omega_1 \equiv \frac{\kappa^2}{4\pi\Sigma_{11}^{\infty}}\left(\frac{\Sigma_{11}^{\infty}\Lambda^2}{\Sigma_{11}^{\infty}\Lambda^2 + v_0}\right) \qquad \omega_2 \equiv \frac{\kappa^2}{4\pi\Sigma_{22}^{\infty}} \tag{3.19}$$

(recall that we have set $k_B T = 1$). Hence from (3.17) and (3.18) we see that the recursion operators are simply related thus:

$$\mathcal{R}_{\Delta\Sigma_{12}}^{(t)} \equiv \mathrm{e}^{-2t}\mathcal{R}_W^{(t)} \tag{3.20}$$

which is our central result. Note that the right-hand side is directly related to the singular contribution to the free energy. In particular at the matching point t^* of the rescaling procedure (where fluctuations can be ignored) we may identify $f_{\mathrm{sing}} \sim \mathrm{e}^{-2t^*}W^{(t^*)}(\langle l_{21}\rangle)$

where $\langle l_{21} \rangle$ is the (renormalized) minimum of the potential. For the two-field model considered here the leading-order singularity in $f_{\rm sing}$ is

$$f_{\rm sing} \sim \left(1 + \frac{\omega_1 + \omega_2}{2}\right) \frac{\bar{h}}{\kappa} \ln \bar{h} \tag{3.21}$$

and shows the influence of coupling to the surface fluctuations through the ω_1-term which is absent in the corresponding capillary-wave (and Fisher–Jin) expression. Thus, as we have noted before, the effective value of the wetting parameter is renormalized in the two-field theory [9]. The final two equations (3.20) and (3.21) establish that the values of the critical amplitudes in the two-field theory are robust against the inclusion of position-dependent stiffness elements.

Finally, from (3.20) we see that the value of the stiffness matrix element $2\,\Delta\Sigma_{12}^{(t)}$ (evaluated at the minimum of $W^{(t)}(l_{21})$) follows that of the free energy under RG flow. Consequently, within the two-field formalism the analogue of the sum rule (2.9) is obeyed beyond the MF level since the coefficient of Q^2 in the renormalized structure factor $S_{11}(Q)$ is now related to the total free energy including fluctuation-induced corrections appearing in (3.21). Explicitly, beyond the MF level we can use the renormalized binding potential and stiffness coefficients in the matrix equation (2.16) to calculate

$$S_{11}(Q) = \frac{1}{v_0 + Q^2[\Sigma_{11} + (\Sigma_{22} + 2\Delta\Sigma_{12}^{(t^*)}(z_{21}))/(1 + Q^2\xi_\parallel^2)]} \tag{3.22}$$

where the correlation length retains its MF divergence $\xi_\parallel \sim \bar{h}^{-1/2}$. Thus the flow of the SMFE relation under renormalization is sufficient to establish that the two-field theory is consistent with the exact sum-rule requirement $\mathcal{G}_2(0,0) \propto \Sigma_{w\alpha}$.

4. Discussion and conclusions

In this paper we have extended our earlier RG study of the two-field effective Hamiltonian for complete wetting to allow for position-dependent stiffness coefficients. Whilst the flow equations are in general rather complicated, we have been able to establish an elegant operator relation (3.20) for the RG flows of the cross-coupling term $\Delta\Sigma_{12}$ and the free energy. With this identity the bare SMFE relation, $2\,\Delta\Sigma_{12}(z_{21}) = W(z_{21})$, may be regarded as the correct initial condition to ensure that at the end of the RG trajectory the renormalized stiffness coefficient is similarly related to the renormalized singular part of the free energy, so that equation (2.25) is still valid including fluctuation effects. Using the renormalized quantities, the structure factor $S_{11}(Q)$ at the wall has the same non-Lorentzian form as is found in explicit MF calculations.

To finish we make some brief remarks about systems with long-ranged fluid–fluid forces which we have thus far neglected. For this it is easier to consider a density functional description pertinent to modelling fluid systems and write the grand-potential functional in the local density approximation:

$$\Omega[\rho(\boldsymbol{r})] = -\int \mathrm{d}\boldsymbol{r}\ \rho(\boldsymbol{r})(\mu - V(\boldsymbol{r})) + \mathcal{F}[\rho] \tag{4.1}$$

where μ is the chemical potential, and the intrinsic Helmholtz free energy is given by

$$\mathcal{F}[\rho] = \int \mathrm{d}\boldsymbol{r}\ f_h(\rho(\boldsymbol{r})) + \frac{1}{2}\int\int \mathrm{d}\boldsymbol{r}_1\ \mathrm{d}\boldsymbol{r}_2\ \rho(\boldsymbol{r}_1)\rho(\boldsymbol{r}_2)w(r_{12}). \tag{4.2}$$

The free energy arising from repulsive interactions between fluid molecules is treated in a local density approximation: $f_h(\rho)$ is the Helmholtz free-energy density of a uniform hard-sphere fluid of density ρ. The second term in (4.2) treats attractive forces in MF fashion: the attractive part of the pairwise potential between two fluid molecules, $w(r)$, has the form

$$w(r) = -\frac{A}{r^6} \qquad r \to \infty \tag{4.3}$$

(where A is a constant) characteristic of the tail of the dispersion forces. The local character of the model is not an important deficiency for wetting by the vapour phase (drying) since there are no significant packing or volume-exclusion effects at the wall. Here we concentrate on the case of complete drying at a hard wall corresponding to an external potential

$$V(z) = \begin{cases} \infty & z < 0 \\ 0 & z > 0. \end{cases} \tag{4.4}$$

The model shows coexistence between bulk liquid (l) and gas (g) phases (with number densities $\rho_l(T)$ and $\rho_g(T)$ respectively) at subcritical temperatures and chemical potential $\mu = \mu_{\rm sat}(T)$, and is known to satisfy an analogue of the second-moment sum rule such that [18]

$$\mathcal{G}_2(0,0) = -\Sigma_{wl} \qquad \mu \to \mu_{\rm sat}^+. \tag{4.5}$$

The MF critical exponents for the complete drying transition with these forces are $\beta_s^{co} = 1/3$, $\nu_\parallel^{co} = 2/3$ and $\alpha_s^{co} = 4/3$ and are valid for $d = 3$ [19]. Consequently, according to the sum rule (4.5) there is a next-to-leading-order singularity in $\mathcal{G}_2(0,0)$ of the form $(\mu - \mu_{\rm sat}(T))^{2/3}$. A MF study by Dietrich and Napiórkowski (DN) [20] recovers the correct singular behaviour of the free energy and identifies the traditional binding potential, $W_{DN}(l)$ say, for a film of thickness l as

$$W_{DN}(l) = \Delta\rho(\mu - \mu_{\rm sat})l + \frac{\pi A \rho_g \, \Delta\rho}{12 l^2} + \cdots \tag{4.6}$$

where $\Delta\rho \equiv \rho_l - \rho_g$. From this expression it is straightforward to calculate $f_{\rm sing}$ and the correlation length $\Sigma_{lg}\xi_\parallel^{-2} = \partial^2 W_{DN}/\partial l^2$.

Now consider a two-field Hamiltonian for this system where l_1 corresponds to a surface of suitable fixed density ρ_1^X whilst l_2 has density $\rho_2^X = (\rho_l - \rho_g)/2$. Thus l_1 is always bound to the wall whereas l_2 is a measure of the thickness of the drying film and may be identified with l in (4.6) above. The two-field Hamiltonian can be written as

$$H_I^{(2)}[l_1(\boldsymbol{y}), l_2(\boldsymbol{y})] = \int \mathrm{d}\boldsymbol{y} \, \left[\frac{1}{2}\Sigma_{\mu\nu}(l_1, l_2)\boldsymbol{\nabla} l_\mu \cdot \boldsymbol{\nabla} l_\nu + W^{(2)}(l_1, l_2) \right] \tag{4.7}$$

with

$$\Sigma_{\mu\nu}(l_1, l_2) = \int_{-\infty}^{\infty}\int_{-\infty}^{\infty} \mathrm{d}z_1 \, \mathrm{d}z_2 \, \frac{\partial \rho_\pi^{(2)}}{\partial l_\mu} C_2(z_1, z_2) \frac{\partial \rho_\pi^{(2)}}{\partial l_\nu} \tag{4.8}$$

where C_2 is the second transverse moment of the direct correlation function (defined analogously to the moment expansion in (2.7)):

$$C(\boldsymbol{r}_1, \boldsymbol{r}_2) = \frac{\delta^2 \mathcal{F}[\rho]}{\delta\rho(\boldsymbol{r}_1)\,\delta\rho(\boldsymbol{r}_2)} = \delta(\boldsymbol{r}_1 - \boldsymbol{r}_2)\frac{\mathrm{d}\mu_h}{\mathrm{d}\rho(\boldsymbol{r}_1)} + w(r_{12}) \tag{4.9}$$

where $\mu_h \equiv \mathrm{d}f_h/\mathrm{d}\rho$ is the chemical potential of the uniform hard-sphere fluid and $r_{12} = |\boldsymbol{r}_1 - \boldsymbol{r}_2|$. For $z_1 \neq z_2$ the second moment is given explicitly by

$$C_2(z_1, z_2) = -\frac{1}{4k_B T}\int \mathrm{d}\boldsymbol{r}_\parallel \; \boldsymbol{r}_\parallel^2 w(\sqrt{(z_2 - z_1)^2 + \boldsymbol{r}_\parallel^2}). \tag{4.10}$$

The profile $\rho_\pi^{(2)}(z; l_1, l_2)$ is the doubly constrained planar one-body distribution. Evaluating the integrals in (4.8) we find that the diagonal elements have negligible position dependence and may be identified with the local wall–gas and liquid–gas tensions:

$$\Sigma_{11} = \Sigma_{wg} \qquad \Sigma_{22} = \Sigma_{lg}. \tag{4.11}$$

On the other hand the off-diagonal element is long-ranged due to the decay of $C_2(z_1, z_2)$. The integrals are dominated by the variation of density near the wall (where it is discontinuous) and near the liquid–vapour interface. Clearly we have

$$\Sigma_{12}(0, l_2) = \rho_g \, \Delta\rho \, C_2(0, l_2) + \cdots \tag{4.12}$$

or explicitly

$$\Sigma_{12}(0, l_2) = \frac{\rho_g \, \Delta\rho \, \pi A}{8l^2}. \tag{4.13}$$

Comparing with the DN binding potential we again observe that

$$f_{\text{sing}} = 2\Sigma_{12}(0, l_2) \tag{4.14}$$

which establishes the SMFE for drying with long-ranged forces for $d = 3$. Recall that the upper critical dimension is less than three for such systems, so further RG analysis is not needed.

Thus for $d = 3$ we have shown that the SMFE relation (2.25) is valid for both short-ranged and long-ranged fluid–fluid forces in the presence or absence of fluctuation effects which alter critical amplitudes. We suspect that analogues of the relation are valid in lower dimensions and that it is in fact a general requirement for thermodynamic consistency in theories of correlation function structure.

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