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Structure, phase transitions and dynamics of interfaces and surfaces†

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Abstract. The structure of the interface between coexistent phases in the planar Ising model is reviewed and some new results are given for interfaces not in principal directions. The Wulff construction is investigated in this respect. A brief review of exactly solvable models of phase transitions in surfaces is given. The paper concludes with new results for the dynamics of interfaces.

1. Statics

1.1. Introduction—Ising model

Consider a cubic lattice with integer-valued coordinates. At each vertex $i \in \mathbb{Z}^d$ there is a spin $\sigma(i) = \pm 1$ and the state of the system is described by all such values for a given lattice Λ, denoted $\{\sigma(\Lambda)\}$. The energy and canonical ensemble probability of such a configuration are given by

$$E\{\sigma(\Lambda)\} = -H \sum \sigma(i) - J \sum_{\langle i,j \rangle} \sigma(i)\sigma(j) \tag{1.1}$$

where $\langle i, j \rangle$ signifies a sum over nearest-neighbour pairs and

$$p\{\sigma(\Lambda)\} = Z_\Lambda^{-1} \exp(-\beta E\{\sigma(\Lambda)\}) \tag{1.2}$$

where $\beta = 1/kT$ introduces the temperature T and Z_Λ is a normaliser. Regarding the spins as magnetic, H is a field and J is an exchange coupling. The model may also be thought of in other contexts.

(*a*) *Lattice gas.* $\sigma(i) = +1$ (-1) corresponds to particle present (absent) in cell i. The intermolecular interaction is infinitely repulsive for two particles in the same cell (a configuration not allowed in the Ising model), attractive with value $4J$ for nearest neighbours and zero otherwise. The fugacity is $\exp(2\beta J + nk)$, where n is the number of nearest neighbours.

(*b*) *Binary mixtures and alloys.* $\sigma(i) = 1$ (-1) means cell i contains species a (species b). The coupling $J > 0$ favours like neighbouring pairs over unlike ones and the field acts as a differential fugacity.

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Most of the remarks which follow will concern the *planar* Ising model which has a rich behaviour which is understood in some detail, mainly because of the interplay of general mathematical results (Lebowitz 1977, Miracle-Sole 1976, Ruelle 1968, Abraham 1987) with exact calculations which go back to the fundamental work of Onsager (1944). An increasing number of applications are emerging in surface science: for instance, the phase diagrams of CH_4 sub-monolayer on graphite (Kim and Chan 1974), the surface reconstruction of (110) facets on Au (a $(2\times 1)\rightarrow(1\times 1)$ transition) (Campuzano *et al* 1985), roughening of Au on (110) W (Kolaczkiewicz and Bauer 1985), to cite but three.

1.2. *Phase diagram of Ising model*

The mean magnetisation is defined by

$$m(h, T) = \lim_{\Lambda\rightarrow\infty} \frac{1}{|\Lambda|} \sum_{i\in\Lambda} \langle\sigma(i)\rangle \tag{1.3}$$

where $\langle\ \rangle$ is the expectation with respect to (1.2). The main features of the diagram are

(i) the obvious symmetry $m(h, T) = -m(-h, T)$,

(ii) concavity in h for $h \geq 0$ (Griffiths *et al* 1970),

(iii) existence of non-zero spontaneous magnetisation for $T < T_c(2)$, the critical temperature (Martin-Lof 1972, Lebowitz and Martin-Lof 1972) and its exact calculation (Onsager 1949, Yang 1952).

The equation of state cannot be calculated exactly for $h \neq 0$ (except for the critical indices γ, γ' and δ), but the behaviour far enough from the critical point can be obtained from rigorous perturbation theory (to all orders) (Ruelle 1968).

1.3. *Interface structure*

To study the spatial relationship between pure phases at coexistence (this means the bulk field $H = 0$, $T < T_c(2)$), a small magnetic field $h(i) = h \operatorname{sgn} i_2$ can be applied. This is equivalent to specifying boundary conditions

$$\{\sigma(i) = +1\ (-1) \text{ if } i \in \partial\Lambda,\ i_2 \geq 1\ (i_2 \leq 0)\}$$

and having $h(i) = 0$ in the bulk (see figure 1). This result is proved by correlation inequalities (Abraham and Issigoni 1980) and makes further calculations, which are

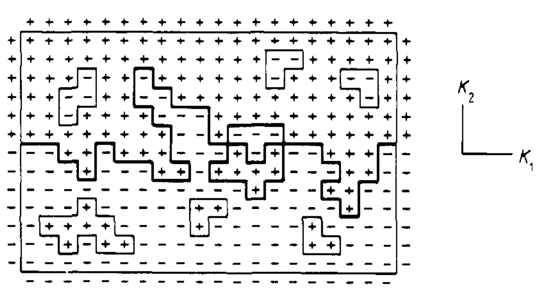

Figure 1. A spin configuration with symmetry breaking boundary conditions. There is a 'long contour' λ indicated by a bold line connecting the points of spin flip on the boundary. The regions above and below λ have their magnetisation altered from ± 1 by the small closed loops shown.

now technically possible, have a precise meaning. For instance, the surface tension is defined by

$$\tau = \lim_{N\to\infty} (1/2N) \lim_{M\to\infty} \log(Z^{++}/Z^{+-}) \tag{1.4}$$

in terms of partition functions Z^i; Z^{++} has all boundary spins +. Since we are at coexistence, the bulk and wall contributions are expected to cancel out in (1.4), leaving the interface term. This is indeed the case, giving

$$\tau = 2(K - K^*) \tag{1.5}$$

with $\sinh 2K \sinh 2K^* = 1$.

According to historically accepted ideas due to van der Waals, the magnetisation $m(x, y)$ should vary with y, but not x, in the thermodynamic limit for laboratory = fixed axes, as defined by the containing vessel in figure 2. Instead, we find (Abraham and Reed 1974)

$$\lim_{N\to\infty} \lim_{M\to\infty} \langle\sigma(x, y)\rangle = 0. \tag{1.6}$$

Evidently, the interface is somewhere else. It turns out that, to recapture it, we have to scale y with the width of the system. The general result is then expressed by theorem 1 (Abraham and Reed 1976).

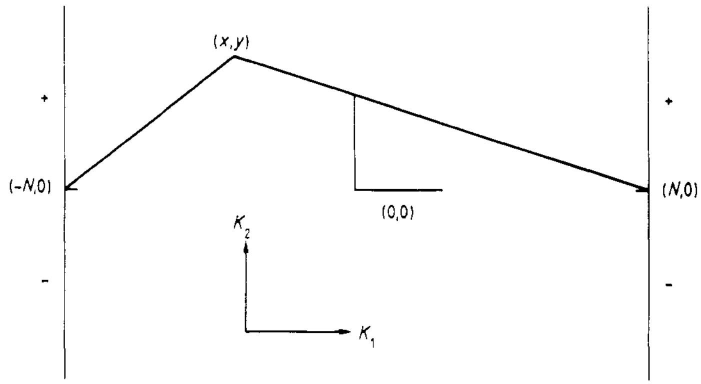

Figure 2. The labelling of axes and interactions for the symmetry breaking boundary conditions used to establish the interfacial profile.

Theorem 1. For all $0 \leq T < T_c(2)$, $\alpha \in \mathbb{R}$, $\beta \in (-1, 1)$ we have

$$\lim_{N\to\infty} \lim_{M\to\infty} \langle\sigma(\beta N, \alpha N^\delta)\rangle = \begin{cases} 0 & \delta < \frac{1}{2} \\ m^* \operatorname{sgn} \alpha & \delta > \frac{1}{2} \\ m^* \operatorname{sgn} \alpha \Phi\left(\dfrac{b(T)|\alpha|}{(1-\beta^2)^{1/2}}\right) & \delta = \frac{1}{2} \end{cases} \tag{1.7}$$

where

$$\Phi(x) = \frac{2}{\sqrt{\pi}} \int_0^x \exp(-u^2)\, du \tag{1.8}$$

and

$$b(T) = (\sinh \tau)^{1/2}. \tag{1.9}$$

1.4. Remarks

(i) Let us draw lines separating antiparallel neighbouring spin pairs, as in figure 3. There will be a long contour γ_0 connecting $(-N, \frac{1}{2})$ to $(N, \frac{1}{2})$. Gallavotti (1972) proved that, for $T \ll T_c(2)$, the minimum distance from γ_0 to any point (x, y) denoted $\delta(\gamma_0, (x, y))$ satisfies

$$\lim_{N\to\infty} \lim_{M\to\infty} P\{d(\gamma_0, (x, y)) < N^{\delta}\} = 0 \tag{1.10}$$

for any $\delta < \frac{1}{2}$. This is consistent with theorem 1 at an intuitive level.

(ii) For an arbitrary product of spins in some Λ sufficiently large, Aizenman (1979, 1980) and Higuchi (1979) proved that a translationally variant correlation function cannot be induced in the thermodynamic limit no matter how spins are fixed on the boundary.

(iii) These results can be generalised to interfaces whose mean direction is any angle $\theta \in (-\pi/2, \pi/2)$ by moving the right-hand side of Λ in figure 4 so that γ_0 ends at $(N, [2N \tan \theta])$. Then the surface tension is (Abraham and Reed 1977)

$$\tau(\theta) = |\cos \theta| \gamma(\omega(\theta)) - \mathrm{i} \sin \theta \omega(\theta) \tag{1.11}$$

where $\omega = \omega(\theta)$ solves

$$\mathrm{i} \tan \theta = \partial\gamma/\partial\omega \tag{1.12}$$

(with $\mathrm{Re}\, \omega = 0$ to fix the branch). The Onsager function is defined by

$$\cosh \gamma(\omega) = \cosh 2K_1^* \qquad \cosh 2K_2 - \sinh 2K_1^* \sinh 2K_2 \cos \omega \tag{1.13}$$

with $\gamma(\omega) \geqslant 0$ for $\omega \in \mathbb{R}$. For typical polar plots of $\tau(\theta)$, see the review article by Rotmann and Wortis (1984). The analogous magnetisation theorem is for the function

$$\lim_{N\to\infty} \lim_{M\to\infty} \langle \sigma(\beta N, (1+\beta) N \tan \theta + \alpha N^{\delta}) \rangle$$

which satisfies the same results for $\delta < \frac{1}{2}$ and $\delta > \frac{1}{2}$. For $\delta = \frac{1}{2}$, the scaling factor $b(T)$ acquires θ dependence, being replaced by

$$b(\theta, T) = [\gamma^2(\omega(0))]^{-1/2} \tag{1.14}$$

(recall (1.11) and (1.12)).

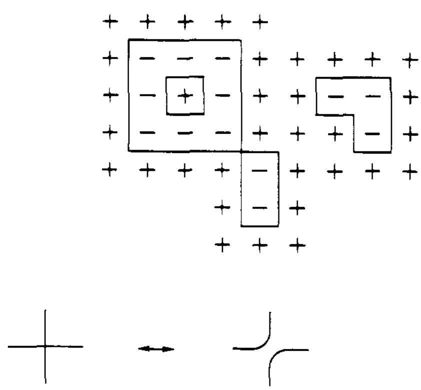

Figure 3. Spin configurations and contours. A unit edge on the dual lattice Λ^* separates any part of antiparallel spins: 0, 2 or 4 lines may meet at interior points of Λ^*. The contours are edge self-avoiding. Where they cross, they can be deformed as shown into self-avoiding loops. To avoid double-counting, the other non-crossing decomposition is suppressed.

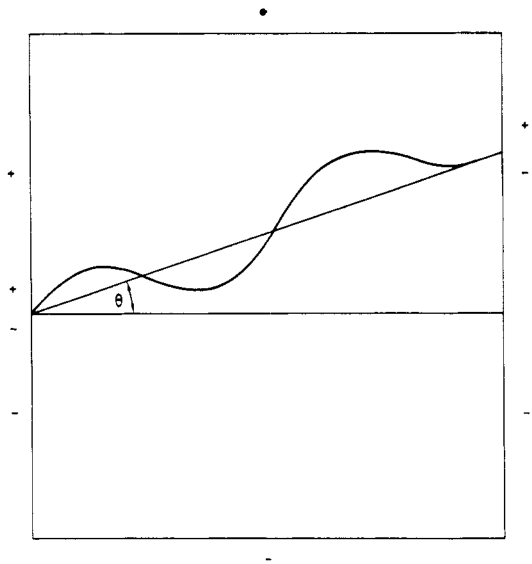

Figure 4. Angle-dependent interfacial free energy. The point of spin reversal is sheared on the right-hand edge. The long contour is shown schematically.

The width of the interface thus satisfies

$$\lim N^{-1/2} W(\theta, T) = (\tau(\theta) + \tau^2(\theta))^{1/2} \tag{1.15}$$

a formula conjectured for the Ising model by Akutsu and Akutsu (1986) and proved by them for SOS models, and in greater generality by De Coninck and Ruiz (1988). This has interesting properties as $T \to 0+$ on $[0, \pi/2]$:

$$\lim_{T\to 0} \lim_{N\to\infty} N^{-1/2} W(\theta, T)^{-} = \sin\theta \cos\theta (\sin\theta + \cos\theta). \tag{1.16}$$

Thus only at $\theta = 0$, $\pi/2$, the scaled width vanishes: we have a facet, as the Wulff construction suggests, but not at intermediate angles.

1.5. The Wulff construction

According to thermodynamical arguments (for a review, see Rottman and Wortis (1984)), the equilibrium mean shape of the crystal surface is given by minimising the line integral for the free energy

$$F(N, \theta) = \int_{(-N,0)}^{(N,[2N\tan\theta])} \tau(\theta(l))\,\mathrm{d}l \tag{1.17}$$

with the fixed boundary conditions, and here no area constraint. For differentiable solutions, $\theta(l) = \theta$ everywhere on the interface line, which is thus straight. As pointed out by Ball, if we allow discontinuous solutions, any non-decreasing function f from $\mathbb{Z} \cap [-N, N]$ to $\mathbb{Z} \cap [0, [2N \tan\theta]]$ with $f(-N) = 0$, $f(N) = [2N \tan\theta]$ is a minimiser.

The $T \to 0$ limit shows that not all these solutions are significant, which is a rather curious fact. One way of viewing theorem 1 is that we derive both the conclusions of the Wulff construction (albeit a rather trivial example) for $T > 0$ and the fluctuations about the mean structure, which are normally tacked on later in an *ad hoc* and not entirely convincing fashion.

1.6. The wetting transition

Consider the arrangement shown in figure 5. With + spins on the top and the bottom we expect a + phase; the effect of the perturbed bonds will be to give a magnetisation which decays from +1 to $+m^*$ on the scale of the bulk correlation length (Abraham 1980). But if we reverse all spins on the lower face, there will be a domain wall. If $0<a<1$, then the minimum energy will be attained with the domain wall *intersecting* the modified bonds. But the entropy of wandering will be lost. This competition produces a phase transition of second order (Abraham 1980) at a temperature $T_w(a)<T_c(2)$ for $0<a<1$, in which the low-temperature phase has the domain wall bound (see figures 6 and 7). This squeezes out the − phase, which may be said to wet partially

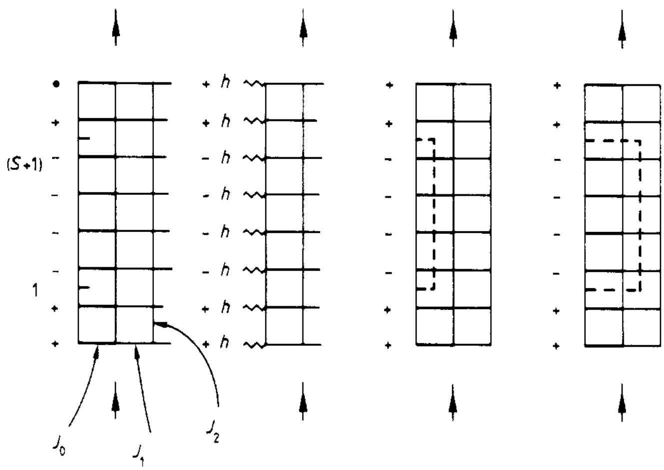

Figure 5. A planar Ising model with modified surface bonds and a domain wall, or long contour beginning and ending in the surface. This is equivalent to the external field arrangement shown in the second drawing with $h=J_0$. The reader should compare the Boltzmann factors for the last two drawings for general s.

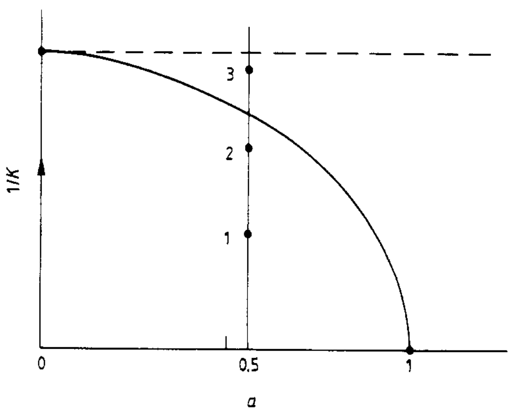

Figure 6. The phase diagram for the system depicted in figure 5. The thermodynamic limit has been taken, followed by $s\to\infty$. Between the curve and the axes, the domain wall is bound to the surface; outside, it fluctuates 'out of sight'. These two regions correspond to the partially wet and wet cases, respectively.

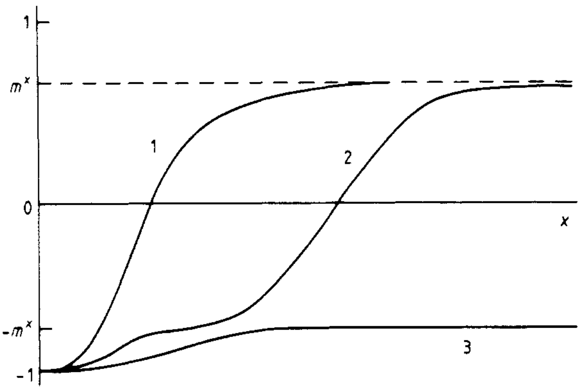

Figure 7. Sketches of magnetisation profile near a wall. The curves are labelled by the corresponding points on the line in figure 6. In curve 2, a new length ξ appears scaling the switch from $-m^*$ to $+m^*$. Curve 3 is typical of the situation $T_c(2) > T > T_w(a)$: $m(x)$ tends to $-m^*$ from below as $x \to \infty$.

the bottom surface. For $T_c(2) > T > T_w(a)$, a macroscopic film of − phase intercalates between the wall and the + phase.

The phase diagram and magnetisation as a function of y are shown in figures 6 and 7. Clearly a new length scale $\xi^x(a, T)$ emerges. As $T \to T_w(a)^-$,

$$\xi^x(a, T) \sim [(T_w(a) - T)/T_w(a)]^{-1}. \tag{1.18}$$

1.7. *Remarks*

(i) This phase transition may also be studied by random walk models (Fisher 1984, Abraham and De Coninck 1983) which show that the droplets of mainly − spins at the wall are *microscopic* in size. Thus the picture advocated by Cahn (1977) in his important work on wetting does not hold. There the thermodynamic stability of a sessile drop with a contact angle $\theta_c(a, T)$ is discussed. In the case discussed here, the contact angle makes no sense for microscopic droplets. But De Coninck and Dunlop (1987) have made sense of this picture for the random walk model by introducing a constant area constraint which forces one drop to be macroscopic. The contact angle so defined satisfied the modified Young equation:

$$\cos\theta\, \tau_\pm(\theta) - \sin\theta\, \partial\tau_\pm/\partial\theta = \tau_{-w} - \tau_{+w}. \tag{1.19}$$

(The modification is the introduction of the derivative term which is appropriate for an underlying lattice.) The least T for which $\theta_c = 0$ may also be considered as a thermodynamic definition of $T_w(a)$. This was investigated in Cahn (1977). Recently, (1.19) has been used to obtain contact angles for the planar Ising model, and its derivation from first principles of statistical mechanics is under way, using a 'fixed area' constraint (Abraham *et al* 1987). It is possible that a sessile drop without the fixed area (volume in 3D) is metastable.

(ii) Binding and wetting of domain walls to other domain walls. The terrace-ledge-kink (TLK) model of a crystal surface is illustrated and explained in figure 8. Because the kinks expose more area (or reduce bonding), they are disfavoured. Under appropriate circumstances, the ledges either repel or attract (this may depend in a very sensitive way on surface impurities, such as adsorbed gases). It turns out that Lieb's ice-rule Bethe ansatz technique (the six-vertex problem) can be used on a suitably adapted model. For attractive ledges, there is a drastic surface reconstruction (Abraham 1983)

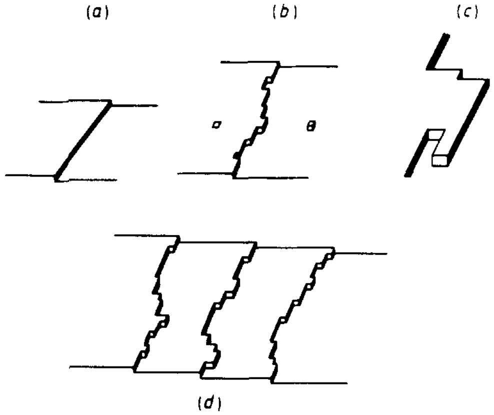

Figure 8. The terrace-ledge-kink (TLK) model. (*a*) shows a ledge separating two terraces having a height difference of units in atomic sizes. (*b*) shows kinks formed on the ledge. The pit on the left-hand terrace and erratic adatom on the right-hand side are not allowed in the model (they are energetically unfavourable). (*c*) shows an overhang on a ledge; this is not allowed. (*d*) shows a group of three ledges forming part of a vicinal section. The ledges cannot cross, but they can touch.

where the vicinal section (cheese wedge) develops macroscopic facets perpendicular to the basal plane. On the other hand, if the ledges repel when the angle of the section is such that there is a unique ground state, there is a low-temperature state with a facet at that angle. This facet breaks up at a roughening transition which is of F type, and thus of infinite order. For other angles, there is no phase transition and no facet.

The topics in § 1 which are not new are reviewed in greater depth in Abraham (1987).

2. Dynamics

We now return to the structure of the interface with which § 1 began and discuss the dynamical stability of the $+-$ domain wall. The Ising model itself has no dynamics but we can introduce the dynamical effect of the underlying heat bath in the following way: on a sufficiently large length scale much greater than the bulk correlation length, the Ising model interface becomes a string with essentially no overhang in the direction normal to the mean interface, which we regard as a straight line from $(-L, 0)$ to $(L, 0)$. For each integral n in $[-L, L]$ we give the displacement $h(n)$ of the string from the mean interface. The free energy functional for a given configuration may be taken as

$$F[\{h(n)\}] = 2L\tau + K \sum_{-L+1}^{L} (h(n) - h(n-1))^2 \tag{2.1}$$

with $h(-L) = h(L) = 0$. Equation (2.1) is only valid physically in the coarse-grained sense, but we shall nevertheless take $h(n) \in \mathbb{R}$. The Onsager–Machlup, or Langevin, dynamics which are clearly explained by Kac and Logan (1979) may be introduced:

$$\begin{aligned} \partial h/\partial t &= -\Gamma \nabla F + \eta(t) \\ &= -K\Gamma A h + \eta(t) \end{aligned} \tag{2.2}$$

where

$$A = \begin{pmatrix} 2 & -1 & & & & & \\ -1 & 2 & -1 & & & 0 & \\ & -1 & 2 & -1 & & & \\ & & & \ddots & & & \\ & 0 & & & -1 & 2 & -1 \\ & & & & & -1 & 2 \end{pmatrix} \tag{2.3}$$

and $\eta(t)$ is Gaussian white noise for which

$$\begin{aligned} \langle \eta_i(t) \rangle &= 0 \qquad \forall i = -L, \ldots, L, \forall t \\ \langle \eta_i(t)\eta_j(t') \rangle &= 2\Gamma K \delta_{ij}\delta(t-t'). \end{aligned} \tag{2.4}$$

First we reduce the problem by normal mode analysis of A: let

$$A = S\Lambda S^{\mathrm{T}} \tag{2.5a}$$

where S is a (real) orthogonal matrix and let

$$\hat{h} = S^{\mathrm{T}}h \qquad \hat{\eta} = S^{\mathrm{T}}\eta. \tag{2.5b}$$

Then

$$\partial \hat{h}_q/\partial t = -\Gamma K \lambda_q \hat{h}_q + \hat{\eta}_q \tag{2.6}$$

where λ_q is an eigenvalue of A. The transformation does not alter the properties of the noise, so the problem may be restated stochastically in terms of the conditional probability $P\{\hat{h}_q(t)\,|\,\hat{h}_q(0)\}$, which we denote P for convenience:

$$\frac{\partial P}{\partial t} = \Gamma K \lambda_q \frac{\partial}{\partial \hat{h}_q(t)}\{\hat{h}_q(t)P\} + \Gamma K \frac{\partial^2 P}{\partial \hat{h}_q(t)^2} \tag{2.7}$$

the solution of which is the Ornstein-Uhlenbeck process:

$$P\{\hat{h}_q(t)\,|\,\hat{h}_q(0)\} = \left(\frac{\lambda_q}{2\pi[1-\exp(-2\Gamma K\lambda_q t)]}\right)^{1/2} \exp\left(-\frac{\lambda_q}{2}\,\frac{[\hat{h}_q(t)-\hat{h}_q(0)\exp(-\Gamma K\lambda_q t)]^2}{1-\exp(-2\Gamma K\lambda_q t)}\right). \tag{2.8}$$

Notice that, as $t \to \infty$, $P \to [(\lambda_q/2\pi)]^{1/2}\exp(-\frac{1}{2}\lambda q h_q^2(\infty))$, the initial condition is forgotten and the limiting Gaussian distribution leads back to theorem 1, through

$$P\{h_0(t)\,|\,h(0)=0\} = \frac{1}{(2\pi\sigma^2(t))^{1/2}}\exp[-h_0^2(t)/2\sigma^2(t)] \tag{2.9}$$

which is obtained by using characteristic functions and Gaussian integration, with

$$\sigma^2(t) = \frac{1}{2L}\sum_1^L \frac{1-\exp(-4\Gamma Kt\{1-\cos[(2k-1)\pi/2L]\})}{1-\cos[(2j-1)\pi/2L]} \tag{2.10}$$

where the explicit form of S and λ_q in (2.5a, b) has been used. The solution separates into several regimes.

(a) If $\Gamma t \ll 1$, with L finite, we get a diffusive regime

$$\langle h_0(t)^2 \rangle \sim t. \tag{2.11}$$

(*b*) If $L \to \infty$, the sum becomes a Riemann integral, giving

$$\sigma^2(t) = \frac{1}{4\hbar} \int_0^\pi \mathrm{d}q \frac{1 - \exp[-2\Gamma K t(1 - \cos q)]}{1 - \cos q} \tag{2.12}$$

which behaves like $\sqrt{\Gamma t}$ for $\Gamma t \gg 1$, unlike the diffusive case.

(c) The scaling limit, denoted s-lim with $t \to \infty$, $L \to \infty$, such that $z = \Gamma t / l^2$ is fixed, s-lim $\sigma^2(t)/L = F(z)$ where

$$F(z) = \frac{2}{\pi^2} \sum_1^\infty \frac{1 - \exp[-z\pi^2(2j-1)^2]}{(2j-1)^2}. \tag{2.13}$$

For $t \gg L^2$, z is large and the limit is approached ultimately *exponentially*:

$$F(z) \sim \frac{1}{2} - \frac{2}{\pi^2} \exp(-z\pi^2). \tag{2.14}$$

As t decreases, more timescales become relevant (or unsaturated). To recapture the small-z result ($t \ll L^2$)

$$F(z) \propto z^{1/2} \tag{2.15}$$

we have to bring in an infinite sequence of timescales:

$$t_j = L^2 / \pi^2 (2j-1)^2 \Gamma K \tag{2.16}$$

through the Poisson summation formula.

To sum up, the main points are that the initially flat interface is unstable to macroscopic fluctuations, but the time is scaled typically by L^2 and an infinite number of timescales are involved as $L \to \infty$.

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