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Relationship between the anisotropic interface tension, the scaled interface width and the equilibrium shape in two dimensions

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Abstract. The formula which relates the scaled interface width to the anisotropic interface tension is generalised for an arbitrary orientation of the interface in two dimensions. The relationship between the scaled interface width and the curvature of the equilibrium crystal shape is also generalised.

1. Introduction

It is well known that the one-dimensional interfaces for two-dimensional systems are rough and delocalised at any temperature $T>0$, as has been proved (Gallavotti 1972, Abraham and Reed 1974, Higuchi 1979, Aizenman 1980) for the two-dimensional square-lattice Ising system. The interface delocalisation width is of the order of $N^{1/2}$, with N being the linear size of the system and then suitably scaled quantities (e.g. scaled interface profile and scaled interface width) have been introduced and analysed. In particular, the exact scaled interface profile in a two-dimensional square-lattice Ising system has been calculated (Abraham and Reed 1976, 1977, Abraham 1981) to be the integral of the Gaussian distribution whose variance, the scaled interface width, σ, is finite when $T<T_c$, the critical temperature of the Ising model, and diverges like $|T-T_c|^{-1/2}$ as $T\to T_c$.

Recently, much attention has been paid to the anisotropy of interface properties. As to the anisotropy of σ, the exact calculation is limited to the special case $\theta=0$, where θ is the mean tilt angle of the interface relative to the crystal axis. For a thermodynamical treatment, there exists an argument by Fisher *et al* (1982, hereafter referred to as FFW) which showed that, by taking into account the correct anisotropy of the interface tension, the modified capillary wave theory (Buff *et al* 1965, Weeks 1977) reproduces the exact results on the Ising model for σ. We note here that the results of FFW are also limited to the case $\theta=0$ and that the generalisation for general θ is not attained by the original argument of FFW itself, since it utilises the special case of a $\theta=0$ interface.

The purpose of this paper is to present the generalised formula which relates the anisotropy of the interface tension to that of the scaled interface width. In § 2, we derive the formula by a thermodynamical argument similar to FFW but which is slightly different from FFW in that we use the Legendre-transformed interface tension introduced by Andreev (1982). In § 3, we perform explicit calculations on the SOS model to verify the formula from a microscopic point of view. In § 4, the formula is applied to the Ising model. In § 5, we will show that the curvature of the equilibrium crystal shape is directly connected to the scaled interface width. The last section is devoted to a summary and discussion.

2. Thermodynamical argument

We first review the argument by FFW. We consider an interface, the reference interface, which extends along the crystal axis. We denote its length (i.e. the distance between the starting point and the endpoint) by L. To cause a tilt to this interface by fixing the starting point and moving its endpoint in the direction normal to the reference interface, excess free energy ΔF is required. Denoting the tilt angle by θ, we can calculate ΔF as

$$\Delta F = \gamma(\theta)(L/\cos\theta) - \gamma(0)L. \tag{1}$$

In the above, $\gamma(\theta)$ is the interface tension (the interface free energy per unit length) and $L/\cos\theta$ is the length of the inclined interface. For small θ, we can expand ΔF in θ to have

$$\begin{aligned}\Delta F/L &= (\gamma(0)+\tfrac{1}{2}\gamma''(0)\theta^2+\ldots)/(1-\tfrac{1}{2}\theta^2+\ldots)-\gamma(0)\\ &= \tfrac{1}{2}\theta^2(\gamma(0)+\gamma''(0)).\end{aligned} \tag{2}$$

The quantity $\gamma(0)+\gamma''(0)$ is sometimes called the stiffness of the interface. In the above, we do not have the θ-linear term $\gamma'(0)\theta$ because of the speciality of the $\theta=0$ interface, $\gamma'(0)=0$, which comes from the crystal symmetry and from the fact that the one-dimensional interface is rough at any finite temperature. FFW pointed out that, in the prediction of the modified capillary wave theory (Buff *et al* 1965, Weeks 1977) for the interface width w,

$$w^{-2} = \beta\gamma(0)/L \tag{3}$$

with β being the inverse temperature $1/(k_BT)$, the interface tension $\gamma(0)$ should be substituted by the 'effective' interface tension $\gamma(0)+\gamma''(0)$ to give

$$w^{-2} = \beta(\gamma(0)+\gamma''(0))/L. \tag{4}$$

In terms of the scaled interface width $\sigma(0) = w/L^{1/2}$, we have

$$\sigma^{-2}(0) = \beta(\gamma(0)+\gamma''(0)) \tag{5}$$

which agrees with the exact calculation in the Ising model (Abraham and Reed 1974, 1977, Abraham 1981).

It is quite natural to suppose that relation (5) admits a simple generalisation for a general inclination angle θ of the reference interface, namely

$$\sigma^{-2}(\theta) = \beta(\gamma(\theta)+\partial^2\gamma(\theta)/\partial\theta^2). \tag{6}$$

However, since the original derivation of FFW utilises the fact $\gamma'(0)=0$, there must be some modification of the derivation of the formula (6) for general θ where $\gamma'(\theta)\neq 0$ in general. For this purpose, we present an argument similar to, but slightly different from, that of FFW, which is suitable for the derivation of (6) for $\theta\neq 0$.

To treat the macroscopically inclined interface and its fluctuation, we introduce the 'generalised free energy' $F(p, \eta)$ defined by

$$F(p, \eta) = L(f(p) - \eta(p)) \tag{7a}$$

$$f(p) = \tilde{\gamma}(p)(1+p^2)^{1/2} \tag{7b}$$

where we have $p = \tan\theta$, $\tilde{\gamma}(p) = \gamma(\theta)$ and the parameter η is the 'field' conjugate to the variable p. Note that, by introducing $F(p, \eta)$, we have made the Legendre transformation of the variable $p \to \eta$. We regard $p = \tan\theta$ as a fluctuating variable and we determine the mean inclination angle from the condition $\partial F(p, \eta)/\partial p = 0$, the minimisation condition of $F(p, \eta)$ with respect to p, which amounts to

$$\partial f(p)/\partial p = \eta. \tag{8}$$

The above condition gives the equilibrium value of p and θ, $p = p^*(\eta)$ and $\theta = \theta^*(\eta)$. We want to consider the fluctuation of the interface around its equilibrium. We put $\Delta p = p - p^*(\eta)$ and expand $F(p, \eta)$ around $p = p^*(\eta)$ to obtain

$$F(p, \eta) = L\tilde{f}(\eta) + \tfrac{1}{2}L\partial^2 f/\partial p^2|_{p=p^*}(\Delta p)^2 \tag{9a}$$

$$\tilde{f}(\eta) = F[p^*(\eta), \eta]/L. \tag{9b}$$

The quantity $\tilde{f}(\eta)$ is the Andreev free energy (Andreev 1982). For notational simplicity we write $p^*(\eta)$ and $\theta^*(\eta)$ as p and θ in the following. The standard thermodynamical fluctuation theory gives

$$\langle(\Delta p)^2\rangle = (1/\beta)(L\partial^2 f(p)/\partial p^2)^{-1}. \tag{10}$$

Remembering $p = \tan\theta$, we have

$$\langle(\Delta p)^2\rangle = \langle(\Delta\theta)^2\rangle/\cos^4\theta \tag{11a}$$

$$\partial^2 f(p)/\partial p^2 = \cos^3\theta(\gamma(\theta) + \gamma''(\theta)). \tag{11b}$$

The deviation of the endpoint of the interface, normal to the reference inclined interface, Δh, is related to $\Delta\theta$ as

$$\Delta h = \tilde{L}\Delta\theta \tag{12}$$

with $\tilde{L} = L/\cos\theta$. Introducing the scaled variable $y = \Delta h/\tilde{L}^{1/2}$ and defining the scaled interface width $\sigma(\theta)$ by $\sigma^2(\theta) = \langle y^2\rangle$, we obtain formula (6).

3. SOS calculations

To amplify the thermodynamical argument presented in the previous section, we perform explicit calculations on an simplified interface model, the so-called SOS (solid-on-solid) model (Burton *et al* 1951). As was pointed out by Huse *et al* (1985), the SOS description of the interface wandering is correct when we discuss the large length scale properties of the interface.

In the SOS model, the 'overhang' configurations are forbidden. The effect of the bubbles in the bulk is also excluded. Then the interface configuration is represented by a set of heights $\{h_i\}_{i=0}^{N}$ as shown in figure 1 (we take $h_0=0$). The Hamiltonian H_N is defined by

$$H_N=\sum_{i=0}^{N-1}|h_i-h_{i+1}|. \tag{13}$$

We define the 'constrained partition function' $Z_N(h)$ by fixing $h_N=h$ and summing over other variables $\{h_i\}$:

$$Z_N(h)=\sum_{h_1=-\infty}^{\infty}\cdots\sum_{h_{N-1}=-\infty}^{\infty}\exp(-\beta H_N). \tag{14}$$

The unconstrained partition function Z_N is just $\Sigma_{h=-\infty}^{\infty} Z_N(h)$. We put $h_N=N\tan\theta$, where θ is the mean inclination angle of the interface. The interface tension $\gamma(\theta)$ is related to $Z_N(h)$ as

$$Z_N(N\tan\theta)=\exp\left(-\frac{N}{\cos\theta}\beta\gamma(\theta)\right). \tag{15}$$

Strictly speaking, the above $\gamma(\theta)$ is N dependent. However, the correction to its $N\to\infty$ limit is $O(1/N)$ and is neglected in the following arguments.

We introduce a probability distribution function $p(n,h)$ defined by

$$\begin{aligned} p(n,h)&=\langle\delta_{h_n,h}\rangle\\ &=\frac{1}{Z_N}\sum_{h_1=-\infty}^{\infty}\cdots\sum_{h_N=-\infty}^{\infty}\delta_{h_n,h}\exp(-\beta H_N) \end{aligned} \tag{16}$$

where $\delta_{i,j}$ stands for the Kronecker delta. As can easily be verified, we have

$$p(n,h)=Z_n(h)/Z_n. \tag{17}$$

The function $p(n,h)$ is the 'transition probability' of the interface position from $(0,0)$

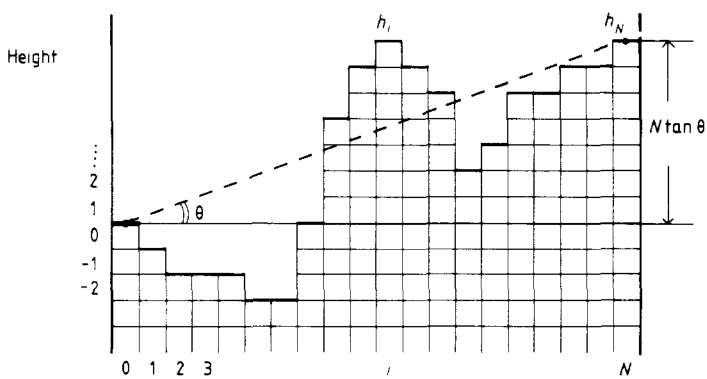

Figure 1. An interface configuration in the SOS (solid-on-solid) model. The site of each column is denoted by i and the column height by h_i. We set $h_0=0$. The mean inclination angle θ is defined by $\tan\theta=h_N/N$.

to (n, h). Then the probability of finding the interface position at (n, h) *in the inclined interface*, which we denote by $P_N(n, h, \theta)$, is calculated as the probability of taking the path $(0,0) \to (n, h) \to (N, N \tan\theta)$ relative to that of taking the path $(0,0) \to (N, N\tan\theta)$:

$$P_N(n, h, \theta) = p(n, h)p(N-n, N\tan\theta - h)/p(N, N\tan\theta) \tag{18a}$$

$$= Z_n(h)Z_{N-n}(N\tan\theta - h)/Z_N(N\tan\theta) \tag{18b}$$

which comes from the Markovian nature of the interface wandering in the SOS model. We want to calculate the deviation probability of the interface position from the reference mean inclined interface. For this purpose, we put $h = n\tan\theta + \Delta h$ in equation (18) and define a function $\tilde{P}_N(n, \Delta h, \theta)$ as

$$\tilde{P}_N(n, \Delta h, \theta) = P_N(n, n\tan\theta + \Delta h, \theta). \tag{19}$$

We are interested in the asymptotic behaviour in the $N \to \infty$ limit. As can be verified by a direct calculation, the function $\tilde{P}_N(n, \Delta h, \theta)$ has a well defined non-trivial (Gaussian) form with the scaled variables

$$y = \Delta h / N^{1/2} \tag{20a}$$

$$x = n/N. \tag{20b}$$

We further define a variable s which measures the deviation of interface position normal to the mean interface, scaled by $(N/\cos\theta)^{1/2}$, with $N/\cos\theta$ being the length of the inclined interface:

$$s = y\cos\theta\cos^{1/2}\theta. \tag{21}$$

From equation (18), after some calculations, we have

$$\lim_{N\to\infty} \tilde{P}_N\left(xN, \frac{s}{\cos^{3/2}\theta} N^{1/2}, \theta\right) = \exp\left(-\frac{s^2}{2x\sigma^2(\theta)}\right)\exp\left(-\frac{s^2}{2(1-x)\sigma^2(\theta)}\right) \tag{22}$$

where, taking into account the relation (15), $\sigma(\theta)$ is found to be related to $\gamma(\theta)$ as

$$\sigma^{-2}(\theta) = \beta\left(\gamma(\theta) + \frac{\partial^2\gamma(\theta)}{\partial\theta^2}\right) \tag{23}$$

which is the desired formula.

4. Application to the Ising model

In the preceding sections we have established the formula (6) for general interface orientation. In this section, we apply the formula to the Ising model to see the actual behaviour of $\sigma(\theta)$.

The exact expression of $\gamma(\theta)$ for the square-lattice Ising model with nearest-neighbour coupling constant K is known (Abraham and Reed 1977, Rottman and Wortis

1981, Avron *et al* 1982) to be

$$\beta\gamma(\theta)=\eta_1\cos\theta+\eta_2\sin\theta \tag{24a}$$

$$\eta_1=\sinh^{-1}[\cos\theta(\alpha(\theta))] \tag{24b}$$

$$\eta_2=\sinh^{-1}[\sin\theta(\alpha(\theta))] \tag{24c}$$

$$\alpha(\theta)=M[1-(2/M)^2]^{1/2}\{1+[\sin^2 2\theta+(2/M)^2\cos^2 2\theta]^{1/2}\}^{-1/2} \tag{24d}$$

$$M=\cosh^2 2K/\sinh 2K. \tag{24e}$$

After some algebra, we have

$$\sigma^{-2}(\theta)=\beta(\gamma(\theta)+\partial^2\gamma(\theta)/\partial\theta^2)$$

$$=\frac{\cos\theta\sinh\eta_1+\sin\theta\sinh\eta_2}{\sin^2\theta\cosh\eta_1+\cos^2\theta\cosh\eta_2} \tag{25a}$$

$$=\frac{\alpha(\theta)}{\sin^2\theta(1+\cos^2\theta\alpha^2(\theta))^{1/2}+\cos^2\theta(1+\sin^2\theta\alpha^2(\theta))^{1/2}}. \tag{25b}$$

In particular, for $\theta=0$, we have

$$\sigma^{-2}(0)=\alpha(0)$$

$$=\sinh 2(K-K^*) \tag{26}$$

where K^* is the dual coupling constant given by $K^*=-\frac{1}{2}\log\tanh K$. As was pointed out by FFW, equation (26) agrees with what has been calculated exactly by Abraham and Reed (1976) using the transfer matrix method. The full form of $\sigma(\theta)$ given by (25) is shown in figure 2 for some temperatures. It is clear that, for θ not corresponding to the crystal axis, $\sigma(\theta)$ does not converge to zero even in the $T\to 0$ limit. This 'residual

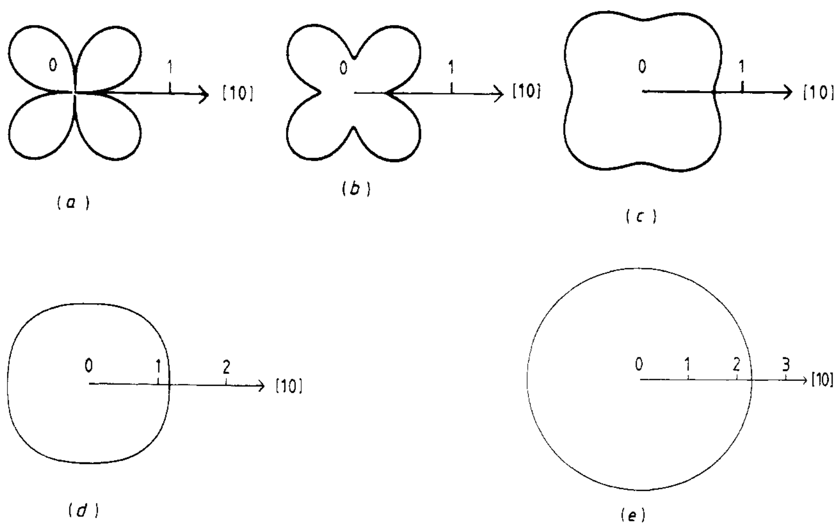

Figure 2. Polar graphs of $\sigma(\theta)$ for the two-dimensional square-lattice Ising model calculated from (25). The temperatures are chosen as (*a*) $T/T_c=0.1$, (*b*) $T/T_c=0.3$, (*c*) $T/T_c=0.5$, (*d*) $T/T_c=0.7$, (*e*) $T/T_c=0.9$.

width' comes from the degeneracy of the ground state configurations of the interface. From the $T \to 0$ limit of the expression (25), we have

$$\sigma^2(\theta) = |\sin\theta|\,|\cos\theta|\,(|\sin\theta| + |\cos\theta|) \qquad (T \to 0) \tag{27}$$

which has a strong anisotropy. Note that the expression (27) is obtained also from the $T \to 0$ limit of $\sigma^2(\theta)$ in the SOS model since the SOS approximation is exact at the $T \to 0$ limit. We can also derive the expression (27) by directly counting the degeneracy of the ground state configurations of the interface. As $T \to T_c$, $\sigma^2(\theta)$ diverges for any θ with the same amplitude of the divergence, reflecting the isotropy of the system in the critical region.

5. Equilibrium shape

In this section, as an application of the formula (6), we present a relation between the scaled interface width and the equilibrium shape. The essential point is the fact that the combination $\gamma(\theta) + \partial^2\gamma(\theta)/\partial\theta^2$ is a constant multiple of the radius of curvature of the equilibrium crystal shape in two dimensions (Wulff 1901, von Laue 1944, Burton *et al* 1951), whose brief derivation, based on the Andreev formulation, is presented in the following.

We draw the equilibrium crystal shape in the xz plane. Andreev (1982) showed that it is determined from the Legendre-transformed free energy $\tilde{f}(\eta)$ defined in (9) via the equation

$$\lambda z = \tilde{f}(-\lambda x) \tag{28}$$

where the parameter λ is the Lagrange multiplier associated with the volume-fixing constraint. Thus the curvature κ is calculated as

$$\begin{aligned}\kappa &= -z''/[1 + (z')^2]^{3/2} \\ &= -\lambda(\partial^2\tilde{f}(\eta)/\partial\eta^2)/[1 + (\partial\tilde{f}(\eta)/\partial\eta)^2]^{3/2}\end{aligned} \tag{29}$$

with $\eta = -\lambda x$. Noting the relations

$$\partial\tilde{f}(\eta)/\partial\eta = -p \tag{30a}$$

$$\partial^2\tilde{f}(\eta)/\partial\eta^2 = -[\partial^2 f(p)/\partial p^2]^{-1} \tag{30b}$$

and rewriting the expressions in terms of the gradient angle θ defined by $p = \tan\theta$, we have

$$\kappa(\theta) = \lambda(\gamma(\theta) + \gamma''(\theta))^{-1} \tag{31}$$

where we have used the relation (11).

By combining the formulae (6) and (31), we see that the curvature $\kappa(\theta)$ of the crystal shape is related to the scaled interface width $\sigma(\theta)$ as

$$\kappa(\theta) = \lambda\beta\sigma^2(\theta). \tag{32}$$

A physically important conclusion from (32) is that, since $\sigma^2(\theta) > 0$ for any θ for $0 < T < T_c$, the equilibrium shape is always convex and cannot be 'dendritic'.

6. Summary and discussion

We have presented a generalised formula which relates the scaled interface width $\sigma(\theta)$ to the anisotropic interface tension for arbitrary interface orientation. We have derived the results both from a thermodynamical argument and from microscopic calculations on the SOS model. We have applied the formula to the Ising model to calculate the explicit form of $\sigma(\theta)$. The formula is also applied to obtain a relation between $\sigma(\theta)$ and the equilibrium shape.

It seems plausible that formulae similar to (6) and (32) hold for a two-dimensional interface in three dimensions. In fact the interface stiffness can be defined for three or more dimensions in the matrix form. The problem is how to relate the stiffness to the interface profile, to which the arguments presented in this paper cannot be applied directly. If we perform the long-wavelength approximation (Weeks 1977, Bedeaux and Weeks 1985) on the problem and adopt the resulting Gaussian Hamiltonian to calculate interfacial quantities, we can derive the generalisation of formulae (6) and (32) for higher space dimensions. Details will be published elsewhere.

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