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THE ROUGHENING TRANSITION

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I.   INTRODUCTION

The idea that there could be a "roughening" of the interface of a crystal in equilibrium with its vapor at a particular temperature T_R was first suggested by Burton and Cabrera (1949) and further developed in a now classic article by Burton, Cabrera and Frank (BCF) (1951). Representing the crystal surface by a two-dimensional (2D) Ising model they suggested that there would be large fluctuations in the surface structure at the Ising model's critical temperature $T_C(2D)$ and a disappearance of the nucleation barrier to crystal growth. Jackson (1958, 1967) further developed and extended these ideas to the case of melt growth and showed that the morphology and growth mechanism of a wide class of crystals could be understood by assuming they were grown above or below the appropriate surface roughening temperature.

Although these ideas were well known to most material scientists and workers in the field of crystal growth, it is only fairly recently that their importance and relevance has been appreciated by condensed matter physicists. The roughening transition is of interest today not only because of its implications for surface physics but also because of its relationship to phase transitions in a number of different systems, several of which are discussed at this Institute.

In these lectures, we will give a brief introduction to the crystal growth models and ideas that lead BCF to suggest the possibility of surface roughening, followed by a review of the modern work relating the roughening transition to phase transitions in a number of 2D systems, including the planar (XY) model, the F

model and the coulomb gas. We describe the application of the
Kosterlitz (1974) renormalization group theory to describe the
statics and dynamics of the roughening transition and the results
of Monte Carlo calculations which seem in good accord with the
theory. More complete discussions and a guide to the literature
of various aspects of these lectures can be found in review arti-
cles by Weeks and Gilmer (1979), Gilmer and Jackson (1977),
Müller-Krumbhaar (1977), and Leamy et al. (1975).

II.   THE SOLID-ON-SOLID MODEL

Consider for simplicity the case of a (001) face of an
impurity-free simple cubic crystal in equilibrium with its vapor.
We model this situation using a restricted version of the usual
lattice gas (Ising model) in which every site is either vacant or
occupied by a single atom whose interaction with another atom in
a nearest neighbor site is ϕ. If we further require that every
occupied site be directly above another occupied site (thus
excluding "overhangs") we obtain a "solid-on-solid" (SOS) model.
A SOS model can thus be thought of as an array of interacting
columns of varying integer heights. The surface configuration is
represented by the 2D array of integers specifying the number of
atoms in each column perpendicular to the (001) face, or equi-
valently by the height of the column relative to the flat $T = 0$
reference surface. Growth or evaporation of the crystal involves
the "surface atoms" at the tops of their columns. As shown in
Fig. 1, complex surfaces with steps and other kinds of disorder
can be represented using the column model.
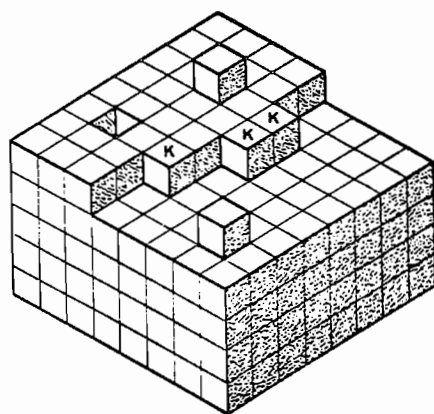

Fig.1 Atoms on a (001) face of a simple cubic crystal. Surface
      atoms may have up to four lateral neighbors. An atom in a
      kink site, indicated by a k in the figure, has two lateral
      neighbors.

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For the restricted lattice gas, the energy between neighboring columns of heights h_j and $h_{j+\delta}$ is determined by counting the number of "broken bonds" and hence is proportional to $\phi|h_j - h_{j+\delta}|$. The equilibrium properties of this particular model (denoted ASOS herein) can be determined from the partition function

$$Z_{ASOS} = \sum_{\{h_j\}=-\infty}^{\infty} \exp\left[-\frac{J}{2kT} \sum_{j,\delta} |h_j - h_{j+\delta}|\right] \tag{1}$$

where $J \equiv \phi/2$ and the summation is over all integer heights for each column. Note that the number of vertical broken bonds is conserved in the excitations permitted in the ASOS model. Hence we can arrive at (1) formally by considering the interface in an anisotropic lattice gas with a vertical bond strength $\phi_\perp$ which tends to infinity (Weeks et al., 1973).

The ASOS model is an accurate approximation to the interface in the unrestricted lattice gas at low T, since the overhanging configurations suppressed are of higher energy. Further we can consider a wider variety of SOS models in which the interaction energy between columns is some increasing function of the height difference $V(h_j - h_{j+\delta})$. At low T, the higher energy multiple height jumps between neighboring columns will be infrequent and the properties of all such models will be very similar. (As will become apparent later, an essential feature of all these models is that the heights can range over all integers - $\infty < h < \infty$.)

Dynamics is introduced into the model by creating or annihilating atoms at random positions on the surface. This process simulates the molecular exchange between the solid and vapor phases. Thus is it reasonable to assume that the rate of creation (deposition) of atoms per site at the surface, denoted k^+, is independent of the neighboring surface configuations. However, the annihilation (evaporation) of a surface atom is an activated process requiring the breaking of nearest neighbor bonds. We assume the evaporation rate of an atom with m lateral neighbors ($0 \leq m \leq 4$ in a cubic lattice) is

$$k_m^- = \nu \exp[-m\phi/kT]$$

where ν is the evaporation rate of an isolated adatom at the surface. It is easy to see that this choice of transition probabilities obeys detailed balance (Gilmer and Bennema, 1972). The equilibrium state, described by Eq (1), is reached when the creation rate, k_{eq}^+, equals the evaporation rate of a kink site, $\nu \exp[-2\phi/kT]$. This must be true since a layer can, in principle, grow (or be removed) by successive creations (or annihilations) of atoms only at kink sites.

We emphasize that the above is a stochastic <u>model</u> for the statics and dynamics of the crystal growth process. Important limitations of the model include the neglect of strain fields, and the need to assume in advance a particular lattice structure. However, the model does give a consistent and physically reasonable description of the cooperative interactions among clusters of atoms that are critical to the crystal growth process. Properly interpreted it thus provides a useful compromise between mathematical simplicity and physical reality.

III. THE BCF ARGUMENT

To gain a qualitative feel for the statics of the roughening transition we make use of the analogy between a lattice gas and a ferromagnetic Ising model, where an occupied site is represented by an "up" spin and a vacant site by a "down" spin. The configuration at $T = 0$ is described by successive 2D layers of up spins representing occupied sites in the crystal followed by layers of down spins representing the vapor. The final (surface) layer of up spins is effectively isolated since the layers above and below are magnetized in opposite directions. Thus following BCF, we might expect the surface layer to behave like a 2D Ising model with large spin fluctuations (i.e., large regions of surface vacancies and adatoms) and thermodynamic singularities near the 2D critical temperature $kT/\phi \cong 0.57$. Note that the cancellation argument holds equally well for the anisotropic lattice gas with $\phi_\perp \gg \phi$. This shows that the roughening transition is not related to the bulk (3D) critical temperature which scales with $\phi_\perp$ (and indeed is infinite for the ASOS model), but rather is a transition unique to the interface.

The BCF picture implies that each crystal face has a distinct roughening temperature, the more loosely packed faces having the lower T_R. Indeed some faces, e.g., the (011) face of a simple cubic crystal, have no connected 2D net of nearest neighbor bonds and their roughening temperature is zero, the result for a 1D Ising model. For most crystal growth applications, the most important faces are the slow-growing close-packed faces.

As one might expect, the critical-like fluctuations occurring at the interface near T_R have an important effect on the crystal growth rate. Crystal growth on a relatively flat surface well below T_R is a difficult process, requiring the formation of a critical nucleus cluster. If a surface is at its roughening temperature, then BCF reasoned that the critical fluctuations produce clusters of arbitrarily large size and hence the nucleation barrier to crystal growth disappears. Another implication is that the crystal grown with $T < T_R$ is faceted with very anisotropic growth rates for the different faces, the close-packed faces growing in a layer-by-layer fashion. Above T_R, essentially isotropic growth should occur (Jackson, 1967).

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It is easy to find fault with this crude argument. There can be an exact cancellation of the interactions from the layers above and below the surface layer only at $T = 0$. Indeed, van Beijeren (1975) used this observation to prove rigorously that $T_C(2D)$ is a lower bound to T_R. Further the restriction of the excitations to only one layer is unrealistic and must be removed for a more exact description. Still, the argument is physically very suggestive and it stimulated experimental work, some of which is described in the next section, which seems in good accord with their physical picture.

IV. EXPERIMENTAL RESULTS

There were initially few attempts to experimentally verify the BCF ideas on roughening because they estimated that a crystal in equilibrium with its vapor would melt before the closest-packed face roughened. However, recent experiments by Jackson and Miller (1977) suggest that for simple van der Waals crystals, the roughening point is well below the melting point. They studied the plastic crystals C_2Cl_6 and NH_4Cl and found dramatic changes in the morphology (faceted to essentially isotropic) of crystals grown for temperatures differing by less than five degrees. These experiments are the only ones we know of in which a crystal in equilibrium with its vapor is taken from below to above its roughening temperature.

Earlier experimental corroboration of the roughening picture involved comparison of a given crystal's structure to predictions arising from estimates of the roughening temperature. Most of this work was for melt growth, and the temperature range over which the crystal growth can be observed experimentally is very small. However, as shown by Jackson (1958, 1967), it is possible to understand both the growth mechanisms (nucleated or continuous) and crystal structure (faceted or isotropic) of a very wide variety of materials by determining whether the crystal as grown was below or above its surface roughening temperature.

These experimental results, and all others we know of, have indirectly observed the roughening transition by its effect on crystal growth. An experimental study of the equilibrium properties of the crystal-vapor interface seems called for. Then one can test a number of the detailed predictions that arise from the new developments in the theory of the roughening transition. These results are discussed in the next part of these lectures.

V. MONTE CARLO CALCULATIONS: QUALITATIVE FEATURES

The BCF one layer model is obviously inadequate in several important respects. To gain a better physical feeling for the roughening transition it is useful to consider the results of Monte Carlo simulations on the ASOS model. Fig. 2 give typical equilibrium surface configurations generated by the MC method at various values of kT/ϕ. At the lower temperature distinct adatom and vacancy clusters are visible but at the highest temperature
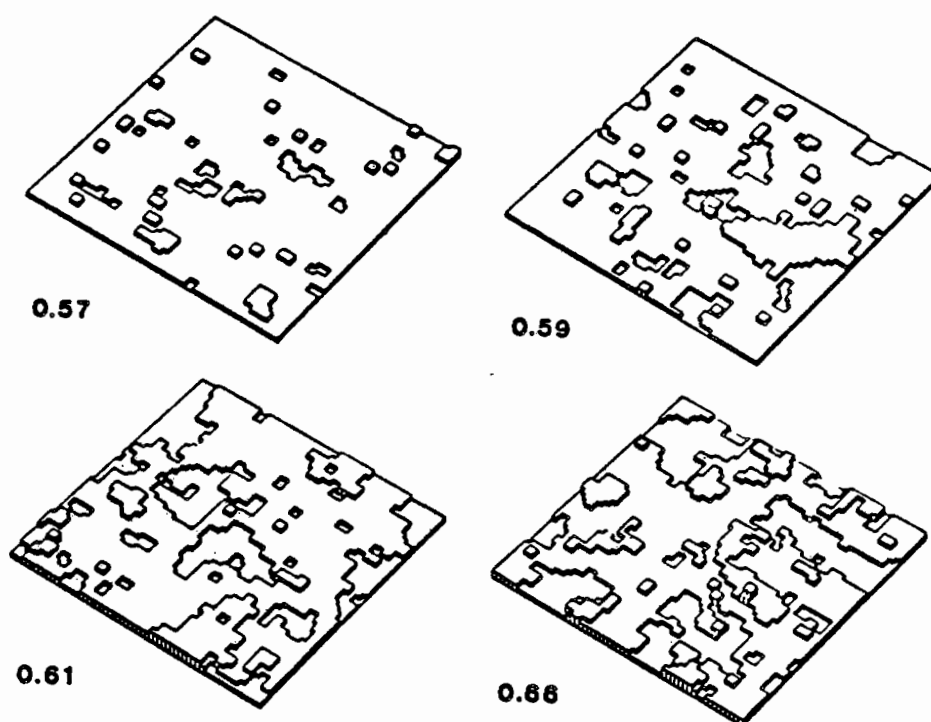
Fig. 2. Computer drawings of typical surfaces generated by the MC
method at the indicated values of kT/ϕ.

the clusters have grown and merged together to such an extent that arbitrarily large clusters are present and indeed the original reference level of the surface is not apparent (Leamy et al., 1975).

Thus the BCF picture of a rapid increase in surface roughness near the 2D Ising model's critical temperature $kT_C(2D)/\phi \simeq .57$ appears confirmed. However an important additional feature becomes clear: the excitations extend over many layers - evidently arbitrarily many in the limit of an infinite system. It is clear that once a large cluster has formed it is just as easy to form another cluster on top of the given cluster as it is to form it in the original layer.

This buildup of large clusters on top of other clusters can be thought of as a long-wavelength distortion of the local position of the solid-vapor interface. Note that there are relatively few double jumps between nearest neighbor columns even at the highest temperature shown. Thus it is the long-wavelength distortions involving a single jump from one large cluster to another that dominate the physics of roughening. The essential idea of BCF is correct: there is a class of low energy excitations possible at the crystal-vapor interface. However these same excitations cause the local position of the interface to wander arbitrarily far from the original reference level.

These considerations suggest there are several equivalent ways of characterizing the roughening transition. Clusters of arbitrarily large size can be found at and above T_R. The formation of these arbitrarily large ridges also implies that the edge free energy and edge energy (per unit length) required to form a step on the crystal surface should vanish at T_R (Leamy and Gilmer, 1974). Since large clusters of adatoms and vacancies are equally probable at T_R, the average density of the surface layer should be 1/2 at and above the roughening temperature. The formation of arbitrarily large clusters in one layer implies a high probability of finding similar large clusters in adjacent layers and the loss of the original reference level. Thus the interface width should diverge at T_R in an infinite system (Weeks et al., 1973). The disappearance of the nucleation barrier implies that the susceptibility (the partial derivative of the average height with respect to an infinitesimal driving force) should diverge at and above T_R. The motion of the interface at and above T_R can be thought of as similar to that of a drumhead, whose normal modes of vibration correspond to the formation of large clusters of adatoms or vacancies on the surface.

VI. STATIC CRITICAL BEHAVIOR

We now make a more formal analysis of the properties of the roughening transition. A literal interpretation of the BCF one layer model suggests that the roughening transition lies in the

same universality class as the 2D Ising model. In fact, because
excitations are not restricted to a single layer, it lies in a very
different universality class. This was first demonstrated in the
work of Chui and Weeks (1976).

Chui and Weeks introduced the discrete Gaussian (DG) model in
which the interactions between nearest neighbor columns is quadratic
in the height difference:

$$H_{DG} = \frac{J}{2} \sum_{j,\delta} \left(h_j - h_{j+\delta}\right)^2 \equiv \frac{J}{2} \sum_{jj'} h_j G_1^{-1}(jj')h_j, \tag{2}$$

$$= \frac{J}{2} \sum_q |h_q|^2 G_1^{-1}(q) \tag{3}$$

where

$$G_1^{-1}(q) = 4 - 2\left(\cos q_x + \cos q_y\right) \tag{4}$$

and

$$h_q = \frac{1}{\sqrt{N}} \sum_j h_j e^{iqj} \tag{5}$$

is the Fourier transform of the height variable h_j. The Fourier
transform of the matrix $G_1^{-1}(jj')$ is explicitly given in Eq. (4).
As argued before, at low T multiple jumps are unimportant and we
expect Eq. (2) to give the same critical behavior as the ASOS model
in Eq. (1). More generally, the roughening transition involves
long wavelength fluctuations in the position of different parts of
the interface. Changes in the interaction energy between columns
that affect only short wavelength properties should be irrelevant
at the roughening point. Furthermore the Gaussian interaction is
in a sense the most fundamental. Note that in Eq. (3) the inter-
action energy for small q goes as $q^2|h_q|^2$. This is characteristic
of a surface tension (Buff et al., 1965). We expect that at high
temperatures the long wavelength properties of the interface arising
from virtually any reasonable microscopic interaction can be de-
scribed using a surface tension. Thus a wide class of microscopic
column hamiltonians should transform under renormalization group
equations to the basic Gaussian interaction as in Eq. (3).

The DG partition function can be written

$$Z_{DG} = \int d\{h_j\} \prod_j W(h_j) \exp\left[-\frac{1}{kT} H_{DG}\right] \tag{6}$$

where

$$W(h_j) = \sum_{n_j=-\infty}^{\infty} \delta(h_j - n_j) \qquad (7)$$

$$= \sum_{k_j=-\infty}^{\infty} \exp\left[ik_j h_j\right] \qquad (8)$$

The <u>weighting function</u> $W(h_j)$ in Eq. (7) restricts the integration in Eq. (6) so that only integer values of h_j contribute. In Eq. (8) we have reexpressed $W(h_j)$ in a more convenient way using a well-known identity (see, e.g., Lighthill, 1959) which is essentially the Poisson summation formula. Here $k_j = 2\pi n$ for integer n. Substituting Eq. (8) into Eq. (6) we have

$$Z_C \equiv \frac{Z_{DG}}{Z_0} = \sum_{\{k_j\}=-\infty}^{\infty} < \exp\left(i \sum_j k_j h_j\right) >_0 \qquad (9)$$

Here Z_0 is the unweighted Gaussian model's partition function [Eq. (6) with $W(h_j) \equiv 1$], which can be evaluated exactly. The angular brackets indicate an ensemble average in the unweighted Gaussian ensemble.

In Eq. (9) we note the characteristic function for the Gaussian distribution. Hence the $\{k_j\}$ also have a Gaussian distribution given by the inverse matrix to G_1^{-1} (see, e.g. Cramer, 1946) and Eq. (9) becomes

$$Z_C = \sum_{\{k_j\}=-\infty}^{\infty} \exp\left[- \frac{kT}{2J} \sum_{jj'} k_j \, G_1\,(jj') \, k_{j'} \right] \qquad (10)$$

where, from Eqs. (2)-(4), the inverse matrix $G_1(jj')$ is

$$G_1\,(j,j') = \frac{1}{2N} \sum_q \frac{e^{iq(j-j')}}{G_1^{-1}\,(q)} \qquad (11)$$

Eq. (10) is in fact the partition function for a neutral 2D lattice Coulomb gas (see Chui and Weeks, 1976 for further details) in which the k_j represents the charges. Note the q^{-2} dependence at small q in Eq. (11) which characterizes the Coulomb interaction.

The reduced temperature kT/J has been inverted in going from the DG
model in Eq. (6) to the Coulomb gas in Eq. (10). The fact that the
Coulomb gas appears is really no mystery: the matrix $G_1^{-1}(jj')$ in
Eq. (2) is the lattice analogue of the Laplacian operator and hence
its matrix inverse, $G_1(jj')$ in Eq. (11), is the 2D lattice Green's
function, i.e., the 2D Coulomb potential.

Since Z_0 is analytic, the singularities in the DG partition
function Z_D are identical with those in Z_C. These had already been
discussed by Kosterlitz and Thouless (1973) and Kosterlitz (1974)
in connection with their analysis of the planar (XY) model and a
dislocation model for 2D melting. They established that the Coulomb
gas has a phase transition from a low temperature dielectric phase
with opposite charges tightly bound together in "diatomic molecules"
to a high temperature metallic phase. The free charges in the
metallic phase come from the now disassociated "molecules" and give
the usual Debye screening. The properties of this transition can
thus be directly related to those of the roughening transition and
differ greatly from those of the 2D Ising model.

The most dramatic differences show up in the behavior of the
correlation length ξ. Define the height-difference correlation
function for two columns separated by a distance r:

$$G(r) = < (h_0 - h_r)^2 > \tag{12}$$

where the angular brackets indicate an ensemble average in the SOS
system. $G(r)$ gives a measure of the average fluctuations in height
between different regions of the interface separated by a distance
r, and the square of the <u>interface width</u> is the $r \to \infty$ limit of
$G(r)$. The <u>correlation length</u> ξ is proportional to the distance r'
at which $G(\overline{r'})$ is approximately equal to its asymptotic value.
The results of Kosterlitz (1974) then imply that below T_R the
interface width is finite with a finite correlation length ξ. At
all temperatures above T_R, however, $G(r)$ is proportional to ln r.
Thus the interface width diverges logarithmically and the correla-
tion length ξ is infinite. It is as if there were a line of
"critical points" for all $T \geq T_R$ where ξ is infinite.

The renormalization group (RG) method of Kosterlitz (1974)
further showed that the correlation length diverges very rapidly
as $T \to T_R$ from below:

$$\xi \propto \exp \left[c/(T_R-T)^{\frac{1}{2}} \right] \tag{13}$$

and of course ξ remains infinite for $T > T_R$. This behavior is very
different from that of the 2D Ising model where ξ diverges by a

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power law only at T_C. Further, the singular part of the free energy has a similar form near T_R:

$$F \propto \exp\left[-C'/\left(|T-T_R|^{\frac{1}{2}}\right)\right] \tag{14}$$

Note the square root dependence on $T-T_R$ in Eqs. (13) and (14). The free energy is non-analytic at T_R but the singularity is a very weak one with all temperature derivatives of the singular part vanishing at T_R. In particular there is no specific heat anomaly at T_R, in contrast to the Ising model. We will discuss the Kosterlitz RG theory and the derivation of some of these results in the next section.

This kind of behavior should apply to a wide class of inter-facial models with different interaction energies between columns. Furthermore the periodic delta function weighting in Eq. (7) can be replaced by other periodic weighting which favor integer positions. A particularly interesting case was analyzed by Ohta and Kawasaki (1978), who took

$$W(h_i) = 1 + 2\ y_o\ \cos 2\pi h_i\ . \tag{15}$$

A Coulomb gas partition function like Eq. (10) again results but the charges k_j now have only the values 0 and $\pm 2\pi$. Using the Kosterlitz RG method, they found critical behavior identical to that described above for the DG model and give further implications for the roughening transition. We discuss in the next section the dynamics of a very similar model.

Another model which has similar critical behavior is the planar model, which Kosterlitz (1974) analyzed by relating it to a 2D coulomb gas. Jose et al., (1977) and Knops (1977) have made the connection between the planar and SOS models quite explicit mathematically by showing they are related by an exact "duality" transformation which we now discuss. Consider a general SOS partition function

$$Z = \sum_{\{h_j\}} \exp\left[-\sum_{j,\delta} V\left(h_j - h_{j+\delta}\right)\right] \tag{16}$$

where $V(h_j - h_{j+\delta})$ is an arbitrary increasing function of the height difference between neighboring columns. If the 2N "bond" variable

$$n_{<ij>} \equiv h_i - h_j \tag{17}$$

(where column i is a nearest neighbor to column j having the smaller x or y coordinate) were all independent then Eq. (16) could be represented as a product of single variable partition functions. Of course there are actually N constraints which the $n_{<ij>}$ variables must obey: around each square of four columns we must have (see Fig. 3)

$$(h_1 - h_2) + (h_2 - h_3) + (h_3 - h_4) + (h_4 - h_1) = 0 \qquad (18)$$

or

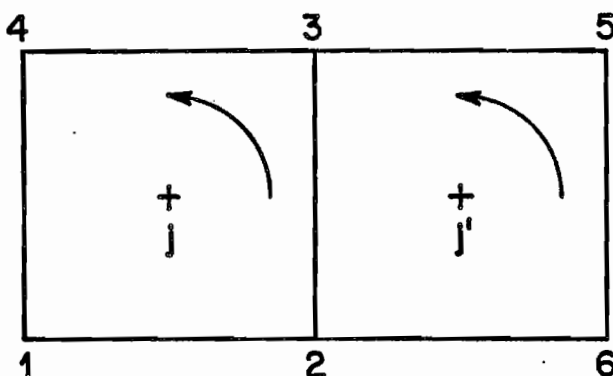$$n_{<12>} + n_{<23>} - n_{<43>} - n_{<14>} = 0 \qquad (19)$$

Fig. 3.   Squares of four columns 1, 2, 3, 4 and 2, 6, 5, 3 and the dual lattice points   j and j' around which the angles ϕ and ϕ' are measured.

We can still introduce the $n_{<ij>}$ variables in Eq. (16) and treat them as independent if we take care of the constraints (19) using the Kronicker δ function:

$$\delta_{0,\Sigma n_{<ij>}} = \int\limits_0^{2\pi} d\phi_j \, \exp\left\{ i\phi_j \left[n_{<12>} + n_{<23>} - n_{<43>} - n_{<14>} \right] \right\} \qquad (20)$$

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The angle ϕ_j can be associated with the center of the jth square of columns, and in general with a point on the dual lattice. We can also index the bond variables using the dual lattice points. For example in Fig. 3, we rename $n_{<23>}$ as $n_{<jj'>}$. Eq. (16) then becomes

$$Z = \sum_{\{n_{<ij>}\}} \int_0^{2\pi} d\{\phi_j\} \exp\left[-\sum_{<jj'>} \left\{V\left(n_{<jj'>}\right)\right.\right.$$
$$\left.\left. + in_{<jj'>}\left(\phi_j - \phi_{j'}\right)\right\}\right]$$
(21)

or

$$Z = \int_0^{2\pi} d\{\phi_j\} \exp\left[-\sum_{<jj'>} \tilde{V}\left(\phi_j - \phi_{j'}\right)\right]$$
(22)

where

$$-\tilde{V}(\phi_j) = \ln\left\{\sum_n \exp\left[-V(n) + in\phi_j\right]\right\}$$
(23)

Eq. (22) is the partition function for a generalized planar model with a 2π periodic angular interaction between neighboring "spins" given by Eq. (23). Just as for the DG to Coulomb gas transformation discussed earlier, the weak and strong coupling limits are interchanged in the transform in Eq. (23). In particular, the DG model transforms exactly into the planar model of Villain (1975) and the usual planar model with $\tilde{V}(\theta) = \frac{J}{kT}\cos\theta$ transforms to a new SOS model with

$$V(n) = -\ln I_n\left(\frac{J}{kT}\right)$$
(24)

Near T_R, the energy for multiple height jumps given in Eq. (24) is bracketed by that given by the ASOS and DG models while at low (planar model) temperatures (i.e., high SOS temperatures), Eq. (24) reduces to the Gaussian model. If these general SOS models are in the same universality class as we have argued, then the corresponding planar models are also.

Another model which almost certainly is in the same universality class is the F model, for which an exact solution is known (Lieb, 1967). We will discuss this model later, after a study of the dynamics of the roughening transition using the Kosterlitz RG method.

VII.   ROUGHENING DYNAMICS AND THE KOSTERLITZ RENORMALIZATION GROUP METHOD

In this section we review a theory of crystal growth dynamics near the roughening point introduced by Chui and Weeks (1978). We are thus dealing with the interesting transition between sub-linear (nucleated) growth below T_R and continuous growth above. We assume some familiarity with recent developments in the theory of dynamic critical phenomena. (See, e.g., Hohenberg and Halperin, 1977.)

The fundamental idea in developing a tractable theory for dynamics at the roughening point is that of dynamic universality (Hohenberg and Halperin, 1977). It is postulated that in addition to all the properties that affect the static roughening behavior, one need consider in addition only the (hydrodynamic) conservation laws and coupling between the conserved variables. Details of the dynamics which do not affect conservation laws are irrelevant for a description of the long-wavelength low-frequency behavior of the system at the roughening point. For example, systems with and without surface diffusion should exhibit similar behavior at their respective roughening transitions.

Our model for crystal growth is particularly simple since there are no conserved quantities such as the energy or momentum density to consider. We have postulated from the first a stochastic and purely relaxational model of crystal growth. Assuming dynamic universality we can thus study, for example, a simple relaxational Langevin model kinetic equation ("Model A") and obtain information about all members of this universality class.

We consider the following generalized SOS model Hamiltonian for the crystal-vapor system

$$H = \frac{J}{2} \sum_{j,\delta} \left(h_j - h_{j+\delta} \right)^2 + Jg^2 \sum_j h_j^2 - \sum_j \Delta\mu_j h_j$$
$$- 2y_0 \, J \sum_j \cos \left(2\pi h_j \right)$$

(25)

The first term gives the interaction energy between a column at site j (and height h_j) and its nearest neighbors at sites $j + \delta$, while the second gives the interaction with a dimensionless "stabilizing field" g^2 which tends to localize the interface near $< h > = 0$. Usually we consider the limit $g^2 \to 0^+$. The third term gives the interaction with "applied fields" $\Delta\mu_j$ which for generality can be different for different lattice sites. We will later associate $\Delta\mu_j$ with the chemical potential driving force for crystal

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growth. The last term, parameterized by the dimensionless quantity y_0 is a <u>weighting function</u> which energetically favors integer values of the h_j. For small y_0 it reduces to the Ohta and Kawasaki (1976) function, Eq. (15). Models similar to Eq. (25) have also been studied by Muller-Krümbhaar (1977), Saito (1978), and Zittartz (1978).

We introduce dynamics through the Langevin equation (See, e.g., Ma, 1976)

$$\frac{\partial h_j}{\partial t} = - \frac{\Gamma}{T} \frac{\delta H}{\delta h_j} + \eta_j$$

$$= - \Gamma K^{-1} \sum_\delta \left(h_j - h_{j+\delta} \right) - \Gamma K^{-1} g^2 h_j + \Gamma \left(\Delta\mu_j / T \right)$$

$$- 2\pi K^{-1} \Gamma y_0 \sin 2\pi h_j + \eta_j \qquad (26)$$

Here $K^{-1} \equiv 2J/T$. (We set Boltzmann's constant equal to unity in this section.) The η_j are Gaussian fluctuating white noises which satisfy

$$< \eta_j(t) > = 0$$

$$< \eta_j(t)\eta_{j'}(t') > = 2\Gamma \delta_{jj'} \delta(t-t') \qquad (27)$$

where the angular brackets indicate an ensemble average. The parameter Γ can be identified with the equilibrium (kink-site) evaporation rate (Weeks and Gilmer, 1979). We assume that the system starts from equilibrium at $t = -\infty$ and allow the applied fields $\Delta\mu_j$ to be time dependent.

If $y_0 = 0$, the Eq. (26) is a linear equation and can be solved exactly by Fourier transform methods in terms of a lattice Green's function which in the long wavelength limit has the form (de Gennes, 1971)

$$G(q,\omega) = \left[K^{-1}\left(q^2 + g^2 \right) - i(\omega/\Gamma) \right]^{-1} \qquad (28)$$

In the limit $g^2 \rightarrow 0^+$, which we consider hereafter, G is the Green's function for 2d diffusion. This, of course, is not surprising since when $y_0 = 0$, Eq. (26) is a finite difference analog of the diffusion equation. $G(q,\omega = 0)$ is proportional to the static Green's function in Eq. (11).

For non-zero y_0, Eq. (25) can be rewritten after taking Fourier transforms as

$$h(q,\omega) = G(q,\omega)\left[\Delta u(q,\omega) + \eta(q,\omega)/\Gamma - 2\pi K^{-1}y_0 \, F\{\sin 2\pi h(s,t)\}\right] \quad (29)$$

Here s is a dimensionless 2d lattice vector (the unit of length being the lattice spacing) locating the center of a column, and F{ } indicates a Fourier transform in space and time.

We will analyze Eq. (29) using linear response theory, assuming that the driving force $\Delta\mu$ is infinitesimally small. Hence we will try to predict the limiting slope of the growth rate curve as the driving force tends to zero. In addition, the linear response analysis gives valuable information about spatial and temporal correlations of the interface at equilibrium when $\Delta\mu = 0$ (Hohenberg and Halperin, 1977).

Expanding the solution of Eq. (27) in powers of $\Delta\mu/T$

$$h(q,\omega) = h_0(q,\omega) + h_1(q,\omega)\Delta u(q,\omega)/T + 0\left(\frac{\Delta u}{T}\right)^2 \quad (30)$$

the linear response function $\chi(q,\omega)$ is given by the ensemble average over the noise

$$\chi(q,\omega) = < h_1(q,\omega) > \quad (31)$$

and using Eqs. (29)-(31), the unperturbed ($y_0 = 0$) response function explicitly is

$$\chi_0(q,\omega) = G(q,\omega) = \left[K^{-1}\left(q^2 + g^2\right) - i(\omega/\Gamma)\right]^{-1} \quad (32)$$

The effect of a non-zero y_0 is conveniently expressed in terms of a self-energy $\Sigma(q,\omega)$, defined as

$$\chi^{-1}(q,\omega) = \chi_0^{-1}(q,\omega) + \Sigma(q,\omega) \quad (33)$$

Substituting Eq. (30) into Eq. (29) we find after some simple manipulation a formally exact expression for Σ given by

$$\Sigma(q,\omega) = \frac{4\pi^2 y_0 K^{-1} F \left\{< \cos\left[2\pi h_0(st)\right]h_1(st,s't') >\right\}}{< h_1(q,\omega) >} \quad (34)$$

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Note that the term transformed is a function only of the differences
s-s' and t-t' since the noise ensemble is stationary.

The behavior of Σ in the limit of very low temperatures is
easy to analyze. The equilibrium fluctuations of h_0 are very small
at low temperatures and the weighting function localizes the inter-
face very near $h_0 = 0$. Linearizing the sine term in Eq. (6.2) then
gives a constant value for Σ of

$$\Sigma(q,\omega) \cong 4\pi^2 y_0 K^{-1} \tag{35}$$

Thus from Eq. (33) there is a finite response even in the $q,\omega \to 0$
limits at low temperature.

At high temperatures $(T > T_R)$ the situation is very different.
Here the weighting function has little effect on the system.
Thermal fluctuations are large enough that the interface wanders
arbitrarily far from its $T = 0$ location (this delocalization
characterizes the roughened phase). When $y_0 = 0$, the weighting
function vanishes altogether and the response function can be
calculated exactly. This divergent response function [Eq. (32)]
presumably gives the limiting high temperature behavior of a system
with a finite y_0.

These qualitative arguments can be put on a much firmer basis
by using the renormalization group method of Kosterlitz (1974) and
José et al. (1977). We consider an expansion of the inverse linear
response function $\chi^{-1}(q,\omega)$ in powers of y_0. Similar expansions
have proved very useful in the static limit. The zeroth order term
$[\chi_0^{-1}(q,\omega)]$ gives the limiting $(T \to \infty)$ behavior and the higher order
terms give corrections arising from a non-zero weighting function.
We will use this expansion to derive differential recursion
relations which relate the response in the original system with
parameters K, Γ and y_0 to that in a system with renormalized para-
meters K', Γ' and y_0'. Integration of the recursion relations will,
in fact, provide a connection for all $T \geq T_R$ between the original
system and the exactly solvable system with $y_0 = 0$.

Expanding h_0, h_1 and Σ in powers of y_0, we find, using
Eqs. (29)-(34), after some straightforward but tedious algebra
[much of which can be found in an article by de Gennes (1971)]
that Eq. (33) can be written to lowest order in q and ω as

$$\chi^{-1}(q,\omega) = \left[K^{-1} + \pi^3 K^{-2} y^2 \int_1^\infty ds \ s^{3-2\pi K} \right] q^2$$

$$-i\omega \left[\Gamma^{-1} + \Gamma^{-1} \frac{\pi^4 y^2}{(\pi K-1)} \int_1^\infty ds \ s^{3-2\pi K} \right] + 0(y^4) \quad (36)$$

where $y \equiv y_0 \exp [-Kc]$ and c is a constant approximately equal to $\frac{1}{2}\pi^2$. We can obtain renormalization group equations from (36) by eliminating the short wavelength parts of the integrals. Divide the range of integration of each integral in Eq. (36) into two parts: 1 to b and b to ∞, with $0 < \ln b << 1$ (i.e., b is very close to unity). The small s (short wavelength) parts of the integration can be combined with the original constant term (either K^{-1} or Γ^{-1}) to yield a new parameter value and the large s part of the integration rescaled so that the integrals again run from 1 to ∞. The scale factor is absorbed in a redefined y variable. Eq. (36) can thus be rewritten in exactly the same functional form with K, y, and Γ replaced by $K(1)$, $y(1)$ and $\Gamma(1)$, with $1 \equiv \ln b$. This equivalence implies the differential recursion relations

$$\frac{dK(1)}{d1} = - \pi^3 y^2(1) \quad (37)$$

$$\frac{1}{2} \frac{dy^2(1)}{d1} = - [\pi K(1)-2] y^2(1) \quad (38)$$

$$\frac{d \ln \Gamma(1)}{d1} = - \frac{\pi^4 y^2(1)}{\pi K(1)-1} \quad (39)$$

subject to the boundary conditions $K(1 = 0) = K$, etc.

The first two equations are essentially identical with the static recursion relations found by Jose et al. (1977) and Nelson and Kosterlitz (1977) in their analysis of the planar model and the 2D coulomb gas. We will study them further before discussing the dynamical implications contained in Eq. (39). Defining the variable $x(1) \equiv \pi K(1)-2$, Eq. (37) can be rewritten

$$\frac{1}{2} \frac{dx^2(1)}{d1} = - \pi^4 x(1) y^2(1) \quad (40)$$

and comparing with Eq. (38), we see there is a conserved quantity

$$x^2(1) - \pi^4 y^2(1) = \text{const} = x^2(0) - \pi^4 y^2(0) \tag{41}$$

As long as $x(1) > 0$, Eq. (38) drives $y(1)$ to zero as $1 \to \infty$. This provides a justification for the original expansion in powers of y in this temperature regime. The roughening point can be thought of as the low temperature end point of this line of "critical" points with $y(\infty) = 0$ and at this end point we must have $x(\infty) = 0$ or $K(\infty) = \frac{2}{\pi}$. This value is universal [i.e. independent of the initial value of y and a number of other modifications in the initial hamiltonian that could be envisioned (Nelson and Kosterlitz, 1978)] and should hold for all roughening models. When applied to other systems, this prediction implies a universal jump in the superfluid density of ^{4}He films as T_C is approached from below and the universal value $\eta = \frac{1}{4}$ for the critical exponent describing the decay of correlations at T_C in the planar model (Nelson and Kosterlitz, 1978).

Another universal feature comes from Eq. (41) when we evaluate it at $1 = \infty$ for temperatures greater than T_R. Then $y(\infty) = 0$ and

$$x^2(\infty) = \left[x^2(0) - \pi^4 y^2(0) \right] \qquad T \geq T_R \tag{42}$$

Very near T_R, We can expand the right hand side in a power series about $T-T_R$, noting that the constant term vanishes since at T_R, $x(\infty) = 0$. We get to lowest order

$$x(\infty) = [A(T - T_R)]^{\frac{1}{2}} \qquad |T - T_R| \ll 1 \tag{43}$$

where A is a nonuniversal constant, but the square root cusp is again universal. It has already shown up in Eqs. (13) and (14). Finally a (nonuniversal) estimate of T_R can be found from Eq. (42) when we set $x(\infty) = 0$. Recalling that $y = y_0 \exp\left[-\frac{1}{2}\pi^2 K\right]$ [see Eq. (36)] we have the equation

$$K = \frac{2}{\pi} + \pi^2 y_0 \exp\left[-\frac{1}{2}\pi^2 K\right] \qquad (T = T_R) \tag{44}$$

Setting $y_0 = 1$ to approximate the DG model, we solve Eq. (44) by iteration and find $kT_R/J \simeq 1.45$. As we will see this value is in excellent agreement with Monte Carlo estimates.

Further analysis of the static equations is possible but we now examine the behavior of the dynamical parameter Γ in Eq. (39). Eliminating $y^2(1)$ between Eqs. (37) and (39) and integrating we have

$$\frac{\Gamma(\infty)}{\Gamma} = \frac{\pi K(\infty) - 1}{\pi K - 1} \tag{45}$$

Hence Γ effectively scales with K, whose behavior we have discussed above. Eq. (45) shows that the renormalized Γ is reduced from its bare value, but does not vanish along the entire fixed line of critical points which characterizes the roughened phase including the end point at T_R. Using the language of Hohenberg and Halperin, (1977), the dynamics is thus <u>conventional</u>. However, the mutual scaling of K and Γ represents an interesting and somewhat unconventional feature of the model. The calculations given above show that $K(\infty)$ has a square root cusp as $T \to T_R$; thus it should be possible to observe a similar anomaly in $\Gamma(\infty)$.

These results have several immediate consequences for the static and dynamic behavior of the crystal-vapor interface. For example, the average growth rate R of a crystal is related to the response to a spatially and temporally uniform driving force when the stabilizing field $g^2 = 0$. To first order in $\Delta\mu$ it is given by

$$R = \lim_{\omega \to 0} - i\omega\chi(q=0,\omega)\frac{\Delta\mu}{T} \tag{46}$$

$$= \Gamma(\infty) \frac{\Delta\mu}{T} \qquad (T \geq T_R) \tag{47}$$

Thus the theory predicts linear growth at and above T_R in agreement with conventional theories of crystal growth.

Below T_R the situation is very different. Approaching the roughening temperature from below, the response function has the limiting form

$$\chi(q,\omega) = \left[K'\left(q^2 + \xi^{-2}\right) - i(\omega/\Gamma')\right]^{-1} \tag{48}$$

with a finite correlation length ξ and renormalized coefficients K' and Γ'. Eq. (46) then predicts a zero growth rate for $T < T_R$ to first order in $\Delta\mu/T$. This result is consistent with the fact that growth at low temperatures occurs by a nucleation mechanism. Nucleation theory gives the result $R \propto \exp(-c/\Delta\mu)$, so in fact below T_R all terms in a power series about $\Delta\mu = 0$ should vanish.

This change in growth mechanisms is directly related to the change in the <u>equilibrium</u> spatial and temporal correlations between different parts of the interface. The height-height correlation

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$$< |h_0(q,\omega)|^2 > = \frac{2}{\omega} \, Im[\chi(q,\omega)] \tag{49}$$

where Im [ ] denotes the imaginary part. In particular, for $T \gtrsim T_R$ and large s or large t,

$$< [h_0(s,t) = h_0(0,0)]^2 > \overset{\sim}{\cong} \frac{K(\infty)}{2\pi} \, \ln \left\{ max \left[s^2, \frac{4\Gamma(\infty)t}{K(\infty)} \right] \right\} \tag{50}$$

where we have used some results of de Gennes (1971). Thus there are logarithmically diverging correlations in space and time above T_R. Note that the coefficient of the logarithm involves only the renormalized coupling constant $K(\infty)$. As discussed before, the RG theory predicts that $K(\infty)$ takes on the universal value $\frac{2}{\pi}$ and from Eq. (43) that there is a square root cusp near T_R. Thus an accurate determination of the equal time correlation function $G(r)$ in Eq. (12) [i.e., Eq. (50) with $t = 0$] provides a direct test of these universal predictions.

The large distance limiting value of the equal time correlation function gives a measure of the interface width. Eq. (50) shows that the interface width diverges logarithmically for $T \gtrsim T_R$. Similar remarks apply to the temporal correlations. Eq. (50) also implies that the correlation length ξ is infinite for all $T \gtrsim T_R$.

Below T_R, Eq. (48) holds and the correlation functions reach finite asymptotic values exponentially fast. In particular, the interface width is finite below T_R and there is a finite correlation length. There are many other interesting features of the roughening point which follow from a more careful analysis of the renormalization group equations. (See, e.g., Ohta and Kawasaki, 1978). We will instead discuss an exactly solvable model where the RG predictions can be checked as well as the results of computer simulations, both of which are in accord with the Kosterlitz RG theory.

VIII. THE FSOS MODEL AND MC CALCULATIONS

Van Beijeren (1977) showed that there is a particular roughening model which is isomorphic to the exactly solvable F model. (The F model is a special case of the symmetric six-vertex model in which the two vertices with no net polarization are given the lowest energy, and hence it describes an antiferroelectric system).

Consider a SOS model for the (001) face of a face-centered-cubic crystal with nearest-neighbor interactions between the atoms. At T = 0 nearest neighbor columns in the x and y directions differ by one atom since half the columns terminate in the layer directly below the outermost surface layer. Now constrain the system such that at all temperatures these nearest neighbor columns can differ by at most ± 1 atom. Thus we are completely suppressing the higher energy multiple height jumps between neighboring columns. As argued before, this should have no effect on the critical behavior and we expect that this model, which we call the FSOS model, is in the same universality class as the other SOS models (ASOS, DG, XY, ...) we have been discussing.

Van Beijeren (1977) showed by a simple argument (which we will not reproduce here since it is very clearly presented in the original work) that the allowed column configurations in the FSOS model can be placed in exact correspondence with the vertex configurations of the F model, and hence the two systems are isomorphic. (Van Beijeren actually considered a bcc crystal with next nearest neighbor forces but his argument applies equally well to the nearest neighbor fcc model, which seems more physically realistic.) Thus one can make use of the results for the exact solution of the F model (Lieb, 1967, and Lieb and Wu, 1972) to test the predictions of the RG theory given in the last section.

Van Beijeren showed that there is indeed a roughening transition in the FSOS model and that at T_R the free energy to form a step vanishes. There is no divergence in the specific heat at T_R but the free energy has an essential singularity of exactly the form [Eq. (14)] predicted by the RG theory.

Furthermore we can make use of the very recent results of Youngblood, Axe and McKoy (1979 as discussed in this Institute) to analyze the exact behavior of the height-height correlation function $G(r)$ [see Eq. (12)] in the FSOS model. They find for all $T \geq T_R$, the exact result for large separation r:

$$G(r) \sim \frac{A(T)}{\pi} \ln r \qquad (51)$$

where at T_R, $A(T_R) = \frac{2}{\pi^2}$ and there is a square root cusp as $A(T)$ approaches its value $\frac{2}{\pi}$ at T_R. These exact results are in precise agreement with the universality predictions of the RG theory as discussed after Eq. (50) and provide a dramatic confirmation of the Kosterlitz RG approach. Unlike most other applications of RG methods, where approximate results are obtained for the system of interest from, say, an ε expansion, the Kosterlitz RG method appears exact for the 2D systems it was designed to treat.

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The success of the theory for the FSOS model suggests that one can use the theory to help predict the transition temperature in other SOS models. Shugard, Weeks and Gilmer (1978) performed MC calculations for $G(r)$ in the FSOS model where the $G(r)$ is known exactly and showed that the MC method using a 60×60 system gave a very accurate representation of $G(r)$ until about $r = 12$ where finite size effects became significant. Their data at T_R could be very accurately fit as in Eq. (51) with $A(T_R) = \frac{2}{\pi}$. Thus accurate MC calculations of $G(r)$ are possible despite the problems of finite system sizes and finite run times.

Shugard et al. then calculated $G(r)$ for the ASOS and DG models and determinined T_R by finding the temperature where the best fit to the curve using Eq. (51) gave an $A(T) = 2/\pi$. They found $kT_R/J = 1.24$ for the ASOS model, in good agreement with the series expansion estimates of Weeks et al. (1973). For the DG model, they estimate $kT_R/J = 1.46$, considerably above the unrenormalized value $4/\pi = 1.28$ predicted by the theory of Zittartz (1978), but in excellent agreement with the KT estimate given after Eq. (44). We believe these values are much more accurate than previous estimates by Swendsen (1977), which were based on an assumed divergence in the specific heat. As discussed before, the KT theory predicts no divergence in the specific heat.

Shugard et al., also studied the planar model by simulating the dual SOS model given in Eq. (24). They find $kT_C/J = 0.90$ which agrees fairly well with the series expansion estimates of 0.95 given by Lambreth and Stanley (1975). The data definitely rules out the value $1.1 \sim 1.2$ given by Miyashita et al., (1978), on the basis of direct MC simulations of the planar model and shows the advantage of a simulation using the dual SOS model with its discrete excitations.

Finally we mention that simulations of time-dependent correlation functions give results in excellent agreement with the theory of Chui and Weeks (1978), Eq. (50). In particular, the diffusion-like s^2-t scaling holds for all $T \geq T_R$.

IX. FINAL REMARKS

Since the time of Burton, Cabrera and Frank (1951) there has been considerable progress in our understanding of the nature of the roughening transition. It is in the same universality class as the phase transition in the planar model and the theory of Kosterlitz and Thouless (1973, 1974) provides precise predictions for a number of experimentally accessible properties. A quantitative experimental study of the roughening transition could provide a crucial test of these important theoretical ideas.

There remain some interesting problems for the theorist. Little work has been done on the roughening transition for multi-component systems. Under certain conditions this may be in a different universality class (Knops, 1979, private communication). Also the analysis of the crystal growth rate has only been done using linear response theory. This is inadequate to uncover the details of the disappearance of the nucleation barrier as $T \rightarrow T_R$ from below. A treatment accurate to all orders in the driving force $\Delta\mu$ could give additional insight into nucleation theory. Finally the study of roughening in models more general than the SOS model would be instructive. For example, we believe very strongly that the roughening transition for the interface in the unrestricted 3D lattice gas lies in the same universality class as the restricted SOS models, but there is no rigorous proof. A related question which the existence of a roughening transition brings up is the degree to which interfacial properties such as the interface width can be thought of as intrinsic (independent of system size and external field strength). Some preliminary thoughts on this subject have been given by Widom (1972), and Weeks (1977), but no rigorous analysis has been done.

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