PHYSICAL REVIEW B VOLUME 30, NUMBER 3 1 AUGUST 1984

Renormalization-group analysis of layering transitions in solid films

David A. Huse
AT&T Bell Laboratories, Murray Hill, New Jersey 07974

(Received 24 February 1984)

The layering critical points of a multilayer adsorbed solid film are examined with the use of a renormalization group. The critical temperature $T_{c,n}$, of the nth layering transition will, for large n, be less than the roughening temperature, T_R, of the corresponding interface between bulk phases by an amount proportional to $1/\ln^2 n$. The layering critical points are in the universality class of the two-dimensional Ising model.

I. INTRODUCTION

Multilayer films adsorbed on attractive substrates may exhibit a variety of possible phase transitions, as has been recently reviewed by Pandit, Schick, and Wortis.[1] One type of transition is the layering transition, in which the thickness of a typically solid film increases discontinuously by one layer as the pressure is increased. Such transitions have been observed in a variety of systems[1] including, for example, ^{4}He (Refs. 2 and 3) and ethylene (Refs. 4 and 5) adsorbed on graphite. The nth layering transition, from an $(n-1)$-layer film to an n-layer film, typically is only present at low temperatures[1] and may terminate as the temperature increases in a *layering critical point* at $T_{c,n}$. For $T > T_{c,n}$ the film may evolve continuously from $n-1$ to n layers as the pressure is increased. A possible phase diagram with such layering transitions and layering critical points is shown in Fig. 1. Ramesh and co-workers[2,3] have explored the phase diagram of solid ^{4}He films adsorbed on graphite (in a Grafoil superleak) using fourth sound as a probe and found results qualitatively similar to Fig. 1. In particular, they have measured $T_{c,n}$ for $n=4$–8 and found that these critical temperatures[3] increase with n.

A simple lattice-gas model with layering transitions and critical points has been introduced and studied in the mean-field approximation by de Oliveira and Griffiths.[6] They proposed that the layering critical points are related to the roughening transition of the corresponding interface between bulk phases. Ebner[7] then carried out Monte Carlo simulations of such a lattice-gas model and found layering critical temperatures $T_{c,n}$ increasing with layer number and approaching an apparent roughening temperature T_R for large n. This relationship between layering criticality and roughening was demonstrated explicitly by Weeks[8] using not a lattice-gas model, but an even simpler interfacial Hamiltonian in which the only degree of freedom is the height $h(\vec{r})$ of the film-fluid interface above the substrate. Such interfacial Hamiltonians, of the form

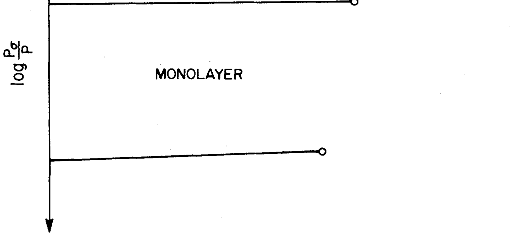

FIG. 1. Possible phase diagram for an adsorbed film with layering transition lines terminating in layering critical points (open circles). The bulk fluid-to-solid phase transition occurs at pressure $P=P_\sigma(T)$ and the roughening transition of the interface separating bulk fluid and solid phases occurs at T_R (solid circle).

$$H=\int d^2r \left[\frac{J}{2} \, |\vec{\nabla} h(\vec{r})|^2 + V(h(\vec{r})) \right] , \tag{1.1}$$

in a continuum representation (of course, lattice versions are also used), have proven important in understanding the roughening transition.[9–12] However, the calculation of the universal critical behavior at the roughening transition has generally[12,13] relied on a duality transformation to the Coulomb gas,[9,10,12] a transformation that is only possible when the potential $V(h)$ is a sufficiently well-behaved function. The duality transformation does not appear to be useful for the type of potential needed to realistically model the effect of a nearby substrate.[14]

In this paper renormalization-group techniques are applied directly to the interfacial Hamiltonian (1.1). This has recently been done for the critical wetting problem by Brézin *et al.*[15] An expansion to first order in the potential $V(h)$ was sufficient for that problem[15] but for roughening and layering criticality it is necesary to go to second order in the potential, as in Sec. III. The results for the layering critical points are qualitatively similar to those obtained by Weeks[8] using a variational approximation, but the precise critical behavior is actually somewhat different from that found in the variational approximation. In particular, the layering critical temperatures behave as

$$T_R - T_{c,n} \sim 1/\ln^2 n \tag{1.2}$$

for large n. This result holds for substrate potentials that fall off algebraically with distance, such as those that arise from van der Waals forces. The variational approximation[8] gives $T_R - T_{c,n} \sim 1/\ln n$. If one inverts (1.2), one finds an essential singularity typical of the roughening transition,[9–13] namely

$$n \sim \exp[B/(T_R - T_{c,n})^{1/2}]\,, \tag{1.3}$$

where B is a nonuniversal constant.

Solid films of arbitrarily large thickness n will generally not be thermodynamically stable, due to elastic contributions to the total energy,[16] and therefore the precise asymptotic behavior of (1.2) may not be confirmable experimentally. On the other hand, the qualitative behavior $T_{c,n} < T_{c,n+1} < T_R$, which should apply to all sufficiently thick films, has already been observed in ^{4}He on graphite.[3] Another theoretical prediction that might be checked experimentally is that each layering critical point is in the universality class of the two-dimensional Ising model.[8]

II. MODEL

Interfacial Hamiltonians such as (1.1) have been used in studies of roughening,[9–13] layering,[8] and wetting.[15] They are useful in problems where the interface undergoes phase transitions while the bulk phases it separates do not.[17] The simple form (1.1) is a coarse-grained Hamiltonian appropriate for modeling universal long-wavelength properties of the interface;[8] terms of the form $|\vec{\nabla} h|^4$, $(\nabla^2 h)^2$, etc., that determine nonuniversal, shorter-distance behavior have been ignored.

The potential $V(h)$ in our interfacial Hamiltonian (1.1) is the total free energy per unit area of a film of thickness h as obtained by integrating out all degrees of freedom *except* translations of the interface.[8] There are contributions to this total that are intrinsic to the film, as well as others that arise from interactions with the substrate. The solid phase, of which the film is comprised, has a free energy per unit volume that exceeds that of the fluid phase by Δf; this will give a contribution of $\Delta f h$ to $V(h)$. The lattice structure of the solid causes a preference for films that are an integral number of layers thick. Fractional layers cost more energy due to strains, dislocations, or steps. Thus, $V(h)$ will also have a component that is oscillatory with h; this term gives rise to the roughening transition in the interface between bulk phases.[9–13] Finally, the van der Waals attractions to the substrate fall off as z^{-3}, where z is the distance from the substrate. These must be integrated over the film to give a contribution to $V(h)$ that varies as h^{-2} for large h. Thus the full potential we consider is essentially the same as that used by Weeks,[8] namely

$$V(h) = \Delta f\, h + c h^{-2} + y \cos(2\pi h) + V_0\,. \tag{2.1}$$

Note that the layer spacing has been taken as unity and higher harmonics in the oscillatory term have been ignored (they are not a factor in determining universal critical behavior).

At low temperatures fluctuations in the film thickness, h, are suppressed and the equilibrium film thickness is determined by the global minimum of the potential $V(h)$. If the oscillatory term y is relatively strong, then $V(h)$ will have local minima only near integral film thicknesses, as illustrated in Fig. 2. As, say, the pressure and consequently Δf are varied, the global minimum of $V(h)$ can change discontinuously. These discontinuities in the film thickness are the layering transitions shown in Fig. 1, where $\Delta f \sim \ln(P_\sigma/P)$ for small Δf. The potential shown in Fig. 2 is for a pressure and temperature near the transition from a three-layer to a four-layer film. If we focus on this transition and assume we are looking at sufficiently large length scales that fluctuations to $h=2$ or 5 are very rare and unimportant, then we may approximate $V(h)$ as $V(h) \simeq -g\tilde{h}^2 + u\tilde{h}^4$, with $\tilde{h} = h - 3.5$. This is simply the potential one inserts in (1.1) to obtain a continuous-spin Ising model.[18] This illustrates a correspondence between the layering-transition line and the first-order line in a two-dimensional Ising model and thus is why we expect each layering-transition line to terminate, as the temperature is increased, in a critical point of the Ising universality class.[8]

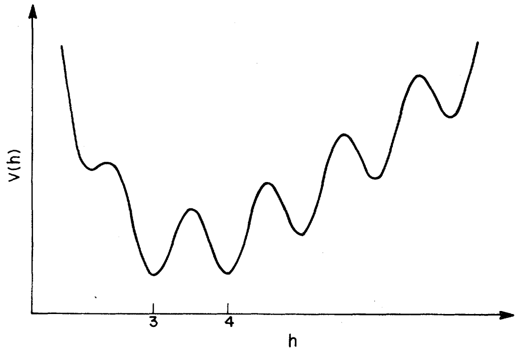

FIG. 2. Potential $V(h)$ in our interfacial Hamiltonian (1.1) for a pressure and temperature near the fourth layering transition. Neglecting fluctuations, the three and four-layer films are nearly degenerate.

As a brief aside, let us consider the possibility of layering transitions in liquid films. Sutton *et al.*[4] have claimed to observe such transitions in ethylene on graphite from two to three and three to four layers, although the evidence is slight. For a fluid film, the smooth part of the potential $V(h)$ will be qualitatively similar to that for the solid film, (2.1), but the oscillatory part will certainly not be present for thick films. However, at low temperatures the fluid in the film will be close packed and highly correlated and presumably has a tendency to form layers within a few atomic distances of the substrate. Thus, the potential $V(h)$ for a fluid film may have an oscillatory component whose amplitude decays exponentially with a correlation length of order one or more atomic spacings. This can give rise to layering transitions in liquid films of very few layers. The first layering transition, from a zero-layer to monolayer fluid film, is simply the condensation of the two-dimensional monolayer liquid and is exhibited by many adsorbate-substrate systems. It will be interesting to see if any further layering transitions in *fluid* films can be experimentally substantiated.

III. RENORMALIZATION

If the potential $V(h)$ vanishes, our interfacial Hamiltonian (1.1) reduces to a Gaussian model,

$$H_0=\frac{J}{2}\int d^2r\,|\vec{\nabla}h(\vec{r})|^2\,, \tag{3.1}$$

for which one can construct exact renormalization-group transformations. The potential may then be treated perturbatively, provided that it is small in the region of interest. In what follows, a renormalization-group transformation for (1.1) is expanded to second order in the potential $V(h)$.

For convenience, we work with a square system with periodic boundary conditions and arbitrarily large area A. The Fourier-transformed height variables are

$$\hat{h}(\vec{k})=A^{-1/2}\int d^2r\,e^{i\vec{k}\cdot\vec{r}}h(\vec{r})\,. \tag{3.2}$$

To avoid ultraviolet divergences we need a short-distance or high-momentum cutoff; a Gaussian cutoff in momentum space is chosen. (Renormalization with a sharp cutoff produces long-range interactions.) This smooth cutoff is such that of the possible degrees of freedom $\hat{h}(\vec{k}\,')$ for $\vec{k}\,'$ near $\vec{k}$, only a fraction $\exp(-k^2/2\Lambda^2)$ are present, where Λ is the cutoff scale. The Gaussian part of the Hamiltonian may then be written as

$$H_0(\Lambda,A)=\frac{J}{2}\sum_{\vec{k}}{}_0^{\Lambda}\,k^2\,|\hat{h}(\vec{k})|^2\,, \tag{3.3}$$

where this sum runs only over those degrees of freedom present with the cutoff scale Λ. The partition function is

$$\begin{aligned}Z(\Lambda,A;H)&=\mathrm{Tr}_{\Lambda}\{e^{-\beta H}\}\\&=\mathrm{Tr}_{\Lambda}\{e^{-\beta H_0}[1-\beta H_1+\tfrac{1}{2}(\beta H_1)^2+\cdots]\}\,,\end{aligned} \tag{3.4}$$

where the interaction part of the Hamiltonian is

$$H_1=\int d^2r\,V(h(\vec{r}))\,. \tag{3.5}$$

To renormalize we must first reduce the cutoff scale to $\Lambda'=\Lambda(1-\epsilon)$, where ϵ is arbitrarily small, by integrating out degrees of freedom with density

$$\Lambda\epsilon\frac{\partial}{\partial\Lambda}\left[\frac{A}{(2\pi)^2}\exp\left[\frac{-k^2}{2\Lambda^2}\right]\right]=\frac{\epsilon k^2A}{(2\pi\Lambda)^2}\exp\left[\frac{-k^2}{2\Lambda^2}\right] \tag{3.6}$$

in momentum space. Then, the system is rescaled by a factor of $1-\epsilon$ and the renormalized Hamiltonian H' is given by

$$Z(\Lambda,A';H')=Z(\Lambda,A;H)\,, \tag{3.7}$$

where $A'=A(1-\epsilon)^2$. The renormalization-group flow equations are given by[18]

$$\dot{H}=\frac{\partial}{\partial\epsilon}H'\,|_{\epsilon=0}\,, \tag{3.8}$$

and thus we need only to expand to linear order in ϵ.

Let us divide $h(\vec{r})$ into the part not to be integrated over,

$$h'(\vec{r})=A^{-1/2}\sum_{\vec{k}}{}_0^{\Lambda}\,e^{-i\vec{k}\cdot\vec{r}}\hat{h}(\vec{k})\,, \tag{3.9}$$

and the part to be integrated out (the "fast" part),

$$h_f(\vec{r})=A^{-1/2}\sum_{\vec{k}}{}_{\Lambda'}^{\Lambda}\,e^{-i\vec{k}\cdot\vec{r}}\hat{h}(\vec{k})\,. \tag{3.10}$$

The momentum-space density of elements in this last sum is given by (3.6). The interaction part of the Hamiltonian may be expanded as

$$\begin{aligned}H_1=\int d^2r\,\Bigg[&V(h'(\vec{r}))+h_f(\vec{r})\frac{dV(h'(\vec{r}))}{dh'(\vec{r})}\\&+\tfrac{1}{2}h_f^2(\vec{r})\frac{d^2V(h'(\vec{r}))}{dh'^2(\vec{r})}+\cdots\Bigg]\,.\end{aligned} \tag{3.11}$$

The expression for the partition function resulting from performing the Gaussian integrals over the fast degrees of freedom is

$$\begin{aligned}Z(\Lambda,A;H)=\mathrm{Tr}_{\Lambda'}\{&\exp[-\beta H_0(\Lambda',A)-\beta\epsilon AE_0']\\&\times[1-\beta\tilde{H}_1+\tfrac{1}{2}(\beta\tilde{H}_1)^2+\cdots]\}\,,\end{aligned} \tag{3.12}$$

where E_0' is just the change in the energy zero, and

$$\begin{aligned}\tilde{H}_1=&\int d^2r\left[V(h'(\vec{r}))+\tfrac{1}{2}A^{-1}\frac{d^2V(h'(\vec{r}))}{dh'^2(\vec{r})}\sum_{\vec{k}}{}_{\Lambda'}^{\Lambda}(\beta Jk^2)^{-1}\right]\\&-\tfrac{1}{2}\beta\int d\vec{r}_1d\vec{r}_2\frac{dV(h'(\vec{r}_1))}{dh'(\vec{r}_1)}\frac{dV(h'(\vec{r}_2))}{dh'(\vec{r}_2)}A^{-1}\sum_{\vec{k}}{}_{\Lambda'}^{\Lambda}e^{-i\vec{k}\cdot(\vec{r}_1-\vec{r}_2)}(\beta Jk^2)^{-1}+\cdots\,.\end{aligned} \tag{3.13}$$

The sums here may be replaced by Gaussian integrals in the thermodynamic limit. Rescaling the system so that $\vec{r}\,'=\vec{r}(1-\epsilon)$ then gives our renormalized Hamiltonian as

$$\begin{aligned}H'=H_0(\Lambda,A')+\epsilon AE_0'+&\int d^2r'\left[V(h'(\vec{r}\,'))+\epsilon\left[2V(h'(\vec{r}\,'))+\frac{1}{4\pi\beta J}\frac{d^2V(h'(\vec{r}\,'))}{dh'^2(\vec{r}\,')}\right]\right]\\&-\frac{\epsilon}{4\pi J}\int d^2r_1'd^2r_2'e^{-(\vec{r}_1-\vec{r}_2)^2\Lambda^2/2}\frac{dV(h'(\vec{r}\,'_1))}{dh'(\vec{r}\,'_1)}\frac{dV(h'(\vec{r}\,'_2))}{dh'(\vec{r}\,'_2)}+\cdots\,.\end{aligned} \tag{3.14}$$

The short-ranged interactions generated at second order in the potential may be Taylor-expanded and integrated out, and, via (3.8), the renormalization-group flow equation is found to be[19]

$$\dot{H} = \int d^2r \left[E_0' + 2V(h(\vec{r})) + \frac{1}{4\pi\beta J}\frac{d^2V(h(\vec{r}))}{dh^2(\vec{r})} - \frac{1}{2J\Lambda^2}\left[\frac{dV(h(\vec{r}))}{dh(\vec{r})}\right]^2 + \frac{1}{4J\Lambda^4}\,|\vec{\nabla}h(\vec{r})|^2\left[\frac{d^2V(h(\vec{r}))}{dh^2(\vec{r})}\right]^2 + O(V^3, V^2(\nabla^2 h)^2)\right] . \tag{3.15}$$

Let us now apply this renormalization group to the potential (2.1) appropriate for modeling a multilayer solid film. More generally, the potential may be divided into a smooth part, $V_s(h)$, reflecting effective interactions between the interface and the substrate, and the oscillatory part, reflecting the interaction between the interface and the lattice structure of the film. Let us expand the smooth part about its minimum at h_0 as

$$V_s(h) = V_s(h_0) + g(h-h_0)^2 + \sum_{n=3}^{\infty} u_n (h-h_0)^n . \tag{3.16}$$

If the potential has the form

$$V_s(h) = V_0 + \Delta f h + c h^{-\alpha} + O(h^{-\beta}) , \tag{3.17}$$

with $0 < \alpha < \beta$, and

$$h_0 \approx (\alpha c/\Delta f)^{1/(\alpha+1)} \tag{3.18}$$

is sufficiently large, then $|u_n| \ll g$. The approximate potential we will then consider is

$$V(h) = g(h-h_0)^2 - y\cos(2\pi h) , \tag{3.19}$$

where the constant and the irrelevant (in the renormalization-group sense of the word) higher harmonics have also been ignored. For large h_0 we have

$$g = g_0 \sim h_0^{-2-\alpha} . \tag{3.20}$$

To first order in g and second order in y the renormalization-group equations (3.15) for this potential are

$$\dot{g} = 2g, \quad \dot{y} = (2-\pi/\beta J)y, \quad \dot{J} = 4\pi^4 y^2/J\Lambda^4 . \tag{3.21}$$

If $g=0$ this is just the roughening problem and the renormalization-group equations are well known.[9,10,12,13] The flow and phase diagram for $g=0$ is illustrated in Fig. 3. The roughening transition and the rough or unpinned (low-βJ or high-temperature) phase are governed by the fixed line $g=y=0$. The lowest-order renormalization-group equations (3.21) are only accurate for sufficiently small g and y. However, even if y is large at some microscopic length scale, it will renormalize to small values provided that βJ is near or below its value at the $g=0$ roughening transition and g is sufficiently small.

Near a substrate, the smooth part of our potential, proportional to g in (3.19), will not vanish. When its minimum, h_0, is not a half-integer, the full potential (3.19) will have a unique global minimum. Under renormalization this minimum will become more pronounced, due to the increase of g, and its location will shift (unless h_0 is an integer). The location of this minimum after renormalization to macroscopic scales determines the film thickness $\langle h \rangle$. As discussed in Sec. II the layering transitions occur when the global minimum of $V(h)$ becomes nonunique. For the present potential, (3.19), such degeneracies only occur when

$$h_0 = \tfrac{1}{2}\ (\mathrm{mod}\,1) . \tag{3.22}$$

Thus, let us now focus on such half-integral values of h_0.[20]

In the smooth phase of the roughening model ($g=0$) the system orders into one of a countable infinity of equivalent ordered states, each with $\langle h \rangle$ an integer. An infinitesimal g breaks this symmetry so that for h_0 a half-integer the system will order in one of only two possible states, with $\langle h \rangle = h_0 \pm \frac{1}{2}$. This ordering is equivalent to that of an Ising model. As g is increased further, these two global minima of $V(h)$ become shallower relative to the local maximum at $h=h_0$ which separates them. Thus increasing g is similar to increasing the temperature in a continuous-spin Ising model. For given values of βJ and y in the $g=0$ ordered or smooth phase, the system will clearly have an Ising critical point at some $g_c(\beta J, y)$, and for $g > g_c$ the system will be disordered, having $\langle h \rangle = h_0$, by symmetry. The schematic location of this Ising critical surface $g_c(\beta J, y)$ is illustrated in Fig. 4 for both $\beta J > \pi/2$ and $\beta J < \pi/2$.

Each layering-transition line in an adsorbate-substrate system is represented in this model by a curve in $(\beta J, g, y)$ space within the ordered phase. The layering critical

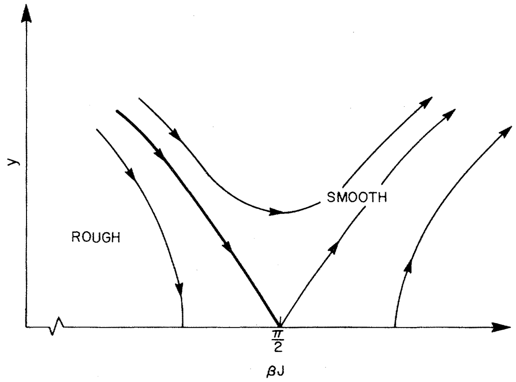

FIG. 3. Renormalization-group flow diagram for the roughening problem ($g=0$). The thick solid line is the roughening transition, separating the high-temperature rough phase from the low-temperature pinned or smooth phase. The rough phase is governed by the fixed line $y=0, \beta J < \pi/2$, while the fixed point $y=0, \beta J = \pi/2$ governs the roughening transition.

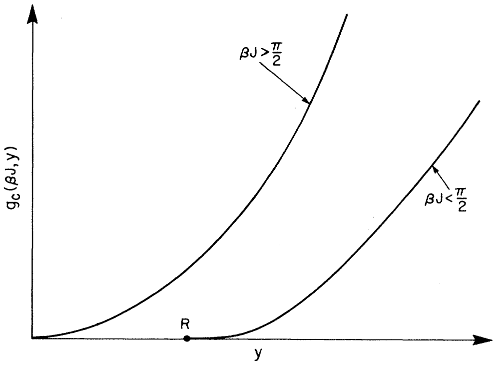

FIG. 4. Schematic location of the manifold of Ising critical points $g_c(\beta J, y)$ for $\beta J > \pi/2$ and $\beta J < \pi/2$. The ordered phase exists for $g < g_c(\beta J, y)$. For $\beta J < \pi/2$ the Ising critical line terminates at the roughening transition point R.

point is the intersection of this line with the surface of Ising critical points, $g_c(\beta J, y)$. The one-phase regions between the layering lines (see Fig. 1) are represented by potentials (3.19) with h_0 not a half-integer. In the limit of thick films the parameters βJ and y will take on values close to those of the free interface (no substrate) and the parameter g will be small by (3.20). For small g and $y > 0$ the surface of Ising critical points, $g_c(\beta J, y)$, goes to the roughening line (thick solid curve in Fig. 3; also see Fig. 4). To obtain some details of the universal behavior expected in this thick-film limit we must now return to the renormalization-group equations (3.21).

The roughening transition is governed by the fixed point $y = 0, \beta J = \pi/2$. Let us expand (3.21) about this point using the variables

$$K = 2 - \pi/\beta J, \quad \tilde{y} = 4\sqrt{2}\beta y/\Lambda^2 , \tag{3.23}$$

and truncate at lowest order, giving

$$\dot{g} = 2g, \quad \dot{\tilde{y}} = K\tilde{y}, \quad \dot{K} = \tilde{y}^2 . \tag{3.24}$$

The renormalization-group trajectory of a system in the thick-film limit (small g), with y and βJ near the $g = 0$ roughening transition, will pass near this fixed point. Thus, at some intermediate length scale, the system is described by g_0, $\tilde{y}_0$, and K_0, where all three parameters are small so we may use (3.24). When parametrized in the usual fashion by l, where $l_0 = 0$ and $\dot{l} = 1$, the renormalization-group flow is (see, e.g., Ref. 10)

$$\begin{aligned} g(l) &= g_0 e^{2l} , \\ \tilde{y}(l) &= A_0 \sec(\psi_0 + A_0 l) , \\ K(l) &= A_0 \tan(\psi_0 + A_0 l) , \end{aligned} \tag{3.25}$$

where

$$A_0^2 = \tilde{y}_0^2 - K_0^2, \quad \psi_0 = \tan^{-1}(K_0/A_0) , \tag{3.26}$$

and $\tilde{y}_0 > |K_0|$ is assumed (this is the case of interest, namely $T < T_R$). The roughening-transition line is $\tilde{y} = -K > 0, g = 0$, and flows into the fixed point $\tilde{y} = K = g = 0$. The parameter A_0^2 measures the deviation from the roughening line and is therefore proportional to $T_R - T$ of the original system.

The surface of Ising critical points $g_c(\tilde{y}, K)$ is an invariant manifold under the flow (3.24). Thus, if one point on a given trajectory (3.25) is critical, the entire trajectory must be critical as well. All of the trajectories with $\tilde{y}_0 > |K_0|$ are asymptotic for $\psi_0 + A_0 l \to \pi/2$ to the plane $\tilde{y} = K$ (see Fig. 3). Let us therefore locate the Ising critical surface near $\tilde{y} = K_f > 0$, where K_f is sufficiently small that our simple expressions (3.25) for the trajectories are still good approximations. The Ising critical surface may be expanded about the point $\tilde{y} = K_f$ as

$$g_c(\tilde{y}, K_f) = g_c(\tilde{y} = K_f) + (\tilde{y} - K_f) g_c^1(\tilde{y} = K_f) + \cdots . \tag{3.27}$$

Trajectories representing systems near roughening will have $A_0 \ll |K_0|, K_f$; let us concentrate on locating such trajectories that are in the critical surface $g_c(\tilde{y}, K)$. The initial point of the trajectory is

$$\psi_0 = -\frac{\pi}{2} + \frac{A_0}{|K_0|} + O\left[\frac{A_0}{K_0}\right]^2 \tag{3.28}$$

and it reaches $K(l) = K_f$ at $l = l_f$, where

$$l_f = \frac{\pi}{A_0} - \frac{1}{|K_0|} - \frac{1}{K_f} + O(A_0) . \tag{3.29}$$

This "final" point of the trajectory is also specified by

$$\begin{aligned} \tilde{y}_f &= K_f + O(A_0/K_f) , \\ g_f &= g_0 \exp\left[\frac{2\pi}{A_0} - \frac{2}{|K_0|} - \frac{2}{K_f}\right][1 + O(A_0)] . \end{aligned} \tag{3.30}$$

This trajectory is in the critical surface only if

$$g_f = g_c(\tilde{y}_f, K_f) , \tag{3.31}$$

or, in the limit of small g_0,

$$A_0 \approx -2\pi/\ln(g_0/\tilde{g}) , \tag{3.32}$$

where

$$\tilde{g} = g_c(\tilde{y} = K_f)\exp\left[\frac{2}{|K_0|} + \frac{2}{K_f}\right] . \tag{3.33}$$

This gives us the form of the Ising critical surface near the roughening-transition line.

Now we must translate this back into the original language of multilayer films to see what we have learned about the layering critical temperatures $T_{c,n}$. The number of layers is $n \approx h_0$, and in the thick-film limit we then have

$$g_0 \sim n^{-2-\alpha} , \tag{3.34}$$

by (3.20). The deviation from the roughening temperature is proportional to A_0^2, so that

$$T_R - T_{c,n} \sim A_0^2 \approx \frac{4\pi^2}{(2+\alpha)^2 \ln^2(n/\tilde{n})} , \tag{3.35}$$

where $\tilde{n}$ is a nonuniversal number determined by the strength of the substrate potential. Note that this result holds for any power-law interaction between the interface and the substrate, with exponent α in (3.17).

IV. DISCUSSION

As mentioned in the Introduction, the limiting functional form of $T_R - T_{c,n}$ as given by (3.35) is not something that one can readily check experimentally. To verify it requires studying a wide range of $\ln n$, which means such *extremely* thick films that it does not appear possible. However, the above renormalization-group treatment makes the simpler, but nontrivial, prediction that for large n, $T_{c,n} < T_{c,n+1} < T_R$. Such behavior has indeed been observed for one system, namely ^{4}He on graphite,[3] and also in a lattice-gas model.[7] Nightingale, Saam, and Schick[21] have independently arrived at the result (3.35).

ACKNOWLEDGMENT

I thank John D. Weeks for useful discussions.

---

[1] R. Pandit, M. Schick, and M. Wortis, Phys. Rev. B **26**, 5112 (1982).

[2] S. Ramesh and J. D. Maynard, Phys. Rev. Lett. **49**, 47 (1982).

[3] S. Ramesh, Q. Zhang, G. Torso, and J. D. Maynard, Phys. Rev. Lett. **52**, 2375 (1984).

[4] M. Sutton, S. G. J. Mochrie, and R. J. Birgeneau, Phys. Rev. Lett. **51**, 407 (1983); S. G. J. Mochrie, M. Sutton, R. J. Birgeneau, D. E. Moncton, and P. M. Horn, Phys. Rev. B **30**, 263 (1984).

[5] S. K. Satija, L. Passell, J. Eckart, W. Ellenson, and H. Patterson, Phys. Rev. Lett. **51**, 411 (1983).

[6] M. J. de Oliveira and R. B. Griffiths, Surf. Sci. **71**, 687 (1978).

[7] C. Ebner, Phys. Rev. A **23**, 1925 (1981).

[8] J. D. Weeks, Phys. Rev. B **26**, 3998 (1982); in *Phase Transformations in Solids,* edited by T. Tsakalakos (Elsevier, New York, 1984).

[9] S. T. Chui and J. D. Weeks, Phys. Rev. B **14**, 4978 (1976).

[10] T. Ohta and K. Kawasaki, Prog. Theor. Phys. **60**, 365 (1978).

[11] Y. Saito, Z. Phys. B **32**, 75 (1978).

[12] For a review of the roughening transition, see J. D. Weeks, in *Ordering in Strongly Fluctuating Condensed Matter Systems,* edited by T. Riste (Plenum, New York, 1980), p. 293.

[13] S. T. Chui and J. D. Weeks [Phys. Rev. Lett. **40**, 733 (1978)] did not use the duality transformation in their renormalization-group study of the dynamics of the roughening transition.

[14] J. T. Chalker [J. Phys. A **15**, 2899 (1982)] has carried through the duality transformation for a very special substrate potential.

[15] E. Brézin, B. I. Halperin, and S. Leibler, Phys. Rev. Lett. **50**, 1387 (1983).

[16] D. A. Huse, Phys. Rev. B **29**, 5031 (1984); Frederick T. Gittes and M. Schick, Phys. Rev. B **30**, 209 (1984).

[17] D. A. Huse, W. van Saarloos, and J. D. Weeks (unpublished).

[18] See, e.g., K. G. Wilson and J. Kogut, Phys. Rep. **12C**, 75 (1974).

[19] The nonlinear terms here, at second order in $V(h)$, are certainly to some extent cutoff dependent. It is not clear what nonlinear terms are universal, if any. I thank Daniel S. Fisher for pointing this out.

[20] The present approximate potential (3.19) is correct only in the limit of very thick films where $|u_n| \ll g$ in the true potential (3.16). Since the "smooth" part of the true potential is not symmetric about $h = h_0$, the layering transitions do not occur precisely at a half-integral h_0. However, in the thick-film limit the asymmetry vanishes and does not alter the asymptotic behavior of the layering critical temperatures (1.2). Furthermore, at each layering critical point the asymmetry may be attributed to the leading "odd" correction-to-scaling field, which is irrelevant at the Ising fixed point and corresponds to the fifth-order term in a Landau expansion. Just as at a liquid-vapor critical point, an Ising-like symmetry that is not present in the microscopic Hamiltonian is attained in the scaling limit.

[21] M. P. Nightingale, W. F. Saam, and M. Schick, Phys. Rev. B (to be published).