Critical Wetting in Systems with Long-Range Forces

In a recent Letter,¹ Nightingale, Saam, and Schick studied wetting transitions in three dimensions in the framework of interface models. The pinning potential for the interface coordinate h was taken as

$$V(h) = \text{const} - Ah^{-2}, \quad h > 0,$$
 (1)

with a hard wall for h < 0. It was found that no depinning transition occurs for (1) and A > 0. A continuous transition was only found as $A \rightarrow 0^+$. Thus, *critical wetting cannot occur for model* (1).¹

The potential (1) is related to the effective interaction between the substrate and the interface. For temperature T = 0, this interaction is given by the difference between the interactions of the adatoms with the real substrate and with a hypothetical

$$F(m) = \int_0^\infty dz \, \{m(z)\chi(z,m) - u(z)m(z) - \ln\cosh[2\chi(z,m)]\},\$$

$$\chi(z,m) = \int_0^\infty dz' \, K(z-z') \, m(z'), \tag{4}$$

K(z-z') is the interaction between the adatoms due to the long-range van der Waals forces after integration over lateral directions. We take K(x) $= (J/T) \{1 + (x/a)^2\}^{-2}$ where J and a are the overall strength and the range of this interaction. The effective substrate potential u(z) in (3) is taken to be

$$u(z) \rightarrow (B/T)z^{-3}, \quad z \rightarrow \infty$$
 (5)

with B > 0.

The model (3)-(5) has a bulk critical point at $T_c = Ja/\pi$. As $T \rightarrow T_c$, the bulk order parameter behaves as $m_{\pm} = \pm M = \pm (3t)^{1/2}$ with $t = 1 - T/T_c$. In the following, "liquid" and "gas" mean m > 0 and m < 0, respectively. The substrate potential (5) can induce a liquid layer of thickness h near the surface while the bulk is still a gas. The free energy of this film is $F(m(z)) - F(m_{-})$ where m(z) solves $\delta F/\delta m = 0$. For large h,

$$\overline{m}(z) = m_{+}\theta(h-z) + m_{-}\theta(z-h)$$
(6)

should be a reasonable approximation for m(z). Thus, the free energy of a thick film should be approximately equal to $F(\overline{m}(z)) - F(m_{-}) = V(h)$. If (6) is inserted into (3)-(5), one finds

$$= \text{const} + (M/T) \{ B - \frac{2}{3} Ja^4 M + O(M^3) \} h^{-2}$$

for large h. Thus, the phase boundary for critical wetting is

$$B^* = \frac{2}{3} Ja^4 M + O(M^3) \rightarrow 2Ja^4 t^{1/2} / \sqrt{3}$$

substrate composed of adatoms.² For general T, however, V(h) is given by the local free-energy density of a rigid interface located a distance h from the substrate. In this Comment, we argue that, for large h, this free energy behaves as

$$V(h) = \text{const} - \{A + A^*(t)\}h^{-2},$$
(2)

where A^* depends on the reduced temperature $t = 1 - T/T_c$. For model (2), critical wetting occurs at $A = -A^*(t)$.³

The potential (2) is obtained as follows. We start from the semi-infinite Ising model defined in Eq. (10) of Ref. 2. Using the standard procedure of a Gaussian transformation,⁴ we obtain a continuum model for the order-parameter field ϕ . Since we will only study mean-field theory, we put $\phi = m(z)$. z is the distance from the substrate surface. This leads to the Landau-Ginzburg free energy

as $T \to T_c$. With A = -MB/T and $A^* = MB^*/T$, one obtains (2).

Finally, note that (2) contains only the leading terms of V(h) for large h. For short-range forces, next-to-leading terms are important in order to determine the complete phase diagram.⁵ They are also necessary in order to explain the results of van der Waals theory for systems with long-range forces⁶ as will be discussed elsewhere.

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³In Ref. 1, a short-range potential was included by $V(h) \equiv 0$ for small h. In this case, critical wetting is found if the constant in (2) is negative near T_c .

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