Pinning of a solid-liquid-vapor interface by stripes of obstacles

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We use a macroscopic Hamiltonian approach to study the pinning of a solid-liquid-vapor contact line on an array of equidistant stripes of obstacles perpendicular to the liquid. We propose an estimate of the density of pinning stripes for which collective pinning of the contact line happens. This estimate is shown to be in good agreement with Langevin equation simulation of the macroscopic Hamiltonian. Finally we introduce a two-dimensional mean field theory that for small strength of the pinning stripes and for small capillary length gives an excellent description of the averaged height of the contact line.

PACS number(s): 68.10.−m, 68.45.Gd, 68.45.Ws

I. INTRODUCTION

The spreading (wetting) of a liquid on a solid is important in a widespread field of practical applications such as lubrication, the efficiency of detergents, oil recovery in a porous medium, and the stability of paint coatings [1]. The motion of the interface is often extremely sensitive to impurities and roughness, which tend to pin (stick) the interface. Different situations arise for the motion of the boundary line between a solid, liquid, and vapor (called the triple line or the contact line) depending on the heterogeneity of the solid and depending on whether the liquid completely wets the solid surface [2] or incompletely wets the solid [3–13]. Due to the presence of a microscopic precursor film that advances ahead of the macroscopic liquid, the case of complete wetting is noticeably insensitive to the heterogeneity of the solid. In the incomplete wetting case obstacles tend to pin the contact line, which makes the statics and dynamics of the contact line highly sensitive to the specific form of the heterogeneity of the solid. The accepted reason for this dependance is the variability of the local wetting angle with roughness or fluctuations in chemical composition. As a consequence of these fluctuations, the contact angles for a proceeding or a receding liquid are not equal, and this hysteresis in the macroscopic wetting angle is the signature of the pinning of the triple line by heterogeneities. Similar kinds of problems are also encountered in other situations where an elastic body is pinned by a random potential [14] such as flux pinning of type-II high-T_c superconductors [15], pinning of charge density waves [16], pinning of magnetic domain walls [17], and dislocation pinning [18].

We have proposed a macroscopic Hamiltonian approach to study the pinning of a solid-liquid-vapor interface when a solid is pulled vertically out of a liquid (called the immersion geometry). We investigate the simple case of the statics of an array of equidistant stripes of obstacles, and we study how the averaged height of the triple line depends on the capillary length and the density and strength of the stripes. Investigating those dependences allows us to determine precisely the conditions for which the interaction of the triple line with the heterogeneities becomes a collective phenomenon. We hope by studying this simple case to understand some of the important physics that is involved in the more complex case of completely random pinning sites.

II. A MACROSCOPIC HAMILTONIAN APPROACH TO THE PINNING OF SOLID-LIQUID-VAPOR INTERFACES

The geometry of the problem is chosen so that the liquid-vapor interface is in the x-y direction and the solid is pulled vertically out of the liquid in the z direction. The origin of the z axis is taken to coincide with the liquid-vapor interface. The local surface energies for the liquid-gas interface γ_{LG}, the solid-liquid interface $\gamma_{SL}(y,z)$, and the solid-gas interface $\gamma_{SG}(y,z)$ determine a local force balance expressed by Young's [19] relation:

$$\gamma_{LS}(y,z) + \gamma_{LG}\cos\theta(y,z) = \gamma_{SG}(y,z). \tag{1}$$

$\theta(y,z)$ is the local macroscopic contact angle between the solid and the liquid.

The Hamiltonian reads

$$\mathcal{H} = \int_0^L \int_0^M dx\,dy\, \frac{1}{2}\rho g h^2(x,y)$$
$$+ \gamma_{LG}\int_0^L \int_0^M dx\,dy\, \sqrt{1+[\vec{\nabla}h(x,y)]^2}$$
$$+ \int_0^M \int_0^{h(0,y)} dy\,dz\, \gamma_{SL}(y,z) + \int_0^M \int_{h(0,y)}^N dy\,dz\, \gamma_{SG}(y,z),$$
$$\tag{2}$$

where ρ is the mass density of the liquid (assumed constant) and g the gravity constant. The first term in this expression is the gravity potential energy, the second term describes the surface energy between the liquid and the gas, and the last two terms account for the surface energies due to contact between the solid wall and the liquid, respectively, gas phase [20].

If one considers how the Hamiltonian changes by a small change in $\delta h(x,y)$, the three parameters $\gamma_{SL}(y,z)$, $\gamma_{SG}(y,z)$, and γ_{LG} that enters the Hamiltonian, can be expressed in terms of just two variables, namely, the liquid-gas surface energy γ_{LG} and the macroscopic contact angle $\theta(y,h(x,y))$ using Young's relation Eq. (1):

$$\frac{\delta \mathscr{H}}{\delta h(x,y)} = \rho g h(x,y) - \gamma_{LG} \nabla^2 h(x,y) \{1+[\vec{\nabla} h(x,y)]^2\}^{-1/2}$$

$$+ \gamma_{LG} \nabla^2 h(x,y) \{1+[\vec{\nabla} h(x,y)]^2\}^{-3/2} [\vec{\nabla} h(x,y)]^2$$

$$- \gamma_{LG} \cos[\theta(y,h(x,y))] \delta(x). \qquad (3)$$

The sole quantity that has the dimensions of a length is called the capillary length and is defined as

$$\kappa \equiv \sqrt{\frac{2\gamma_{LG}}{\rho g}}. \qquad (4)$$

On the length scales for which the Hamiltonian Eq. (2) is supposed to be valid, one can safely ignore fluctuations due to the temperature, so the problem of finding the configuration $h(x,y)$ that minimizes $\mathscr{H}$ is a zero temperature problem.

One way to obtain the equilibrium configuration $h(x,y)$ described by the Hamiltonian Eq. (2) is to perform simulated annealing using Monte Carlo simulation. Notice that since only the *change* of the total energy is needed in a Monte Carlo update, the equilibrium state is completely specified by the three variables ρg, $\theta(y,z)$, and γ_{LG}. In Ref. [4] simulated annealing was carried out for the case $\theta(y,z) = $ const, and for the case of equidistant stripes of obstacles (with contact angle θ') in the z direction. The case $\theta(y,z) = $ const served as a check of the validity of Eq. (2) since the profiles $h(x,y)$ can be directly compared to various analytical results [21]. Initial findings for the case of stripes of obstacles indicated that the averaged height of the triple line, $\langle h(0,y) \rangle$, was linear in the density of pinning sites, c, for small values of c and with a crossover to nonlinear behavior for $c \rightarrow 1$. The density for which the crossover happened, c^*, was an increasing function of $1/\kappa$. This is to be expected, since for small c a given pinning stripe does not feel the presence of the other pinning stripes, and $\langle h(0,y) \rangle$ can be obtained as a simple superposition over all the pinning stripes of the profiles of individual stripes that pin the liquid-vapor interface. On the other hand when c becomes larger the pinning stripes mutually (collectively) lift the liquid, and the resulting profile $h(0,y)$ cannot be obtained as a simple superposition over the pinning stripes. Therefore one expects that collective pinning sets in once the averaged distance between pinning stripes, d, becomes of the order of the capillary length κ, giving $c^* \sim 1/d \sim 1/\kappa$. We show in this paper that another length scale enters the problem, so that the above mentioned argument has to be modified.

It turns out that simulated annealing has the disadvantage of requiring large amounts of computing time, since one needs to perform a slow annealing sequence $T(t)$ [22] in order to bypass metastable configurations. Furthermore, the optimal sequence $T(t)$ depends on the parameters θ, θ', c, κ, and the lattice constant a, and one should in principle determine the optimal sequence $T(t)$ and the optimal step

size Δh for each set of parameter values used in a simulation. In this paper we have instead used Langevin equation simulations of Eq. (3) since it was found to be computationally more efficient than the Monte Carlo simulations. That is, we have numerically solved the equation

$$\frac{\delta h(x,y)}{\delta t} = -\Gamma \frac{\delta \mathscr{F}}{\delta h(x,y)}. \qquad (5)$$

Γ is a mobility constant, t is the time that we let go to infinity in order to find the equilibrium state, and $\mathscr{F}$ is the free energy of the system, which we will assume is $\mathscr{F} \approx \mathscr{H}$, since we neglect fluctuations due to the temperature. In order to take proper account of the boundary condition for the contact of the liquid with the solid plate one has that

$$\left. \frac{\partial h(x,y)}{\partial x} \right|_{x \rightarrow 0} = \cot[\theta(y,h(0,y))]. \qquad (6)$$

This condition has to be introduced via a Langrange multiplyer as an extra term to the Hamiltonian Eq. (2), or equivalently one leaves out the contact terms in the energy Eq. (2) (which amounts to leaving out the term $-\gamma_{LG} \cos[\theta(y,h(x,y))] \delta(x)$ in Eq. (3) and expresses them instead via Eq. (6)).

When discretizing Eq. (5) the functional form of θ for a realization of stripes of obstacles takes the form

$$\theta(y,z) = \delta_{\text{modulus}(y,i),0} \theta' + (1 - \delta_{\text{modulus}(y,i),0}) \theta, \qquad (7)$$

with θ' the value for the contact angle on the stripes of obstacles, θ the value of the contact angle between the stripes of obstacles, and $1 \leq i \leq L/a$ an integer determining the density ($0 \leq c \leq 1$) of pinning stripes. One notices that due to translational invariance in the z direction, there is no pinning force acting when the liquid is pulled vertically out of the liquid, whereas a rotation of the solid about an axis in the x-y plane or a translation of the solid in the y direction produces a pinning force.

III. RESULTS

In Fig. 1 is shown the averaged height of the triple line $\langle h \rangle$ versus the density c for different values of κ from a numerical calculation of Eqs. (5)–(7). The discretization parameter was chosen $a = 0.04$ (the same for all the results represented in this paper), a typical time step $\Delta t \approx 10^{-4} - 10^{-2}$ depending on the values of $\kappa, \theta, \theta', c$; the lattice sizes were chosen between $L/a \times M/a = 100 \times 100$ and $L/a \times M/a = 100 \times 400$ with the larger lattice size for larger κ and the number of time steps to reach equilibrium in the range $2 \times 10^4 - 10^6$. The distance between two pinning stripes, d, is given by the total length of the solid in the y direction, divided by the total number of stripes:

$$d = \frac{L}{cL/a} = \frac{a}{c}. \qquad (8)$$

Therefore the critical density c_κ^* for which collective pinning should set in, given by equating this distance d with the capillary length κ, is
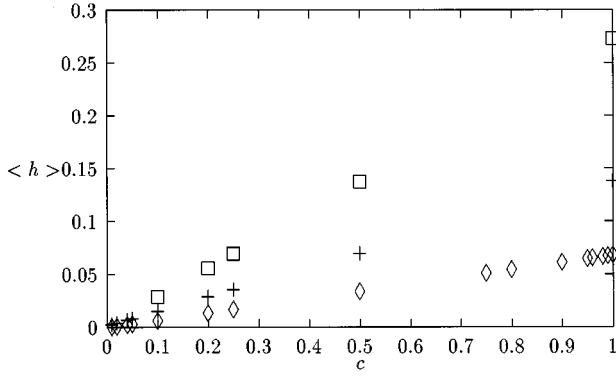
FIG. 1. The averaged height of the triple line $\langle h \rangle$ vs the density c for different values of κ. $\kappa=4$ ($\square$), 2 (+), and 1($\diamond$), respectively. $\theta=\pi/2$ rad. $\theta'=1.5$ rad.

$$c_\kappa^* = \frac{a}{\kappa}. \qquad (9)$$

For the given variables this gives the critical densities $c_\kappa^*=0.01, 0.02, 0.04$. Since $\langle h \rangle$ increases linearly with c for all three values of κ without showing any sign of crossover, we conclude that we are in a single pinning regime, contrary to the simple argument that we suggested above.

The reason for this discrepancy is that the pinning stripes introduce a new length scale in the problem, namely, the distance between the height to which the liquid would rise without pinning stripes $\langle h_0 \rangle$ minus the height to which the liquid would rise with a density of one of pinning stripes, $\langle h_0' \rangle$:

$$H \equiv \langle h_0' \rangle - \langle h_0 \rangle = \kappa[\sqrt{1-\sin(\theta)} - \sqrt{1-\sin(\theta')}]. \quad (10)$$

Therefore if H plays a role in the onset of the single-collective pinning regime one should be able to go from one pinning regime to another by changing *either* $|\theta' - \theta|$ *or* κ. We have confirmed this statement by keeping κ constant and increasing the quantity $|\theta' - \theta|$ for various values of κ (see
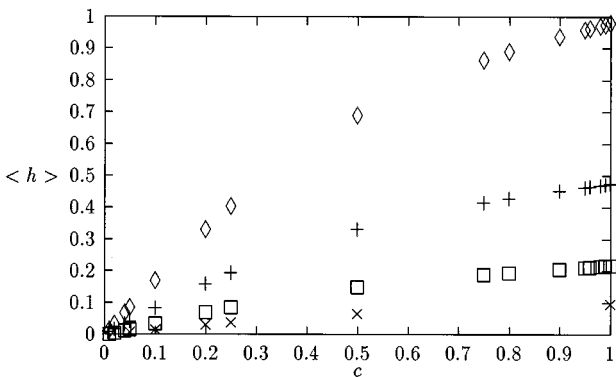

FIG. 2. The averaged height of the triple line $\langle h \rangle$ vs the density c for different values of κ. $\kappa=1$ ($\diamond$), 0.5 (+), 0.25 ($\square$), and 0.125 ($\times$), respectively. $\theta=\pi/2$ rad, $\theta'=0.5$ rad. From Eq. (11) one finds $\widetilde{c}_{\text{curvature}}(\kappa=1.0)=0.15$, $\widetilde{c}_{\text{curvature}}(\kappa=0.5)=0.30$, $\widetilde{c}_{\text{curvature}}(\kappa=0.25)=0.60$, and $\widetilde{c}_{\text{curvature}}(\kappa=0.125)>1$.
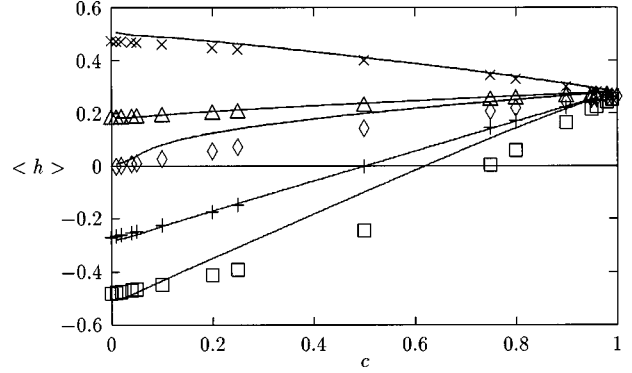

FIG. 3. The averaged height of the triple line $\langle h \rangle$ vs the density c for different values of θ. $\kappa=0.5$, $\theta'=1.0$ rad. $\theta=0.5$ rad ($\times$), 1.2 ($\triangle$), $\pi/2$ ($\diamond$), -0.5 (+), and -1.0 ($\square$), respectively. The solid lines are obtained from the mean field solution Eq. (19). From Eq. (11) one finds $\widetilde{c}_{\text{curvature}}(\theta=0.5)=0.67$, $\widetilde{c}_{\text{curvature}}(\theta=1.2)>1$, $\widetilde{c}_{\text{curvature}}(\theta=\pi/2)=0.55$, $\widetilde{c}_{\text{curvature}}(\theta=-1.0)=0.23$, and $\widetilde{c}_{\text{curvature}}(\theta=-0.5)=0.27$.

the discussion after Fig. 3). Furthermore, in Fig. 2 is shown the averaged height of the triple line $\langle h \rangle$ versus the density c for different values of κ for a fixed value of $|\theta' - \theta|$. One notices a crossover from a single pinning regime to a collective pinning regime as one increases c, and with the crossover appearing for smaller c the larger the value of κ. For given values of κ, θ, θ' and for small c's a stripe does not feel the presence of its neighbor stripes. As c increases the curvature of the triple line between two stripes increases. This enhances the surface area and thereby the surface energy term in Eq. (2). Eventually the curvature becomes so large that the cost in gravitational energy by lifting the liquid between two stripes is outbalanced by the resulting decrease in curvature and thereby surface energy. When this happens the system is in the collective pinning regime. Assuming that the decay of the triple line away from a stripe is exponential [23], we estimate that curvature effects become important when the distance the triple line has decayed after one correlation length becomes on the order of the averaged distance between two stripes:

$$\frac{a}{\widetilde{c}^*} = d^* = H \exp(-1),$$

$$\widetilde{c}^* = \frac{a}{\kappa[\sqrt{1-\sin(\theta)} - \sqrt{1-\sin(\theta')}]\exp(-1)}. \quad (11)$$

Assuming the form of the triple line in between two pinning stripes can be described as a segment of a circle, another way of stating Eq. (11) is to say that collective pinning happens once the radius of curvature between the stripes becomes on the order of the averaged distance between the stripes. In Figs. 2 and 3 we indicate the critical densities $\widetilde{c}^*$ for the various parameters used in the simulation. Giving that Eq. (11) is only based on a simple order of magnitude argument, the agreement with the onset of the collective pinning deduced from the simulations is striking. Besides, using Eq. (11) for the simulations performed in Fig. 1, all give a $\widetilde{c}^*$ larger than one, meaning that collective pinning should not

occur for these values of parameters, which is in agreement with Fig. 1. Based on these results, we conjecture that Eq. (11) should also give a good estimate in the case of random pinning sites, with θ describing the averaged value of the strengths of pinning sites, and θ' the fluctuations about it.

IV. MEAN FIELD THEORY

We now propose a simple mean field theory in order to obtain the averaged height of the triple line, $\langle h_0(c,\kappa,a_0,\theta,\theta')\rangle$, as a function of the density of pinning stripes c. The mean field assumption amounts to considering just one pinning stripe, and finding the minimum energy of this configuration. Furthermore we simplify the problem, and consider only a two-dimensional projection of the three-dimensional liquid meniscus onto the solid; we will assume the functional form for the height h as a function of distance y away from the pinning stripe:

$$y \in [0:a]: h(y) = h_0; y \in [a:d]: h(y) = h_0 \exp\left(-\frac{y}{\kappa}\right) + h_1.$$
(12)

The two constants h_0 and h_1 will be determined from the requirement that the mean field energy E_{mean} of the system attains it minimum. E_{mean} is given by

$$E_{\text{mean}} = \frac{1}{2}\rho g t h_0^2 a - \gamma_{\text{LG}} \cos(\theta') h_0 a$$

$$- \int_a^{a+d} dy\, \gamma_{\text{LG}} \cos(\theta) h_0 \exp\left(-\frac{y}{\kappa}\right) - \gamma_{\text{LG}} \cos(\theta) h_1 d$$

$$+ \int_a^{a+d} dy\, \frac{1}{2}\rho g t h_0^2 \exp\left(-\frac{2y}{\kappa}\right) + \int_a^{a+d} dy\, \frac{1}{2}\rho g t h_1^2$$

$$+ \int_a^{a+d} dy\, \rho g t h_0 h_1 \exp\left(-\frac{y}{\kappa}\right).$$
(13)

The variable t is supposed to take into account the contribution in energy of the three-dimensional meniscus on the two-dimensional projection due to interfacial and gravitational energy. The form of t will be determined by the requirement

$$\langle h_0(c,\kappa,a,\theta,\theta')\rangle \to h_0, c \to 0;$$

$$\langle h_0(c,\kappa,a,\theta,\theta')\rangle \to h_1, c \to 1.$$
(14)

Minimizing the energy

$$\frac{\partial E_{\text{mean}}}{\partial h_0} = 0; \quad \frac{\partial E_{\text{mean}}}{\partial h_1} = 0,$$

$$\rho g t h_0^2 a - \gamma_{\text{LG}} \cos(\theta') a [-\gamma_{\text{LG}} \cos(\theta) + \rho g t h_1]$$

$$\times \kappa \left[\exp\left(-\frac{a}{\kappa}\right) - \exp\left(-\frac{a+d}{\kappa}\right)\right]$$

$$+ \frac{1}{2}\rho g t h_0 \kappa \left[\exp\left(-\frac{2a}{\kappa}\right) - \exp\left(-\frac{2(a+d)}{\kappa}\right)\right] = 0;$$

$$[-\gamma_{\text{LG}} \cos(\theta) d + \rho g t h_0] \kappa \left[\exp\left(-\frac{a}{\kappa}\right) - \exp\left(-\frac{a+d}{\kappa}\right)\right]$$

$$+ \rho g t h_1 d = 0.$$
(15)

Equation (15) is two equations with two unknowns from which h_0, h_1 can be determined. Now $\langle h_0(c,\kappa,a_0,\theta,\theta')\rangle$ is given by the integral over the functional form Eq. (12):

$$(a+d)\langle h_0(c,\kappa,a_0,\theta,\theta')\rangle$$

$$= \int_0^a dx\, h_0 + \int_a^{a+d} dx \left[h_0 \exp\left(-\frac{x}{\kappa}\right) + h_1\right],$$
(16)

which from the solutions of Eq. (15) can be written as

$$\langle h_0(c,\kappa,a_0,\theta,\theta')\rangle = \frac{\kappa^2 \gamma_{\text{LG}} \cos(\theta') ac}{2t\{a - [\kappa^2/a(1/c-1)][\exp(-a/\kappa) - \exp(-a/c\kappa)]^2 + (1/2)\kappa[\exp(-2a/\kappa) - \exp(-2a/c\kappa)]\}}$$

$$+ \frac{\gamma_{\text{LG}} \cos(\theta)(1-c)}{\rho g t}.$$
(17)

In order to fulfill the condition Eq. (14) it can be seen from Eq. (17) that a proper choice of t is

$$t = \sqrt{\frac{\gamma_{\text{LG}}}{2\rho g[c(1-\sin(\theta')) + (1-c)(1-\sin(\theta))]}} [c\cos(\theta') + (1-c)\cos(\theta)].$$
(18)

Thus we end up with the form for $\langle h\rangle$ given by

$$\langle h_0(c,\kappa,a_0,\theta,\theta')\rangle = \frac{\kappa^2 \gamma_{\text{LG}} \cos(\theta') ac}{2t\{a - [\kappa^2/a(1/c-1)][\exp(-a/\kappa) - \exp(-a/c\kappa)]^2 + (1/2)\kappa[\exp(-2a/\kappa) - \exp(-2a/c\kappa)]\}}$$

$$+ \frac{\gamma_{\text{LG}} \cos(\theta)(1-c)}{\rho g t}.$$
(19)

Finally we will assume that the width of the pinning stripes a depends on the density of pinning stripes c in the following way:

$$c \in [0:0.5]: a = a_0; \quad c \in [0.5:1]: a = a_0 L 2(c - 0.5), \quad (20)$$

which just states that no two pinning stripes will be neighbors until $c = 0.5$, after which the width of a stripe increases linearly with c.

In Fig. 3 is shown the averaged height of the triple line $\langle h \rangle$ versus the density c for different values of θ with a fixed value of κ and θ'. One notices that the density for which collective pinning begins clearly depends on θ and is well described by Eq. (11). Furthermore the two-dimensional mean field solution gives a good description of the three-dimensional Langevin solution, with the best agreement for small H in Eq. (10). This is to be expected since for large H the three-dimensional nature of the liquid meniscus becomes more important and the two-dimensional mean field picture breaks down.

In conclusion we have proposed a macroscopic Hamiltonian approach to the pinning of solid-liquid-vapor interfaces due to the presence of stripes of obstacles. We find that curvature effects play a crucial role for the transition from the single pinning regime to the collective pinning regime. We have proposed an estimate of the density of pinning stripes for which the collective pinning happens, which is in good agreement with the simulations of the Langevin equation. We conjecture the same estimate to be valid in the case of random pinning sites. Finally a two-dimensional mean field solution has been introduced that for small values of $|\theta' - \theta|, \kappa$ gives an excellent approximation for the height of the triple line.

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[22] T is the temperature as a function of Monte Carlo time t; in Ref. [4] T was chosen proportional to t, i.e., $T(t) = T_0(1 - t/N)$ with N the total number of Monte Carlo steps and T_0 the initial temperature.

[23] The deformation of the contact line is known to be logarithmic *without* the gravity term in Eq. (2) [3], but since gravity introduces a length scale in the problem (the capillary length) the decay of the contact line becomes exponential [21].