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LETTER TO THE EDITOR

The growth equation of a multi-holes system in partial wetting

Yoshihisa Enomoto

Department of Physics, Faculty of Science, Nagoya University, Nagoya 464-01, Japan

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Abstract. We study the dynamical behaviour of a multi-holes system in partial wetting. Applying the method recently developed by us to the problem, we derive for the first time the equation of motion of the contact lines, where the free surface of the wetting liquid meets the solid surface, in the presence of multi-holes. The equation obtained is found to take into account the cooperative effects generated by the spatial interactions among holes.

Recently Sekimoto *et al* [1] have developed a macroscopic static theory of the morphological stability of partial wetting [2]. The system they considered is the partial wetting of a non-volatile liquid on a solid surface that is rigid and microscopically flat. The free surface of the liquid meets the solid surface at the contact line at a certain contact angle. Sekimoto *et al* first derived an expression for the wetting energy involving the effect of gravity as a functional of the contact line. Using the free energy functional and the approximation of a nearly flat free surface of the liquid, they have found that there are at least two fundamental morphologies, which we call a hole and a ridge, which are thermodynamically unstable against certain infinitesimal deformations of the contact lines. The hole-type instability has also been found by Srolovitz and Safran [3] for the case of thin film rupture where the gravity effect is not so important. Moreover, they have discussed the dynamical behaviour of an array of holes. They have, however, only discussed an ideal system where holes are arranged with arbitrary, but uniform, initial size and spacing.

In the present letter we thus consider a general situation where not only the positions of holes but also their initial radii are randomly distributed. By using a Green function method we obtain the growth equation of the multi-holes system of partial wetting. This method has been developed recently by us to discuss the growth equation of the three-dimensional Ostwald ripening [4] and the multi-nuclei system in a vacuum-deposited thin film [5].

Here we consider a system where the non-volatile liquid covers the whole solid surface, except for disc-shaped domains (called holes) where the solid is exposed. The geometry considered is shown in figure 1. In the following discussion, we assume that: (i) the total volume of the wetting liquid is conserved; (ii) the contact line of each hole is circular with radius $R_j(t)$ $(1 \leqslant j \leqslant N(t))$ where $N(t)$ is the number of holes at time t—moreover, its centre $\boldsymbol{X}_j$ is fixed—and (iii) the mean distances between holes are larger than their radii. (iv) The approximation of a nearly flat liquid surface is made, that is, $|\nabla f(\boldsymbol{r}, t)| \ll 1$ where $f(\boldsymbol{r}, t)$ denotes the liquid height at the position $\boldsymbol{r} = (x, y)$ on the solid

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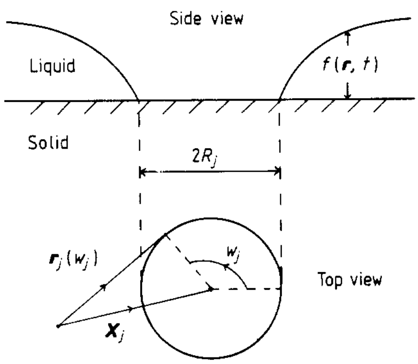

Figure 1. The geometry of the jth hole considered here.

surface, and (v) following Cox [6], the motion of the jth hole's contact line is assumed to be described by [7]

$$(\mathrm{d}/\mathrm{d}t)R_j = s(\theta_e - \theta_j(w_j, t)) \tag{1}$$

where s is a positive constant, θ_e the equilibrium contact angle given by Young [8], and $\theta_j(w_j, t)$ the contact angle at the position of the contact line $\boldsymbol{r}_j(w_j)$, given within assumption (iv) by

$$\theta_j(w_j, t) \equiv -\tan^{-1}(\boldsymbol{n}_j(w_j) \cdot \boldsymbol{\nabla}_j f(\boldsymbol{r}_j, t)) \simeq -\boldsymbol{n}_j(w_j) \cdot \boldsymbol{\nabla}_j f(\boldsymbol{r}_j, t). \tag{2}$$

Here $\boldsymbol{r}_j(w_j)$ and w_j are, respectively, the position and the corresponding angle variable on the jth hole's contact line, as is shown in figure 1. Moreover, $\boldsymbol{\nabla}_j f(\boldsymbol{r}_j, t)$ denotes $\boldsymbol{\nabla} f(\boldsymbol{r}, t)$ evaluated at $\boldsymbol{r} = \boldsymbol{r}_j$, and $\boldsymbol{n}_j(w_j)$ is the outward unit vector normal to the jth contact line. Note that, as can be seen later, the contact angle θ_j is independent of w_j because of assumption (iii) and thus the contact line remains circular during the time evolution.

Under the above assumptions the liquid surface profile $f(\boldsymbol{r}, t)$ is obtained and used to solve the following equation with a Lagrange multiple f_0 [1]:

$$(\nabla^2 - L^2)f(\boldsymbol{r}, t) + L^2 f_0 = 0 \tag{3}$$

with boundary conditions

$$f(\boldsymbol{r}_j, t) = 0 \tag{4}$$

$$f(\boldsymbol{r}, t) \to d(t) \qquad \text{for } |\boldsymbol{r}| \to \infty \tag{5}$$

where $L^{-1} = \sqrt{\gamma/\rho g}$ is the capillary length with γ being the surface energy of the free surface of the liquid, ρ the density of liquid and g the gravitational constant. Despite considering thin film rupture [3], the capillary length L^{-1} and/or the gravity effect g has been found to be important in the partial wetting [1]. Here $d(t)$ denotes the liquid height far from holes and is determined later such that the total liquid volume is conserved.

Equations (3)–(5) are formally analogous to those of [5] and can thus be solved by using the same two-dimensional Green function method. Thus, we shall quote only the final results omitting the intermediate calculations. Therefore, we obtain

$$f(\boldsymbol{r}, t) = d(t) + \sum_j \int \mathrm{d}w_j \, G(\boldsymbol{r} - \boldsymbol{r}_j)c_j(w_j) \tag{6}$$

$$c_j(w_j) = -d(t)\,A(j) - \sum_{i\neq j} A(i)G(X_{ij}) \int \mathrm{d}w_i\, c_i(w_i) \tag{7}$$

$$A(j) = (I_0(J)K_0(J))^{-1} \tag{8}$$

$$X_{ij} = |\boldsymbol{X}_i - \boldsymbol{X}_j| \tag{9}$$

where the argument J of the Bessel functions in equation (8) is defined by $J \equiv LR_j$ and the two-dimensional Green function $G(r)$ satisfying $(\nabla^2 - L^2)G(\boldsymbol{r}) = -\delta(\boldsymbol{r})$ is given by

$$G(\boldsymbol{r}) = (1/2\pi)K_0(L|\boldsymbol{r}|). \tag{10}$$

Then, using (2), (6) and (7), the contact angle $\theta_j(t)$ is obtained from

$$\theta_j(t) = Ld(t)B(jj) - \sum_{i\neq j} B(ij)K_0(LX_{ij})\theta_i(t) \tag{11}$$

with

$$B(ij) = \begin{cases} K_1(J)/K_0(J) & \text{for } i = j \\ I_0(J)K_1(J)/(I_0(I)^2K_0(I)K_1(I)) & \text{for } i \neq j. \end{cases} \tag{12}$$

To obtain these results, we have used the following approximations:

$$G(\boldsymbol{r}_i - \boldsymbol{r}_j) \simeq G(X_{ij}) \tag{13}$$

$$\boldsymbol{n}_j(w_j)\cdot\nabla G(\boldsymbol{r} - \boldsymbol{r}_i)|_{\boldsymbol{r}=\boldsymbol{r}_j} \ll \boldsymbol{n}_j(w_j)\cdot\nabla G(\boldsymbol{r} - \boldsymbol{r}_j')|_{\boldsymbol{r}=\boldsymbol{r}_j}. \tag{14}$$

These approximations may be allowed, because of assumption (iii). Here we remark that due to the above approximations θ_j becomes independent of w_j. The term $\theta_e - Ld(t)B(jj)$ in (1) and (11) is a mean-field term which is analogous to that of [5]. On the other hand, the last term in (11) represents the spatial interactions among holes via the field $f(\boldsymbol{r}, t)$, which result in the statistical correlations beyond the mean-field theory. Such correlations have been found to play important roles in the dynamical behaviour [4].

Finally, the above equations must be supplemented with the conservation law of the liquid volume:

$$Sd_0 = Sd(t) - V_e \tag{15}$$

$$V_e \equiv \iint (d(t) - f(\boldsymbol{r}, t))\,\mathrm{d}^2r = \pi d(t)\sum_j R_j^2 + \iint (d(t) - f(\boldsymbol{r}, t)) \times \prod_j H(|\boldsymbol{r} - \boldsymbol{X}_j| - R_j)\,\mathrm{d}^2r \tag{16}$$

where S is the solid surface area, d_0 the uniform liquid height in the absence of holes, V_e the excluded volume due to holes, and $H(x)$ the usual step function. As a result, we obtain the equation of motion (1) of the contact line with (11) and (15), which is a starting equation for studying the dynamical behaviour of the multi-holes system.

These equations of motion are, however, still difficult to handle. A considerable simplification is obtained if $LX_{ij} \gg LR_j \gg 1$. In this limiting case we have

$$B(jj) \simeq 1 + 1/2LR_j \tag{17}$$

$$K_0(LX_{ij}) \simeq 0 \tag{18}$$

$$V_e \simeq \pi d(t) \sum_j R_j^2. \qquad (19)$$

Then, after some algebraic calculations, equations (1), (11) and (15) reduce to

$$(\mathrm{d}/\mathrm{d}t)R_j = (sd(t)/2)(\Delta(t)/\alpha - 1/R_j) \qquad (20)$$

$$\Delta(t) + \pi \sum_j R_j^2 = Q \qquad (21)$$

with $\Delta(t) \equiv Sd_0(1/d(t) - L/\theta_c)$, $\alpha \equiv Sd_0/2\theta_e$ and $Q \equiv S(1 - Ld_0/\theta_e)$. These simplified equations are analogous to those of the mean-field theory of two-dimensional Ostwald ripening [9], if R_j is regarded as the jth precipitate radius, Q as the total precipitate volume fraction and so on, except for the time-dependent kinetic coefficient $sd(t)$ in (20). Sekimoto has also pointed out such an analogy.

To study the effects of the spatial interactions on the dynamical behaviour of the multi-holes system, we are currently performing molecular dynamics simulation of (1) with (11) and (15) directly. The results, together with a comparison between the present results and those of Srolovitz and Safran, will be published in the future.

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