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Capillary wrinkling scaling laws of floating elastic thin film with a liquid drop

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The deformation patterns of elastic membranes under tension is called wrinkling. Wrinkling, which is caused by capillary surface tension, is called capillary wrinkling (Figure 1).

In recent years wrinkling patterns have drawn particular attention [1-11], since it can be an useful tool to infer material parameters that might otherwise be inaccessible. For example, the commonly observed tearing instability of an elastic sheet, adhered to a rigid substrate, can be used to characterize the adhesion energy. The capillary-driven wrinkle formation can also be used as the basis for a metrology of both the elastic modulus and the thickness of ultrathin films as well as for the study of dynamical relaxation phenomena in ultrathin films [5].

There has also been considerable interest in understanding fundamental aspects of elasto-capillarity [9, 10] and capillary wrinkling, including wrinkling of a compressed elastic film on a viscous layer [1], wrinkling mechanics and the geometry of an elastic sheet under tension [2,3], the size and number of wrinkles [5], the analytic analysis of capillary wrinkling of the circular elastic membranes [6], wrinkling of pressurized elastic shells [7], and capillary buckling of slender rods [11].

Regarding the capillary wrinkling of thin film, a milestone has been laid by Huang et al. [5] and Vella et al. [6]. Huang et al. [5] obtained the expression of capillary wrinkling length ℓ and numbers N by curve-fitting from experimental data, while Vella et al. [6] theoretically proved the capillary wrinkling length expression obtained by Huang et al. [5] for a small deformation of a circular film. A novel experiment, combining both fundamental and applied aspects of the interaction between surface tension and elasticity, was presented by Huang et al. [5]. In their experiment, a small liquid drop was placed onto an elastic membrane that floated in a bath of the same liquid. Before adding the drop, the membrane was stretched by the surface tension of the liquid bath. Once the drop was added, owing to the opposing tension, the contact line of the drop caused radial wrinkles. Experimentally, it was found that the wrinkling length $\ell = 0.031 r\left(\frac{Eh}{\gamma}\right)^{1/2}$, where r is the radius of the drop and γ is the surface tension coefficient of the liquid-gas interface. This pure empirical

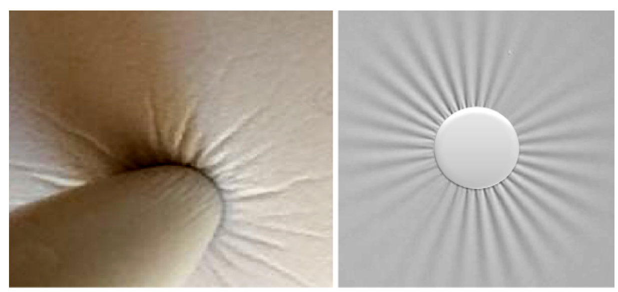
(a) (b)

Figure 1 (Color online) (a) Typical wrinkling of elastic film; (b) typical capillary wrinkling of elastic film. The problem is to find the deformation wrinkling pattern pair (N, ℓ), where the wrinkling number is N and the wrinkling length is ℓ.

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relationship is confirmed by a theoretical justification of Vella et al. [6], who used the Föppl-von Kárman thin plate theory, however, the wrinkling number N expression proposed by Huang et al. [5] has not been theoretically verified yet.

Although Huang et al. [5] successfully obtained the formula for the pair (N, ℓ) by curve-fitting, some questions are still to be answered: are these relationships universally applicable? If not, then under what circumstances can it be approximately valid? How can we promote those to other materials such as axisymmetric anisotropic materials without doing further experiments? How can we extend the pair (N, ℓ) to capillary wrinkling dynamics? If the characteristic scale of the drop is greater than the capillary scale κ^{-1}, gravity begins to dominate the wrinkling process, so what is the scale laws beyond the capillary scale κ^{-1}?

The paper first introduce the research topic before presenting the general expression of the pair (N, ℓ) by using dimensional analysis and determining the controlling parameters of the problem, while proposing linear approximated scaling laws for the pair based on the understanding of the test's data, and then extending to the film made of composite materials. For the spreading of a liquid drop, capillary wrinkling dynamics scaling laws are proposed based on Tanner's law. Finally, the paper concludes with a formulation of wrinkling scaling laws in the gravity regime.

Generally speaking, both the wrinkling number N and length ℓ are functions of the Young modulus E, Poisson ratio ν, film thickness h, radius of liquid drop r and surface tension γ, namely, $N = F(E, h, \nu, \gamma, r)$ and $\ell = G(E, h, \nu, \gamma, r)$, where the F and G are unknown functions. The dimensions of those parameters are listed in Table 1.

Since the film will be in a bending and stretching state with a liquid drop, by dimensional analysis [12-16], the above formula can be further expressed into the following forms: $N = F(D, K, \gamma, r)$ and $\ell = G(D, K, \gamma, r)$, where the bending stiffness $D = Eh^3/[12(1-\nu^2)]$ and the in-plane stiffness $K = Eh/(1-\nu^2)$. They can further be expressed in dimensionless format by suing the the Buckingham π theorem [12-16], as follows:

$$N = F\left(r\sqrt{\frac{\gamma}{D}}, \sqrt{\frac{K}{\gamma}}\right), \tag{1}$$

$$\frac{\ell}{r} = G\left(r\sqrt{\frac{\gamma}{D}}, \sqrt{\frac{K}{\gamma}}\right). \tag{2}$$

These are the general relations for capillary wrinkling of the floating thin film with a liquid drop. However, both functions F and G were not able to be finalized only by the dimensional analysis, therefore other methods such as experimental and numerical ones must be used. Although we still do not know the functions F, G, these relations in eqs. (1) and (2) can still give us an important information, namely, they reveal that the pair (N, ℓ) are controlled by two combined dimensionless parameters, namely $r\sqrt{\frac{\gamma}{D}}$ and $\sqrt{\frac{K}{\gamma}}$, instead of the individual parameters.

Regarding the capillary wrinkling of elastic film, Huang et al. [5] conducted a good test. From their test data we found that the wrinkling number N mainly depends on $r\sqrt{\frac{\gamma}{D}}$; however, the wrinkling length ℓ mainly depends on the in-plane stiffness $\sqrt{\frac{K}{\gamma}}$. Therefore, if we ignore the interaction between bending and stretching, then eqs. (1) and (2) can be simplified as follows:

$$N = F\left(r\sqrt{\frac{\gamma}{D}}\right), \tag{3}$$

$$\frac{\ell}{r} = G\left(\sqrt{\frac{K}{\gamma}}\right). \tag{4}$$

Theoretically, these are the relations that we can obtain by dimensional analysis for small/moderate deformation. Generally speaking, the functions F and G might not be in power forms, which means that capillary wrinkling phenomena has no power laws and/or scaling laws. Nevertheless, these relations can still provide some useful information such as the fact that dimensionless parameter $r\sqrt{\frac{\gamma}{D}}$ is a control parameter for the wrinkling number N; while the parameter K/γ will be a control parameter for the wrinkling length ℓ.

For a small/moderate deformation, eqs. (3) and (4) could be approximately expressed in power laws of the controlling parameters as shown below:

$$N \approx \left[C_N\left(r\sqrt{\frac{\gamma}{D}}\right)^{\alpha}\right], \tag{5}$$

$$\frac{\ell}{r} \approx C_\ell\left(\sqrt{\frac{K}{\gamma}}\right)^{\beta}, \tag{6}$$

where $[Y]$ represents the integer part of real number Y, while the constants C_N, C_ℓ and exponents α, β can be determined by either numerical simulations or experiments.

For a small deformation, by curve-fitting using the experimental data of Huang et al. [5], which produces $C_\ell = 0.033$, $C_N = 3.62$ and $\alpha = 1/2$, $\beta = 1$. Hence, the wrinkling length is finally as follows:

$$\frac{\ell}{r} = 0.033\left(\frac{K}{\gamma}\right)^{1/2}, \tag{7}$$

and the wrinkling number N:

$$N = \left[3.62\left(r\sqrt{\frac{\gamma}{D}}\right)^{1/2}\right]. \tag{8}$$

Table 1 Parameters and dimensions

N	ℓ	h	E	ν	γ	r
1	L	L	$\mathrm{L^{-1}MT^{-2}}$	1	$\mathrm{MT^{-2}}$	L

It is worth pointing out that eqs. (7) and (8) are universal and are valid for all thin flat film that have a small deformation. In recognition of the pioneer contribution tot he problem, we suggest to call eqs. (7) and (8) as Huang's scaling laws of capillary wrinkling eqs. (7) and (8).

The Huang scaling laws can be used to facilitate measurements of the bending stiffness and in-plane stiffness K. If you can firstly get the N, ℓ and radius r, hence, we can calibrate the D and K as follows $D = (\frac{3.62}{N})^4\gamma r^2$ and $K = 918.27\gamma(\frac{\ell}{r})^2$ and the film thickness $h = \sqrt{\frac{12D}{K}} = 1.498N^{-2}\ell^{-1}r^2$.

With the help of eqs. (3) and (4), the above scaling laws of N and ℓ can be extended to the film made of composites materials by simply replacing D and K with a corresponding equivalent or effective bending stiffness D_{eff} and in-plane stiffness K_{eff}; hence, $N = \left[3.62\left(r\sqrt{\frac{\gamma}{D_{\mathrm{eff}}}}\right)^{1/2}\right]$ and $\frac{\ell}{r} = 0.033\left(\frac{K_{\mathrm{eff}}}{\gamma}\right)^{1/2}$. These expressions provide a pretty accurate estimation for those materials without further investigation. For example, if the thin film is made of axial symmetric orthogonal materials, $D_{\mathrm{eff}} = D_r$ and $K_{\mathrm{eff}} = K_r$, we have the pair $N = \left[3.62\left(r\sqrt{\gamma/D_r}\right)^{1/2}\right]$, and $\frac{\ell}{r} = 0.033(K_r/\gamma)^{1/2}$, with the equivalent/effective bending stiffness $D_r = E_r h^3/[12(1-\nu_r\nu_\theta)]$ and the equivalent/effective in-plane stiffness $K_r = E_r h/(1-\nu_r\nu_\theta)$, radius r, the Young modulus E_r and the Poisson ratio ν_r, and the circle Poisson ratio ν_θ.

Regarding dynamical capillary wrinkling, only Huang et al. [5] has studied the problem and proposed a scale law of capillary length as $L(t) \sim \mathrm{e}^{-t^{0.5\pm0.02}}$ by curve-fitting. In the following discussion, we will propose a different scaling law on the capillary wrinkling dynamics.

A drop on a film surface, within a complete wetting regime will slowly spread. Typically, the spreading lasts from a few hours for ordinary liquids, to several weeks for highly viscous fluids such as heavy silicone oils Figure 2.

This dynamic process can be expressed in terms of a contact angle θ_{D}, which depends on the spreading time t. When surfaces are smooth and clean, and for non-volatile liquids, Tanner [17] obtained a remarkable universal law $\theta_{\mathrm{D}} \sim t^{-3/10}$.

The measurements reveal a highly surprising fact, namely that the angle θ_{D} is completely independent of the spreading parameter $S = \gamma_{\mathrm{SO}} - \gamma_{\mathrm{SL}} - \gamma$ as long as S is positive, that is to say, as long as we are in a total wetting regime, where γ_{SO} is the surface tensions at the solid/air, γ_{SL} is the solid/liquid surface tension, and γ is the liquid/air tension, respectively. This is surprising because the force F that acts on the system of interest is essentially equal to spreading parameter $F = \gamma_{\mathrm{SO}} - \gamma_{\mathrm{SL}} - \gamma\cos\theta_{\mathrm{D}} \approx \gamma_{\mathrm{SO}} - \gamma_{\mathrm{SL}} - \gamma = S$. There are two wetting regimes for sessile drops. Partial wetting ($S < 0$): The drop does not spread, but instead forms a spherical cap at equilibrium, resting on the substrate with a contact angle θ_{D}. A liquid is said to be "mostly wetting" when $\theta_{\mathrm{D}} < \pi/2$, and "mostly non-wetting" when $\theta_{\mathrm{D}} > \pi/2$. Total wetting ($S > 0$) : if the parameter S is positive, the liquid spreads completely in order to lower its surface ($\theta_{\mathrm{D}} = 0$). The Young's law: $\gamma\cos\theta_{\mathrm{D}} = \gamma_{\mathrm{SO}} - \gamma_{\mathrm{SL}}$, and $S = \gamma(\cos\theta_{\mathrm{D}} - 1)$.

The precursor film is evidence of the great force F that acts on its boundary. The liquid is rapidly drawn towards the periphery in the form of a film whose thickness is roughly a pancake's thickness and, which is defined as $e = 2\kappa^{-1}\sin(\theta_{\mathrm{D}}/2)$ as shown in Figure 3.

But, behind the film the forces that are involved are quite different. Within the drop are forces of traction $-\gamma_{\mathrm{SL}} - \gamma\cos\theta_{\mathrm{D}}$, whereas within the film (characterized by a zero angle) there are proper forces $\gamma_{\mathrm{SL}} + \gamma$. The net force that acts on the drop is then only $F = \gamma(1-\cos\theta_{\mathrm{D}}) \approx \frac{1}{2}\gamma\theta_{\mathrm{D}}^2$, velocity $V = (V^*/6l)\theta_{\mathrm{D}}^3$, where the dimensionless coefficient $15 < l < 20$, $V^* = \dot{\gamma}/\eta$, and viscosity η. From conservation of the volume $\Omega = (\pi/4)R^3\theta_{\mathrm{D}}$ of the drop, it is easy to obtain the angle $\theta_{\mathrm{D}} \approx (\Omega^{1/3}/V^*)^{3/10}t^{-3/10}$, and the radius $R(t) \approx \Omega^{1/3}(V^*/\Omega^{1/3})^{1/10}t^{1/10}$.

Therefore, the radius of the drop is given by

$$r(t) \approx e\tan\theta_{\mathrm{D}} \approx \kappa^{-1}\theta_{\mathrm{D}}^2 = \kappa^{-1}\left(\frac{\Omega^{1/3}}{V^*}\right)^{3/5} t^{-3/5}. \tag{9}$$

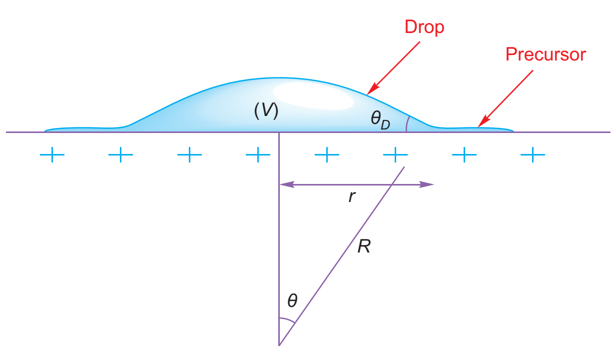

Figure 2 (Color online) Spreading of a drop on a film surface in a total wetting regime. Tanner's law: $\theta_{\mathrm{D}} \sim t^{-3/10}$. In this paper, we obtained $r \sim t^{-3/5}$.

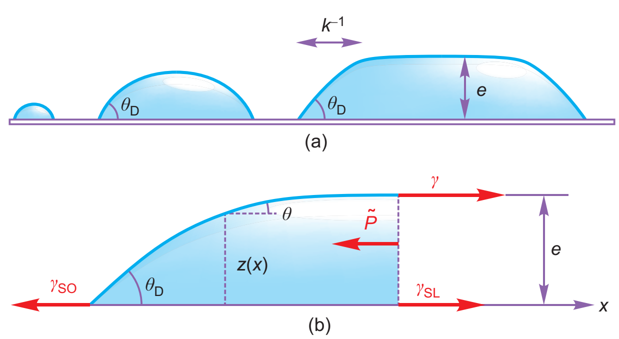

Figure 3 (Color online) (a) Liquid drops of increasing size on a sheet of film. Gravity causes the largest drops to flatten. (b) Equilibrium of the forces (per unit length of the line of contact) act on the edge of a puddle. $\tilde{P} = (1/2)\rho g e^2 = -S$ is the hydrostatic pressure. The equilibrium of forces that act on the line of contact, $\gamma(1-\cos\theta_{\mathrm{D}}) = (1/2)\rho g e^2$, gives the thickness $e = 2\kappa^{-1}\sin(\theta_{\mathrm{D}}/2)$, where the capillary length, $\kappa^{-1} = \sqrt{\gamma/(\rho g)}$.

Substituting this into eqs. (7) and (8), the wrinkling length is given by

$$\ell(t) = 0.033\left(\frac{K}{\gamma}\right)^{1/2}\kappa^{-1}\left(\frac{\eta\Omega^{1/3}}{\dot{\gamma}}\right)^{3/5} t^{-3/5}, \tag{10}$$

which can be considered a first order approximation of Huang et al. [5]. The wrinkling number is given by

$$N(t) = \left[3.62\left(\frac{\gamma}{D}\right)^{1/4}\sqrt{\kappa^{-1}}\left(\frac{\eta\Omega^{1/3}}{\dot{\gamma}}\right)^{3/10} t^{-3/10}\right]. \tag{11}$$

They are illustrated in Figure 4. It must be pointed out that both $\ell(t)$ and $N(t)$ have singularity that stems from the Turner's law at $t = 0$. In order to keep in line with Huang et al. [5], the Turner's law may have to be modified to $\theta_{\mathrm{D}} = \theta_{\mathrm{c}}\exp(-t^{1/4})$, where $\theta_{\mathrm{c}} = 1 + S/\gamma$. Hence, the radius $r(t) = \kappa^{-1}\theta_{\mathrm{c}}^2\exp(-2t^{1/4})$, the wrinkling length $\ell(t) = 0.033\left(\frac{K}{\gamma}\right)^{1/2} r(t)$ and wrinkling number $N(t) = \left[3.62(\frac{\gamma}{D})^{1/4}\sqrt{r(t)}\right]$. Those exponential singularity-free laws are needed to be confirmed in the future.

From eqs. (10) and (11), it is interesting to note that both $\ell(t)$ and $N(t)$ fade away with spreading time, and will stop at critical time $t_c = \Omega^{1/3}/V^* = \eta\Omega^{1/3}/\dot{\gamma}$. Beyond the critical time, the spreading enters the gravity regime. It is important to bear in mind that this equation applies only when r is less than the capillary length $\kappa^{-1} = \sqrt{\gamma/(\rho g)}$. When $r > \kappa^{-1}$, gravity must be taken into account [18]. The length κ^{-1} is generally of the order of a few mm. If one wants to increase the length κ^{-1}, it is necessary to work in a microgravity environment or, more simply, to replace air with a non-miscible liquid whose density is similar to that of the original liquid [18].

Gravity is negligible for sizes $r < \kappa^{-1}$. When this condition is met, it is as though the liquid is in a zero-gravity environment and capillary effects dominate. The opposite case, when $r > \kappa^{-1}$, is referred to as the "gravity" regime. In the critical situation $r = \kappa^{-1}$ and beyond, the scaling laws eq. (12) of the wrinkling length ℓ will be independent of the surface tension γ as follows:

$$\ell = 0.033\left(\frac{K}{\rho g}\right)^{1/2}, \tag{12}$$

and the wrinkling number N, given by

$$N = \left[\frac{3.62\gamma}{\sqrt{\rho g D}}\right]. \tag{13}$$

It is clear that both N and ℓ are constant within the gravity regime.

In summary, this study used dimensional analysis to define the general expression of the pair (N, ℓ), which reveals that there are no universal scaling laws for capillary wrinkling. Only in the case of small and moderate deformation, special universal scaling laws can be formulated. Regarding the bending and in-plane stiffness, it was found that the wrinkling number N is mainly controlled by the ratio of bending stiffness and surface tension, and the wrinkling length ℓ is controlled by the ratio of in-plane stiffness and surface tension. As a natural extension, we gave the pair (N, ℓ) of a thin film made of axisymmetric anisotropic materials. By using Tanner's scaling laws, we obtained dynamical scaling laws for the pair (N, ℓ), which show that the pair (N, ℓ) will fade away with the time. Finally, the pair (N, ℓ) was also obtained within the gravity regime.

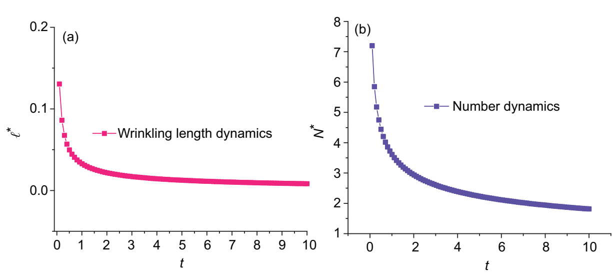

Figure 4 (Color online) (a) Capillary wrinkling length dynamics $\ell^* = \ell/\left[\left(\frac{K}{\gamma}\right)^{1/2}\kappa^{-1}\left(\frac{\Omega^{1/3}}{V^*}\right)^{3/5}\right]$; (b) capillary wrinkling number dynamics $N^* = N/\left[(\frac{\gamma}{D})^{1/4}\sqrt{\kappa^{-1}}\left(\frac{\eta\Omega^{1/3}}{\dot{\gamma}}\right)^{3/10}\right]$.

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