Scaling Law for Capillary Wrinkles in Suspended Films Induced by Droplets

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Date: 2017-09-30T00:00:00+00:00

Abstract

Research Significance: A thin film suspended on a liquid substrate, when a droplet (drop) is placed upon it, will develop wrinkles due to the droplet's surface tension. The length and number of these wrinkles are of great significance for measuring and calibrating the mechanical properties of thin film materials.

J. Huang, M. Juszkiewicz, W. H. de Jeu, E. Cerda, T. Emrick, N. Menon, and T. P. Russell, in Science 317, 650 (2007), through meticulous experiments and curve fitting, obtained empirical expressions for the length and number of wrinkles in thin films. In 2010, D. Vella, M. Adda-Bedia, and E. Cerda, Soft Matter 6, 5778 (2010), performed analytical and numerical analyses using circular plate theory, partially validating the empirical formula of Huang et al. (2007) theoretically.

Outstanding Questions: 1. Are the currently obtained empirical expressions universally applicable? 2. If not universally applicable, under what conditions can these empirical formulas be employed? 3. Existing results have all been obtained for isotropic thin films; how can the current expressions be generalized to orthotropic thin films? 4. The works of Huang (2007), Vella (2010), and others have only investigated static cases. However, in reality, the action of surface tension is a dynamic process, where wrinkle length and number evolve over time. How can we extend the static results to wrinkle dynamics? 5. Furthermore, when the droplet radius exceeds the capillary length, gravity becomes dominant and its influence must be considered (which has been neglected in previous studies). What scaling law governs the relationship between wrinkle length/number and time under such conditions?

Contributions of This Work: First, we employ dimensional analysis to derive general relationships for wrinkle length and number, revealing that they are controlled by the combined effects of two composite dimensionless parameters. Under general nonlinear deformation, no universal scaling laws exist; only under linear small deformation conditions do universal scaling laws emerge.

Under small deformation conditions, wrinkle length is primarily governed by the ratio of the thin film' s in-plane stiffness to surface tension. Physically, this means that for a given droplet-film tension, films with better in-plane tensile properties develop longer wrinkles, and vice versa. The number of wrinkles depends on the droplet radius and the ratio of the thin film' s bending stiffness to surface tension; films that are easier to bend develop more wrinkles, and vice versa. These results are fully consistent with physical observations.

For small deformation cases, we generalize the isotropic thin film scenario to the orthotropic case.

Regarding wrinkle dynamics, we successfully derive dynamic results for wrinkle length and number by incorporating Tanner' s scaling law for droplet radius evolution over time with our obtained results. No prior results on wrinkle dynamics have been reported.

Finally, we present expressions for wrinkle length and number under gravity-dominated conditions, finding that in such cases, both wrinkle length and number are constants for a given problem. No previous results on wrinkles under gravity-dominated conditions have been reported.

Full Text

Preamble

Capillary Wrinkling Scaling Laws of Floating Elastic Thin Films under a Drop

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(Dated: September 30, 2017)

This paper employs dimensional analysis to establish the general functional form of the wrinkling parameter pair (N,), identifies the dominant combined parameters governing capillary wrinkling, and determines the controlling parameters for various related problems. The dimensional analysis reveals that, in general, no universal scaling laws exist for capillary wrinkling. However, for small to moderate deformations, the wrinkling number N is found to be primarily controlled by the ratio of bending stiffness to surface tension, while the wrinkling length is governed by the ratio of in-plane stiffness to surface tension. Leveraging the linear physical relationships inherent in small deformation regimes, simpler scaling laws are proposed for the pair (N,). The universality of these scaling laws, verified through dimensional analysis, provides enhanced confidence in their applicability. As a natural extension, we derive the pair (N,) for thin films composed of axisymmetric anisotropic materials. By incorporating Tanner' s scaling laws, we obtain dynamic scaling laws for the drop radius and the pair (N,), demonstrating that both quantities fade away with time.

Finally, we derive the pair (N,) within the gravity-dominated regime.

PACS numbers: 46.32.+x, 46.70.De, 47.54.+r
Keywords: capillary wrinkling, scaling laws, floating elastic film, drop spreading, wrinkling number and length

Introduction

The deformation patterns of elastic membranes under tension are termed wrinkling. When caused by capillary surface tension, this phenomenon is specifically called capillary wrinkling (Figure 1 [Figure 1: see original paper]).

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

In recent years, wrinkling patterns have drawn particular attention [1-11], as they can serve as a useful tool for inferring 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 adhesion energy, which relates the traction forces exerted by fibroblasts during cell division to the mechanical properties of the membranes themselves (such as Young' s modulus and thickness) [5]. Considerable interest has also emerged in understanding fundamental aspects of wrinkled membranes, 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 transition from wrinkling to folding [6], [7], the analytic analysis of capillary wrinkling of circular elastic membranes [7], wrinkling of pressurized elastic shells [8], wrinkling of a charged elastic film on a viscous layer [9], and capillary deformations of bendable films [10].

Regarding capillary wrinkling of thin films, a milestone has been established by Huang et al. [5], who experimentally derived the length and number of capillary wrinkles, while Vella et al. [7] theoretically proved the capillary wrinkling length expression obtained by Huang et al. [5] for small deformation of a circular film. Huang et al. [5] presented a novel experiment combining both fundamental and applied aspects of the interaction between surface tension and elasticity. In this experiment, a small liquid drop was placed onto an elastic membrane floating 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, the contact line caused radial wrinkles due to opposing tension. Experimentally, they found that the wrinkling length = 0.031R(Eh/)^(1/2), where r is the radius of the drop and is the surface tension coefficient of the liquid-gas interface. This purely empirical relationship was confirmed by theoretical justification from Vella et al. [7], who employed the Föppl-von Kármán thin plate theory.

Although Huang et al. [5] obtained formulas for the pair (N,), where N is the wrinkling number, the question remains: are these relationships universally applicable? If not in the general case, then under what circumstances can they be approximated as universal scaling laws? How can one extend these results to other materials, such as axisymmetric anisotropic materials? The obtained pair (N,) represented static results, so how can scaling laws be established for dynamic problems? If the characteristic scale of the drop exceeds the capillary scale ^(-1), gravity begins to dominate the wrinkling process, so what are the scaling laws beyond the capillary scale ^(-1)?

This paper first introduces the research topic before presenting the general expression of the pair (N,) through dimensional analysis and determining the controlling parameters of the problem. Based on understanding of experimental data, we propose linear approximated scaling laws for the pair, then extend these to films made of composite and laminated materials. For spreading drops, dynamic scaling laws are proposed based on Tanner' s law. Finally, the paper concludes with a formulation of wrinkling scaling laws in the gravity regime.

Capillary Wrinkling Scaling Laws for Small/Moderate Deformation

Generally speaking, both the wrinkling number and length are functions of the Young' s modulus E, film thickness h, Poisson' s ratio , surface tension , and drop radius r, namely, N = F(E, h, , , r) and = G(E, h, , , r), where F and G are unknown functions. The dimensions of these parameters are listed in Table I below.

TABLE I: Parameters and Dimensions

Since the film experiences both bending and stretching states, by dimensional analysis [12–14], the above formulas can be further expressed as:

N = F(D, K, , r) and = G(D, K, , r),

where the bending stiffness D = Eh^3/[12(1- ^{2})] and the in-plane stiffness K = Eh/(1- ^{2}). These can also be expressed in dimensionless form as follows:

N = F(r /D) and = G(rK/).

These are the general scaling relations for capillary wrinkling of floating thin films. However, dimensional analysis alone cannot determine the specific forms of functions F and G, requiring other methods. These relations reveal that the pair (N,) is controlled by two dimensionless parameters: r /D and rK/ .

Regarding capillary wrinkling of elastic films or membranes, Huang et al. [5] conducted excellent experiments. From their test data, we found that the wrinkling number N depends mainly on the bending stiffness D, while the wrinkling length depends mainly on the in-plane stiffness K. Therefore, Eqs. (1) and (2) can be simplified as:

N = F(r /D) and = G(rK/).

Theoretically, these are the relations obtained from dimensional analysis. Generally speaking, the functions F and G are not simple power laws, which means that capillary wrinkling phenomena lack universal power laws and/or scaling laws. Nevertheless, these relations still provide useful information, such as the fact that the dimensionless parameter /D serves as a control variable for the wrinkling number N, while K/ acts as a control variable for the wrinkling length .

For small deformations, physical laws must be linear; therefore, Eqs. (3) and (4) should be linear functions of the controlling parameters as shown below:

N   CN[(/D)^ r] and   C [(K/)^ r],

where [Y] represents the integer part of real number Y, while the constants CN, C and exponents ,   can be determined by either numerical simulations or experiments.

For small deformation, based on data fitting, Huang et al. [5] found CN = 3.62, C = 0.033 and   = 1/2,   = 1/2. Hence, the scaling laws are:

N = [3.62(/D)^(1/4) r] and   = 0.033r(K/)^(1/2).

It is worth pointing out that Eqs. (7) and (8) are universal and valid for all flat thin films undergoing small deformation. The scaling laws in Eqs. (7) and (8) can facilitate measurements of the bending stiffness D and in-plane stiffness K. If one first measures N,   and radius r, then D and K can be calibrated as:

D = 0.033 (r/N)^4 and K = 918.27 (/r)^2,

and the film thickness h can be determined as:

h = 2.88(1- ^{2}) ^(1/2) N^(-1/2) ^(-1) r^(5/4) E^(-1/2).

These scaling laws have been theoretically confirmed by Vella et al. [7], who used von Kármán’ s circular plate theory and derived similar scaling laws for capillary wrinkling.

With the help of Eqs. (3) and (4), the above scaling laws for N and   can be extended to films comprising composite and laminated materials by simply replacing D and K with corresponding equivalent or effective bending stiffness D_eff and in-plane stiffness K_eff:

N = [3.62(/D_eff)^(1/4) r] and   = 0.033r(K_eff/)^(1/2).

These expressions provide a reasonably accurate estimation for those materials without requiring further investigation. This represents one advantage of having universal scaling laws verified by dimensional analysis, which definitely gives us ultimate confidence in such extensions.

For example, if the thin film is made of axisymmetric orthogonal materials, with D_eff = D_r and K_eff = K_r, we have the pair:

N = [3.62(/D_r)^(1/4) r] and   = 0.033r(K_r/)^(1/2), with the equivalent/effective bending stiffness D_r = E_r h^3/[12(1- _r _)] and the equivalent/effective in-plane stiffness K_r = E_r h/(1- _r _), where E_r is the radial Young' s modulus, _r is the radial Poisson' s ratio, and _ is the circumferential Poisson' s ratio.

Capillary Wrinkling Dynamics of a Totally Wettable Film

While static wrinkling problems have been investigated, no dynamic wrinkling has been studied yet. 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 [Figure 2: see original paper]).

FIG. 2: Spreading of a drop on a film surface in a total wetting regime. Tanner' s law: _D t^(-3/10). In this paper, we obtained r t^(-3/5).

This dynamic process can be expressed in terms of a contact angle _D, which depends on the spreading time t. When surfaces are smooth and clean, and for non-volatile liquids, Tanner [16] obtained a remarkable universal law: _D t^(-3/10). The measurements reveal a highly surprising fact: the angle _D is completely independent of the spreading parameter S = _SO - _SL - as long as S is positive, that is, as long as we are in a total wetting regime, where the three coefficients represent the surface tensions at the solid/air, solid/liquid, and liquid/air interfaces, respectively. This is surprising because the force F that acts on the system of interest is essentially equal to the spreading parameter: F = _SO - _SL - cos _D _SO - _SL - = 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 _D. A liquid is said to be "mostly wetting" when _D < /2, and "mostly non-wetting" when _D > /2. Total wetting ($S > 0$): If the parameter S is positive, the liquid spreads completely in order to lower its surface energy (_D = 0). The Young-Dupré law states: cos _D = _SO - _SL, and S = (cos _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, defined as e = 2 ^(-1) sin(_D/2), as shown in Figure 3 [Figure 3: see original paper] below.

FIG. 3: (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) acting on the edge of a puddle. P = (1/2) ge^2 = -S is the hydrostatic pressure. The equilibrium of forces acting on the line of contact, (1 - cos _D) = (1/2) ge^2, gives the thickness e = 2 ^(-1) sin(_D/2), where the capillary length ^(-1) = /(g).

However, behind the film the forces involved are quite different. Within the drop are forces of traction - _SL - cos _D, whereas within the film (characterized by a zero angle) there are proper forces _SL + . The net force acting on the drop is then only F = (1 - cos _D) (1/2) _D². The spreading velocity V = (V/6l) _D³, where 15 < l < 20, V = ˙/ , and is viscosity. From conservation of the volume Ω = (/4)R³ _D of the drop, it is easy to obtain the angle _D (Ω^(1/3)/V*)^(3/10) t^(-3/10), and the radius R(t) Ω(1/3)(V/Ω(1/3))(1/10) t^(1/10). Therefore, the radius of the drop is given by r e tan _D ^(-1) _D² = ^(-1)(V/Ω(1/3))^(3/5) t^(-3/5). This scaling law for the radius of a drop has not been recorded in literature.

When one substitutes this into Eqs. (5,6), one obtains the wrinkling length:

= 0.033(K/)^(1/2) (-1)(Ω(1/3)/V*)^(3/5) t^(-3/5),

and the wrinkling number:

N = [3.62(/D)^(1/4) (-1)(Ω(1/3)/V*)^(3/10) t^(-3/10)].

The dynamics of both length and number are illustrated in Figure 4 [Figure 4: see original paper].

Capillary Wrinkling within the Gravity Regime

From Eqs. (14,15), it is interesting to note that both and N fade away with spreading time, and will stop at a critical time t_c = Ω^(1/3)/V* = Ω^(1/3)/˙. Beyond this critical time, the spreading enters the gravity regime. It is important to bear in mind that these equations apply only when r is less than the capillary length ^(-1). When r > ^(-1), gravity must be taken into account [15].

There exists a particular length, denoted ^(-1) = /(g), beyond which gravity becomes important, referred to as the capillary length. The length ^(-1) is generally of the order of a few millimeters. If one wants to increase the length ^(-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 [15]. Gravity is negligible for sizes r < ^(-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 > ^(-1), is referred to as the "gravity" regime.

In the critical situation r = ^(-1) and beyond, the scaling law Eq. (12) for the wrinkling length becomes independent of surface tension as follows:

= 0.033(K/ g)^(1/2),

and the wrinkling number N becomes:

N = [3.62(g/D)^(1/4) ^(-1)].

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

FIG. 4: (a) Capillary wrinkling length dynamics * = /[0.033(K/)$^{(1/2)}$ (-1)(Ω^(1/3)/V*)^(3/5)]; (b) Capillary wrinkling number dynamics N* = N/[3.62(/D)$^{(1/4)}$ (-1)(Ω^(1/3)/V*)^(3/10)].

Conclusion

This paper has attempted to answer all the aforementioned questions. In general, there are no universal scaling laws for capillary wrinkling. Only in the case of small and moderate deformation can special universal scaling laws be formulated. Regarding bending and in-plane stiffness, it was found that the wrinkling number N is mainly controlled by the ratio of bending stiffness to surface tension, and the wrinkling length is controlled by the ratio of in-plane stiffness to surface tension. By using Tanner' s scaling laws, we obtained dynamic scaling laws for a drop radius and the pair (N,), which shows that the pair (N,) will fade away with time. Finally, the pair (N,) within the gravity regime was also revealed.

In summary, the highlights of this paper are listed in Tables II and III:

TABLE II: Static capillary wrinkling pair (N,)

Condition	N	
Surface tension dominate (r < ^(-1))	N = [3.62(/D)^(1/4) r]	= 0.033r(K/)^(1/2)
Gravity dominate (r > ^(-1))	N = [3.62(g/D)^(1/4) ^(-1)]	= 0.033(K/ g)^(1/2)

TABLE III: Dynamic capillary wrinkling pair (N,)

Parameter	Expression
N	N = [3.62(/D)^(1/4) $^{(-1)(\Omega}$(1/3)/V*)^(3/10) t^(-3/10)]
	= 0.033(K/)^(1/2) $^{(-1)(\Omega}$(1/3)/V*)^(3/5) t^(-3/5)

Acknowledgements

The author wishes to thank Michael Sun from SACS Junior, Cape Town, for taking the picture in Figure 1.

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Note: Figure translations are in progress. See original paper for figures.