Elastic sheet on a liquid drop reveals wrinkling and crumpling as distinct symmetry-breaking instabilities

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Smooth wrinkles and sharply crumpled regions are familiar motifs in biological or synthetic sheets, such as rapidly growing plant leaves and crushed foils. Previous studies have addressed both morphological types, but the generic route whereby a featureless sheet develops a complex shape remains elusive. Here we show that this route proceeds through an unusual sequence of distinct symmetry-breaking instabilities. The object of our study is an ultrathin circular sheet stretched over a liquid drop. As the curvature is gradually increased, the surface tension stretching the sheet over the drop causes compression along circles of latitude. The compression is relieved first by a transition into a wrinkle pattern, and then into a crumpled state via a continuous transition. Our data provide conclusive evidence that wrinkle patterns in highly bendable sheets are not described by classical buckling methods, but rather by a theory which assumes that wrinkles completely relax the stress field. With this understanding we recognize the observed sequence of transitions as distinct symmetry breakings of the shape and the stress field. The axial symmetry of the shape is broken upon wrinkling but the underlying stress field preserves this symmetry. Thus, the wrinkle-to-crumple transition marks symmetry-breaking of the stress in highly bendable sheets. By contrast, other instabilities of sheets, such as blistering and cracking, break the homogeneity of shape and stress simultaneously. The onset of crumpling occurs when the wrinkle pattern grows to half the sheet’s radius, suggesting a geometric, material-independent origin for this transition.

The richness and beauty of the patterns exhibited by a thin sheet may be viewed from the perspective of nonequilibrium pattern formation theory (1), which posits that morphological diversity emanates from successive instabilities of a highly symmetric state. A simple example is the buckling of a sheet of paper compressed along two edges. The mechanics of this familiar Euler instability reflects the disparity between weak bending forces and strong compression forces, which becomes greater as the sheet gets thinner (2, 3). Beyond the mechanical point of view, this instability can be recognized as a spontaneous breaking of the translation symmetry of the sheet in the confinement direction. The conceptual link between the Euler instability, symmetry breaking, and pattern formation is more obvious when the sheet is placed under tension or is compressed on a soft substrate (4, 5). These conditions produce a periodic wrinkle pattern with a finite wavelength. The wrinkle pattern may be subject to secondary instabilities that break the translational symmetry further by introducing modes of larger wavelength (6, 7), or by superimposing on the wrinkles a localized fold shape (8–10). This sequence of pattern development parallels that of prototypical driven systems such as Rayleigh–Benard convection and Taylor–Couette flow (1).

When sheets are subjected to a curved, two-dimensional (2D) confinement (11, 12), higher levels of shape complexity are obtained. Whereas the stress field is essentially uniform in the one-dimensional (1D) examples above, it is spatially varying and strongly coupled to the shape in 2D geometries. In this article, we study the sequence of patterns that develop when an ultrathin, circular sheet is placed on a fluid drop. The surface tension stretching the sheet over the curved drop leads to destabilizing compressive forces in the azimuthal direction. When the curvature of the drop is steadily increased, the axial symmetry of the initial geometry is first broken by a transition to a wrinkled state. In addition to the primary wrinkling instability, we discover yet another instability at higher curvature, in which the wrinkle pattern gives way to a state of lower azimuthal symmetry with localized crumpled features. Two unique features associated with this sequence set it apart from the traditional progression of pattern formation theory: First, the wrinkled state must be viewed as a perturbation about the compression-free limit of a sheet, which can bend at no energetic cost (13–15), rather than through a perturbation about the unstable axisymmetric state (16) that is the basis of traditional near-threshold (NT) postbuckling methods. Although axial symmetry might be expected to be completely lost in the presence of a well-developed wrinkle pattern, remarkably, the stress field retains this symmetry. Our measurements of the wrinkle length provide conclusive evidence for this far-from-threshold (FFT) theory. Second, we show that crumpling emerges from wrinkling through a continuous transition whereby the stress field breaks symmetry. These observations suggest that the wrinkle-to-crumple transition constitutes an unexplored class of morphological phase transition that reflects a primary symmetry breaking of the force field rather than merely lowering the azimuthal symmetry of the shape further. We discover that the onset of the wrinkle-to-crumple transition does not depend explicitly on the elastic moduli of the sheet or the exerted tension. Instead, it occurs when an emergent geometric observable—the wrinkle length—reaches a threshold value that is close to half the radius of the sheet.

Physical System

Our setup is sketched in Fig. 1. A circular polystyrene (PS) sheet is delivered to the free surface of water at the top of a tube of radius 2.5 mm. We use sheets of thickness $t$ from 49 to 137 nm, and radius $W$ of 0.79 or 1.5 mm. The shape of the exposed air-water interface shows no measurable deviations from a spherical cap with radius $R = 2γ/P$, where $γ$ is the liquid-vapor surface tension that stretches the sheet radially at its perimeter and $P$ is the Laplace pressure that acts normal to its surface. The pressure

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*Although the words “wrinkle” and “crumple” are interchangeable in colloquial usage, we use them in their current technical sense to mean a sinusoidal, smooth deformation (3) and a stress-focusing deformation (5), respectively.

†In our experiment, the gravitational contribution to the pressure over the height of the drop is much smaller than the Laplace pressure.

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modulus (the sheet is much smaller than the radius of curvature or experience increasing confinement along latitudes of the sphere. If the sheet is sufficiently large or the tension sufficiently weak, the hoop stress becomes compressive close to its outer edge, leading to a wrinkling instability of the axisymmetric state. These qualitative expectations are borne out by an analysis of the axisymmetric Föppl-von Kármán (FvK) equations (18), which yields the explicit form of the confinement parameter (Materials and Methods and SI Appendix):

\[ \text{confinement: } \alpha \equiv \frac{YW^2}{2\gamma R^2}. \]  

The FvK equations express force balance on a sheet in the transverse (radial and azimuthal) and normal directions (2), and their axisymmetric form is, as follows:

radial: \[ d(r\sigma_r)/dr - \sigma_{0\theta} = 0 \]  

normal: \[ B\Delta^2\zeta - \sigma_r d^2\zeta/dr^2 - \sigma_{0\theta} r^{-1} d\zeta/dr = P. \]

where \( \sigma_r(r), \sigma_{0\theta}(r) \) are the radial and hoop stress components, respectively, \( \zeta(r) \) is the normal displacement of the sheet, and \( \Delta_r \equiv r^{-1} \frac{d}{dr}(r \frac{d}{dr}) \). The confinement parameter \( \alpha \) emerges from balancing the tensional terms with the pressure in Eq. 3, and ignoring the bending force. For \( \alpha \) smaller than a critical value \( \alpha^{\text{cr}} \), a very thin sheet is everywhere under tension in both radial and azimuthal directions, but for \( \alpha > \alpha^{\text{cr}} \approx 5.16 \) the confinement leads to azimuthal compression in an annulus \( L(a) < r < W \) near its outer rim.

Bendability. The bending force (~B) in the normal force balance, Eq. 3, is negligible if the sheet is sufficiently thin or the surface tension is sufficiently large. Comparing the bending and tensile forces in Eq. 3, which scale, respectively, as \( B/W^2 \) and \( \gamma/W^2 \), we find the bendability parameter:

\[ \text{bendability: } \epsilon^{-1} \equiv \gamma W^2/B. \]

Our experiments, which use ultrathin sheets, explore the high-bendability regime, \( \epsilon^{-1} > 2 \cdot 10^5 \), where the shape is primarily governed by the balance of stretching and the resistance of the drop to deviate from its favored sphericity. It is the bendability parameter that differentiates our system from the “capillary origami” experiment (12), where thicker sheets were used to wrap fluid droplets. For those low-bendability films, it was demonstrated that the drop’s pressure was balanced by bending forces.

The confinement and bendability parameters span a phase diagram that is shown later in the article in schematic form. Large enough confinement \( \alpha > \alpha_{\text{cr}}(\epsilon) \) leads to an instability of the axisymmetric state [where \( \alpha_{\text{cr}}(\epsilon) \rightarrow \alpha_{\text{cr}}^\text{cr} \) as \( \epsilon \rightarrow 0 \)]. However, the bendability parameter radically affects the wrinkling pattern (17) that emerges from this instability. In the low-bendability regime, the sheet accommodates compression even in its buckled shape, and a proper description is obtained by a traditional buckling analysis (16) that considers the wrinkled state as an NT perturbation of the axisymmetric state with compression \( \sigma_{0\theta}(r) < 0 \) at an annulus \( L(a) < r < W \). By contrast, in the high-bendability regime only tiny levels of compression (which vanishes as \( \epsilon \rightarrow 0 \)) can be supported in the sheet. Here the wrinkled state must be described by an FFT perturbation theory around the singular membrane limit of a sheet with zero bending modulus (but finite stretching modulus), where wrinkles do not cost any bending energy and compression can be completely relaxed (13–15).

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**Confinement.** When a thin, flat disc is projected onto an undeformable sphere, material elements at larger radii on the disc experience increasing confinement along latitudes of the sphere. If the sheet is much smaller than the radius of curvature \( W \ll R \), or if the radial tension exerted along the edge \( (\gamma) \) is large, the sheet will be under tension everywhere in both radial and azimuthal (hoop) directions. In this case the sheet preserves its axisymmetry and stretches smoothly into a spherical cap. However, if the sheet is sufficiently large or the tension sufficiently weak, the hoop stress becomes compressive close to its outer edge, leading to a wrinkling instability of the axisymmetric state. These qualitative expectations are borne out by an analysis of the axisymmetric Föppl-von Kármán (FvK) equations (18), which yields the explicit form of the confinement parameter (Materials and Methods and SI Appendix):

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Notably, the stress field in the FFT limit does preserve axial symmetry and is obtained as a solution of the axisymmetric FvK Eqs. 2 and 3. The calculational framework of the FFT approach is outlined in Materials and Methods section (see also SI Appendix), where we explain that it leads to markedly different predictions for the size of the wrinkled annulus, and the number of wrinkles, than does an NT buckling analysis.

In contrast to the purely geometric von Kármán number, $W^+Y/B \sim (Wt)^{1/2}$, which is the familiar measure of thinness (2), the bendability parameter (4) incorporates mechanics through $\gamma$. The demarcation of the low- and high-bendability regimes resembles a basic idea in fluid mechanics, where the Reynolds number defines the viscous (low Re) and inviscid (high Re) limits of the Navier–Stokes equations. Similarly, the bendability parameter defines two distinct limits of the FvK equations of thin sheets. Although the low-bendability regime has been the subject of classical buckling analysis, the high-bendability regime, where the FFT theory is required, has remained largely unexplored.

Results and Discussion

In Fig. 2A we show a set of images (I–V) that from left to right indicate the patterns obtained upon increasing the pressure of the droplet. The sheet first smoothly stretches (I), and then starts wrinkling at the edge (II). The wrinkles grow toward the center of the sheet and decrease modestly in number (III). Further in the progression, some of the wrinkles develop sharp cusp-like patterns (IV). Finally, most of the original wrinkles recede, leaving a few sharply defined crumpled features (V) that are approximately uniformly distributed around the sheet. (Higher resolution images and the corresponding side views are available in SI Appendix.)

This progression is quantitatively characterized in terms of $L = L/W$, the fractional extent of the unwrinkled zone in the interior of the sheet, and $2\pi m$, the average angle between wrinkles. Both are plotted as functions of the confinement $\alpha$ for a sheet with thickness $t = 77$ nm. Fig. 2 shows that wrinkles emerge at a sharply defined value, $\alpha_{wr}$. The wrinkles grow inward for $\alpha > \alpha_{wr}$ in very good agreement with the power law $L \sim \alpha^{-1/3}$ predicted by the FFT analysis, Eq. 5, over about 1.5 decades of confinement values $\alpha$. Over the same range, the number of wrinkles increases slightly as shown in Fig. 2B. From similar plots of $L$ for other sheets, we obtain values of $\alpha_{wr}$ that vary between 3 and 11 from sample to sample. This range includes the predicted value $\alpha_{wr}^0 \approx 5.16$, and there is no systematic dependence of the measured $\alpha_{wr}$ on $t$ and $W$.

For $\alpha \gtrsim \alpha_{cr} \approx 30\alpha_{wr}$, a deviation is seen from the power law scaling of the wrinkle extent, at approximately the same values of pressure at which sharp, crumple-like features begin to appear at the tips of some wrinkles. It is difficult to pinpoint the exact value $\alpha_{cr}$ at which these features appear, but beyond this value, multiple length scales appear in the wrinkle pattern. We show in Fig. 2C the length of the leading crumples and the length of the receding wrinkles.

In Fig. 3A, we once again show the extent of the unwrinkled zone, but for a number of sheets of varying thickness and size, now plotted against the scaled confinement parameter $\alpha/\alpha_{wr}$, where $\alpha_{wr}$ is the measured value at the wrinkling threshold. The data for all thicknesses collapse well for $\alpha \leq \alpha_{cr}$, indicating that the length of the wrinkles is independent of the bendability $\epsilon^{-1}$ as anticipated in the FFT theory. There is no regime of $\alpha$ in which the NT behavior (indicated in Fig. 3C by a dotted line) is seen, an absence consistent with the high-bendability values of the sheets used here ($\epsilon^{-1}$ varies from $2 \times 10^5$ to $5 \times 10^6$). As previously discussed, the FFT theory expands about a limit in which the wrinkles completely relax the compressive stress. Thus, the success of the FFT theory implies that the stress field remains largely axisymmetric throughout the wrinkled regime, which is not at all obvious from inspection of the images in Fig. 3.

In Fig. 3B we show a similar plot of the wrinkle separation angle $2\pi m$ scaled by the anticipated FFT dependence of $\epsilon^{-1/3}$, Eq. 5. The bendability $\epsilon^{-1}$ thus affects the scaling of the wave-
lengths of the wrinkling pattern (5, 19), but not the stretching energies that control the wrinkle extent (17). A detailed numerical prediction for the wrinkle angle is not yet available (17) (SI Appendix), but the data collapse suggests that the wrinkle number \( m \) depends only weakly on \( \alpha \).

As Fig. 3A shows, the deviation from the power law scaling becomes evident for \( \alpha > \alpha_{cr} \approx 30 \alpha_{cr} \). The spread in the angle and length data beyond this value reflects the complexity of the crumpled state (Fig. 2A, V) that ultimately replaces the wrinkle pattern. A representative localized shape (Fig. 3C) shows that one of the motifs of the crumpled state has the shape of a developable cone (d-cone), which is a fundamental stress-focused element seen through interference fringes (3). The emergence of these localized structures strongly modifies the overall shape of the drop and sheet: The broken azimuthal symmetry of the wrinkle pattern gives way to an even lower symmetry polygonal shape.

We propose that crumpling emerges from an instability of wrinkling upon large confinement. Thus, a proper description of the crumpling transition requires a nonlinear, FFT description of the wrinkled state. The continuous deviation of \( L \) from the predicted FFT wrinkle extent (Fig. 3A) and the gradual emergence of focused structures from the tips of wrinkles suggests that the crumpling transition is of second order (1). Furthermore, the apparently continuous nature of the crumpling transition is intimately related to the symmetry-breaking nature of this transition. As our theoretical discussion emphasized, the FFT wrinkling solution of FvK equations satisfies two distinct properties: a vanishing compressive stress and an axisymmetric stress field. However, stress focusing is obviously incompatible with axial symmetry. We thus conclude that wrinkling is a primary instability that breaks the axial symmetry of the pattern but not of the stresses, whereas crumpling emerges from a nonstandard second instability that breaks the axial symmetry of the stress field rather than just producing a lower level of azimuthal symmetry in the shape.

In the crumpled state, our measurements show a spread in the scaled extent and angle that is not collapsed by the scaled confinement \( \alpha/\alpha_{cr} \). Furthermore, we note that the onset of the crumpling transition occurs at values of \( \alpha_{cr} \) that vary by a factor of 2 or 3 (black circles in Fig. 3A) for different samples. However, the crumpling transition is more sharply defined in terms of the ordinate of Fig. 3A and is seen to occur when the radius of the unwrinkled zone shrinks to about a half the sheet radius \( L \approx 0.5 \pm 0.02 \). We recall that the threshold value for the wrinkling transition, \( \alpha_{cr} \), is controlled by both mechanics (\( Y, \gamma \)) and geometric length scales (\( W, R \)). The observation that the crumpling transition occurs when an emergent length scale \( L \) becomes a finite fraction of the sheet’s radius \( W \) leads us to infer that this transition is geometric in origin. The wrinkles can no longer minimize elastic energy efficiently, and a broken-symmetry, localized stress field becomes the favored state.

**Conclusions**

Confinement of a sheet on a deformable sphere leads to a richness of form that is useful to view through the lens of pattern development theory. Our observations and analysis suggest the schematic phase diagram plotted in Fig. 4. The sheet is unwrinkled for low confinement values, \( \alpha < \alpha_{cr} \). As is evidenced by our measurements of the extent and number of wrinkles in ultrathin, highly bendable sheets, the FFT scenario becomes operative even at confinement values that are only slightly above threshold. This result emphasizes the importance of the correct application of FFT behavior to the many modern applications that use sheets of nanoscopic dimensions as mechanical elements. A notable feature of the finite-amplitude FFT wrinkle pattern, which distinguishes it from other deformations such as blisters

\[ \alpha \approx \alpha_{cr} \]
geometry of a thin annular sheet under an axisymmetric radial tension gradient (17). However, the imposed curvature leads to a qualitatively unique feature in the phase diagram: a crumpled phase at large values of confinement. An important advantage of our system is the ability to test the scaling of wrinkle length for confinements much larger than the threshold value, whereas experiments in the planar, Lamé geometry are limited to no more than two to three times the threshold value (17, 19, 20). Despite the central role of bendability and confinement, we expect that other morphologically relevant parameters will be required to characterize instabilities that are induced by substrate stiffness and boundary effects. Examples include the folding in floating sheets (8, 21), period-doubling (7), and n-tupling (6) in sheets attached to elastomers, a subcritical creasing instability in neo-Hookean solids (22, 25), and multiscale wrinkling cascades (24, 25).

As far as we are aware, our study provides a unique experimental scenario that exhibits a controlled transition between wrinkled and crumpled shapes. This second instability is associated with a primary symmetry breaking of the stress field. The data indicate a continuous transition determined by the inherent frustration of the geometry rather than by mechanical forces. Analogous studies of 2D crystals on droplet surfaces (26, 27) show that point and line lattice defects may also resolve geometric frustration. The relationship between those defects and the localized structures emerging from a continuum remain to be explored. Although our motivation in choosing a spherical substrate was to frustrate the sheet at all points in space, it is not clear that the spherical geometry was a necessary ingredient to achieve crumpling. One-dimensional patterns do not appear to crumple, and neither do all 2D geometries (17, 19, 20). It remains a puzzle as to what the generic conditions are under which a wrinkled state gives way to crumpling.

Materials and Methods

Apparatus. The tube depicted in Fig. 1 was set in a hole through an acrylic base. The hole was sealed on the bottom by a glass slide to keep the setup watertight and transparent to allow illumination from beneath. The surface symmetry was a necessary ingredient to achieve crumpling. For example, crumpling patterns do not appear in curved sheets (8, 21), period-doubling (7), and n-tupling (6) in sheets attached to elastomers (22, 25), and multi-scale wrinkling cascades (24, 25). Despite the central role of bendability and confinement, we expect that other morphologically relevant parameters will be required to characterize instabilities that are induced by substrate stiffness and boundary effects. Examples include the folding in floating sheets (8, 21), period-doubling (7), and n-tupling (6) in sheets attached to elastomers, a subcritical creasing instability in neo-Hookean solids (22, 25), and multiscale wrinkling cascades (24, 25). As far as we are aware, our study provides a unique experimental scenario that exhibits a controlled transition between wrinkled and crumpled shapes. This second instability is associated with a primary symmetry breaking of the stress field. The data indicate a continuous transition determined by the inherent frustration of the geometry rather than by mechanical forces. Analogous studies of 2D crystals on droplet surfaces (26, 27) show that point and line lattice defects may also resolve geometric frustration. The relationship between those defects and the localized structures emerging from a continuum remain to be explored. Although our motivation in choosing a spherical substrate was to frustrate the sheet at all points in space, it is not clear that the spherical geometry was a necessary ingredient to achieve crumpling. One-dimensional patterns do not appear to crumple, and neither do all 2D geometries (17, 19, 20). It remains a puzzle as to what the generic conditions are under which a wrinkled state gives way to crumpling.

Film Preparation. Films were prepared by spin-coating from a dilute solution of PS in toluene (atactic, number-average molecular weight Mn = 91 K, weight-average molecular weight Mw = 95 K) (Polymer Source Inc.) on glass substrates. Different film thicknesses were obtained either by varying spin speed (between 800 and 2000 rpm) or the concentration of polystrene in solution (between 1 to 2% by mass). Circular cuts of radius W = 0.79 or 1.5 mm were made in the film. The circular film was released on to an air-water interface and then transferred onto the experimental set up (SI Appendix).

The thickness was measured with a precision of 0.2 nm using a filmic white-light interferometer, and was uniform to ±1 nm over the area of the film.

Images. Two types of raw data were collected: top-view photographs, in which features of the film could be measured, and side-view photographs of the menisci of the three fixed tubes, from which the pressure under the film could be calculated. The top-view photographs were taken by a Nikon D5000 dSLR camera equipped with a bellows unit for macrophotography. The camera was attached to a microscope base which provided light from below. Contrast in the images convey the local slope of the film-air interface.

The intensity as a function of angle was analyzed at all radial positions to determine wrinkle length and wrinkle angle (SI Appendix).

FFT Scheme. The calculation proceeds in two steps (17) (SI Appendix):

i. We find the asymptotic stress field of the system in the membrane limit by solving Eqs. 2 and 3 under the constraint that the stress field is compression-free (i.e., σzz ≥ 0) (13). The stress therefore remains axisymmetric in the FFT wrinkled state even though the geometry of the shape is broken. The compression-free stress field determines the FFT extent of wrinkles Lfft. In Fig. 5 we present a numerical solution of the FvK Eqs. 2 and 3 in the NT and FFT limits (Fig. 5 B and C, respectively) for a representative value of α > D1. The stress field is axisymmetric in the FFT limit. The mean field therefore remains axisymmetric in the FFT wrinkled state even though the geometry of the shape is broken.

ii. We determine the number of wrinkles, mfft, from a perturbation theory around the membrane limit. The distinct nature of the reference stress fields results in markedly different predictions for the number and width of wrinkles in the NT and FFT regimes, as we explain in detail in SI Appendix. We predict that the normalized extent of the unwrinkled zone L = L/W, and the number of FFT wrinkles are, as follows:

\[ L_{FFT}(\alpha) = \sqrt{\alpha n_{w}/\alpha}; \quad m_{FFT} \approx k(\alpha)\alpha^{-1/4}. \]  

The predicted extent of the FFT wrinkles is plotted in Fig. 3 A (solid black). The scaling law for the wrinkle number is similar to the one discovered by Cerda and Mahadevan (5) for stretched rectangular sheets.

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