Supplementary materials 1 - Cut-out disclinations

FIG. 1:
FIG. 2:
FIG. 6:
The shape of the glycerol-oil interfaces in our experiments, is determined by the shape of the boundary and the difference in Laplace pressure between the two sides of the interface. The colloidal particles do not wet the interface and the elastic energy of the crystal lattice is not sufficient to compete with the surface tension and deform the droplet. Typically our boundaries are azimuthally symmetric about the $z$ axis and, since the height of the droplets is much smaller than the capillary height, the difference in pressure is constant.

The combination of azimuthal symmetry and constant Laplace pressure (which is proportional to the mean curvature of the surface) restricts the shape to either sections of spheres, as in the case of our domes, or a family of surfaces known as unduloids, nodoids and cantenoids (See Fig.1). These are surfaces of revolution that have constant mean curvature\cite{1, 2}.

We have found that in the parameter region considered in our experiments, the following function approximates the shape of the surface well, and has the advantage of being more amenable to analytical calculations, even though it does not have constant mean curvature:

\[ r = (R_\phi - R_z) + R_z \cosh \left( \frac{z}{R_z} \right) \]

where $R_\phi$ and $R_z$ are the principal radii of curvature at the neck.

Figure 2 shows two examples of unduloids, superposed with our trial function. The corresponding expression for the Gaussian curvature is:

\[ G(z) = -\frac{\text{sech}^3 \left( \frac{z}{R_z} \right)}{R_z \left( (R_\phi - R_z) + R_z \cosh \left( \frac{z}{R_z} \right) \right)} \]
FIG. 1: Capillary bridges take the form of Nodoids (blue), Unduloids (red), Spheres (green) or Catenoids (black), depending on the wetting angle, volume and Laplace pressure of the droplet.
FIG. 2: Comparison between our simplified expression for the surface and nodoids in two cases.


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Supplementary materials 3: Energetics of dislocations and pleats on curved capillary bridges

In this supplementary material we investigate, within continuum elastic theory [1–4], the energetics of defected crystals on capillary bridges and calculate the integrated Gaussian curvature above which dislocations and pleats first appear. The stress tensor \( \sigma_{ij} \) and the interaction of topological defects with curvature can be determined from the Airy stress function \( \chi(x) \) whose sources are the Gaussian curvature \( G(x) \) and the disclinations [5]:

\[
\frac{1}{Y} \nabla^4 \chi(x) = \sum_{\alpha} q_\alpha \delta(x - x_\alpha) - G(x) ,
\]

where \( q_\alpha \) and \( x_\alpha \) are the disclination charges and positions respectively, and \( Y \) is the Young modulus. The corresponding elastic energy reads:

\[
F = \frac{1}{2Y} \int dA \left( \nabla^2 \chi \right)^2 .
\]

In the special case where there are no disclinations, and the elastic deformations are only caused by the Gaussian curvature, we denote the solution of Eq. (1) as \( \chi^G(x) \) and the corresponding stress tensor as \( \sigma^G_{ij}(x) \).

The first step in determining the energetics of our system is to solve Eq. (1) for \( \chi^G(x) \) in the absence of defects. This can be achieved in two steps [2, 4]. We first introduce an auxiliary function \( \rho(x) \) that satisfies:

\[
\nabla^2 \rho(x) = G(x) .
\]

The second step is to set \( \rho(x) \) as a source in the Poisson equation for \( \chi^G(x) \)

\[
\frac{1}{Y} \nabla^2 \chi^G(x) = -\rho(x) + \rho_H(x) ,
\]

where \( \rho_H(x) \) a harmonic function that is used to satisfy boundary conditions.

The covariant Poisson equations (3–4) can be solved by the method of conformal maps [6], as illustrated for capillary bridges in the next sections.

I. GEOMETRY OF CAPILLARY BRIDGES

We now proceed to write down explicit expressions for the metric properties of capillary bridges that are required for our elastic calculations. The capillary bridges are surfaces of constant mean curvature known as unduloids, nodoids and catenoids. In Supplementary materials 2 we show that their shape is well approximated by a surface of revolution whose height \( r(z) \) is a function of the height \( z \):

\[
r(z) = \left( R_\phi - R_z \right) + R_z \cosh \left( \frac{z}{R_z} \right) ,
\]

where \( R_z \) and \( R_\phi \) are the radii of curvature at the neck in the vertical and azimuthal directions respectively. For later use, we define the half height of the capillary bridge to be \( H/2 \). Equation (5) is more amenable to analytical treatment than the full expressions for unduloids and nodoids. Note that the case \( R_z = R_\phi \) corresponds to a catenoid, which has constant vanishing mean curvature.

The corresponding metric is:

\[
ds^2 = \cosh \left( \frac{z}{R_z} \right)^2 dz^2 + \left( R_\phi - R_z + R_z \cosh \left( \frac{z}{R_z} \right) \right)^2 d\phi^2
\]

Upon a change to conformal coordinates the metric can be readily recast in the convenient form [6]

\[
ds^2 = e^{-2\rho(x)} (dv^2 + du^2) ,
\]

where the conformal factor \( \omega \) is given by

\[
\rho(z) = -\log \left( \frac{R_\phi - R_z + R_z \cosh \left( \frac{z}{R_z} \right)}{R_\phi} \right) .
\]

The corresponding Gaussian curvature is:

\[
G(z) = -\frac{\sech^3 \left( \frac{z}{R_z} \right)}{R_z \left( (R_\phi - R_z) + R_z \cosh \left( \frac{z}{R_z} \right) \right)} .
\]

Note that the covariant Laplacian of the conformal factor is equal to the Gaussian curvature [6], see Eq. (3).

II. GEOMETRIC FORCE ON AN ISOLATED DISLOCATION

In this section, we present two complementary approaches to calculating the geometric force experienced by an isolated dislocation starting with the Airy stress function \( \chi^G(x) \). The first is based on considering the Peach-Koeler force on a dislocation, induced by the elastic pre-stress \( \sigma^G_{ij}(x) \) that the curvature generates prior to the introduction of defects [2]. The second relies on an effective theory of disclinations in curved space [3].

A. Geometric Peach-Koehler force

According to standard elasticity theory, a dislocation in an external stress field \( \sigma_{ij}(x) \) experiences a Peach-Koeler force, \( f_k(x) \), given by

\[
f_k(x) = \epsilon_{kj} b_l \sigma_{ij}(x) ,
\]

where \( \epsilon_{kj} \) is the Levi-Civita symbol.
where \( \vec{b} \) is the Burgers vector of the dislocation. A dislocation introduced into the curved 2D crystal will experience a Peach-Koehler force as a result of the pre-existing stress field of geometric frustration \( \sigma_{ij}^G(x) \). The stress tensor \( \sigma_{ij}^G(x) \) in an arbitrary set of curvilinear coordinates \( x = \{x_1, x_2\} \) can be expressed in terms of \( \chi^G(z) \) and the metric tensor \( g_{ij} \) as:

\[
\sigma_{11} = \frac{\tau_{11}}{g_{11}}, \quad \sigma_{22} = \frac{\tau_{22}}{g_{22}}, \quad \sigma_{12} = \frac{\tau_{12}}{\sqrt{g_{11}g_{22}}},
\]

(11)

where \( \tau_{ij} = \gamma^{mn} \gamma_{jp} g_{im} g_{pr} (\partial_n \partial_r \chi^G(z) - \Gamma^q_{nr} \partial_q \chi^G(z)) \).

The Christoffel symbols are denoted by \( \Gamma^k_{ij} \) and \( \gamma_{ij} = \varepsilon_{ij}/\sqrt{g} \) is the covariant antisymmetric tensor, with the determinant of the metric \( g = g_{11}g_{22} \).

For the capillary bridges, we can calculate \( \sigma_{zz}^G(z) \) from Eqs. (11-12) giving:

\[
\sigma_{zz}^G(z) = \frac{\text{sech} \left( \frac{z}{R_z} \right) \tanh \left( \frac{z}{R_z} \right)}{\left( R - R_z \right) + R_z \cosh \left( \frac{z}{R_z} \right)} \partial_z \chi^G,
\]

(13)

and

\[
\sigma_{\phi\phi}^G(z) = \frac{1}{\cosh^2 \left( \frac{z}{R_z} \right)} \left[ \partial^2_z \chi^G - \frac{1}{R_z} \tan \left( \frac{z}{R_z} \right) \partial_z \chi^G \right].
\]

(14)

If we choose \( \vec{b} \) along its minimum-energy orientation (azimuthal, corresponding to a dislocation having its 7-fold defect closer to the neck), we obtain a Peach-Koehler force \( f_z(z) \) pointing in the \( z \) direction away from the neck and of magnitude given by:

\[
f_z(z) = -b \sigma_{\phi\phi}^G(z).
\]

(15)

As we shall see, pleats and dislocations appear in the regime of small integrated Gaussian curvature \( H/R_z \ll 1 \), for which the capillary bridge can be viewed as a weakly deformed cylinder. In this regime, we Taylor expand all previous expressions in powers of \( z/R_z \) to find a closed-form solution for \( \chi^G(z) \) in Eq. (4), subject to the free boundary condition \( \sigma_{zz}^G(z = \pm H/2) = 0 \).

The dislocation can be regarded as a charge neutral pair of disclinations whose dipole moment \( q \) is a lattice vector perpendicular to \( \vec{b} \) that connects the two points of 5 and 7-fold symmetry along the surface. The geometric potential of a dislocation can then be obtained by taking the product of \( b \) times the gradient of \( U(z) \) in the \( z \) direction, including the factor \( \sqrt{g_{zz}} = \cosh(\frac{z}{R_z}) \).

The force \( f_z(z) \) follows upon taking a second gradient with the result:

\[
f_z(z) = -\frac{1}{\sqrt{g_{zz}}} \partial_z \left[ \frac{d}{\sqrt{g_{zz}}} \partial_z U \right].
\]

(21)

One can easily verify that Eq. (21) is equivalent to Equations (15-14), once the identification \( U(z) = q \chi^G(z) \) is made. This gives an explicit expression for the geometric potential \( U(z) \) of an isolated disclination of topological charge \( q \) in the limit of small deformations, see Eq. (22):

\[
U(z) = -\frac{q Y}{24 R_z R_{\phi}} \left( z^4 - \frac{H^2 z^2}{2} + \frac{H^4}{16} \right).
\]

(22)

Negative (positive) disclinations are attracted (repelled) from the neck of the capillary bridge at \( z = 0 \). If the negative (positive) disclination within the dipole is closer to the neck, the total energy is negative (positive). This means that a dislocation in its minimum energy orientation is aligned with its negative disclination facing the neck at \( z = 0 \).

### III. Energetic Criteria for Pleating

We now estimate the threshold integrated Gaussian curvature required to make pleating energetically favorable. The first elastic instability, observed in the experiment of Fig. 4 lower panel, is the appearance near the

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**B. Effective theory of disclinations in curved space**

The energy of a distribution of unbound disclinations with “topological charges” \( \{q_\alpha = \pm \frac{2\pi}{\alpha} \} \) reads

\[
F = \frac{Y}{2} \int dA \int dA' N(x) \frac{1}{\Delta_{xx'}} N(x') \,,
\]

(18)

where \( \frac{1}{\Delta_{xx'}} \) is the Green’s function of the biharmonic operator and the source \( N(x) \) reads

\[
N(x) = \sum_{\alpha} q_\alpha \delta(x, x^\alpha) - G(x) \,.
\]

(19)

Inspection of Eq. (18) reveals that the potential \( U(z) \), experienced on a curved surface by an isolated disclination of topological charge \( q = \pm \frac{2\pi}{\alpha} \) satisfies the same biharmonic equation as the Airy stress function \( \chi^G(z) \) namely

\[
\nabla^4 U(z) = -q G(z) \,.
\]

(20)

The dislocation can be regarded as a charge neutral pair of disclinations whose dipole moment \( q \) is a lattice vector perpendicular to \( \vec{b} \) that connects the two points of 5 and 7-fold symmetry along the surface. The geometric potential of a dislocation can then be obtained by taking the product of \( b \) times the gradient of \( U(z) \) in the \( z \) direction, including the factor \( \sqrt{g_{zz}} = \cosh(\frac{z}{R_z}) \).

The force \( f_z(z) \) follows upon taking a second gradient with the result:

\[
f_z(z) = -\frac{1}{\sqrt{g_{zz}}} \partial_z \left[ \frac{d}{\sqrt{g_{zz}}} \partial_z U \right].
\]

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One can easily verify that Eq. (21) is equivalent to Equations (15-14), once the identification \( U(z) = q \chi^G(z) \) is made. This gives an explicit expression for the geometric potential \( U(z) \) of an isolated disclination of topological charge \( q \) in the limit of small deformations, see Eq. (22):

\[
U(z) = -\frac{q Y}{24 R_z R_{\phi}} \left( z^4 - \frac{H^2 z^2}{2} + \frac{H^4}{16} \right) \,.
\]

(22)

Negative (positive) disclinations are attracted (repelled) from the neck of the capillary bridge at \( z = 0 \). If the negative (positive) disclination within the dipole is closer to the neck, the total energy is negative (positive). This means that a dislocation in its minimum energy orientation is aligned with its negative disclination facing the neck at \( z = 0 \).
top and bottom boundaries of a pair of dislocations with opposite Burgers vectors oriented so that the 7s face the neck of the catenoid. The onset of this instability is calculated below by applying the formalism detailed in the previous section to the experimentally relevant limit of small deformations from a cylindrical geometry.

A. Curvature induced unbinding of dislocations

When the two dislocations appear in the system, they will be located at the minimum energy positions \( z_{\text{min}} \approx \pm H/\sqrt{12} \), where the Peach-Koheler force of Eqs. (15) and (17) vanishes [8]. The difference in energy, \( \Delta E \), between the defected curved crystal and the curved crystal without any defects is given by

\[
\Delta E = \frac{Y b^2}{4\pi} \ln \left( \frac{2z_{\text{min}}}{a} \right) - 2 \int_0^{z_{\text{min}}} f(z') dz' + 2E_c \ , \quad (23)
\]

where \( a \) is the lattice spacing and \( E_c \) is the core energy of each dislocation. The first term represents the interaction energy between two dislocations and the second is twice the geometric potential of a dislocation interacting with curvature. We can obtain an explicit expression for the latter by substituting Eq. (17) into Eq. (23)

\[
- \int_0^z f(z') dz' \approx \frac{Y b}{2R_c R_\phi} \left( \frac{z^3}{3} - \frac{H^2}{12} z \right) \ . \quad (24)
\]

When \( \Delta E \leq 0 \), the nucleation of a pair of oppositely oriented dislocations occurs [9]. Evaluating Eq. (24) at \( z_{\text{min}} \approx H/\sqrt{12} \) leads to the threshold for the instability to occur:

\[
\frac{\pi}{9\sqrt{3}} \frac{H^2}{R_\phi R_z} \approx \frac{b}{H} \ln \left( \frac{H}{a'} \right) , \quad (25)
\]

where \( a' = \sqrt{3/2e^{-8\pi E_c/Y b^2}} \). Since the Burgers vector \( b \) is of the order of the lattice spacing \( a \) we obtain, after a simple rearrangement,

\[
\frac{2\pi R_c H}{R_\phi R_z} \approx \frac{a}{H} \ln \left( \frac{H}{a'} \right) \frac{18 R_\phi}{\sqrt{3} H} , \quad (26)
\]

which can be rewritten as a threshold integrated Gaussian curvature:

\[
\left| \int G \, dA \right| \sim \frac{a}{H} \ln \left( \frac{H}{a'} \right) \frac{R_\phi}{H} . \quad (27)
\]

The modulus sign arises from the fact that the curvature of the bridge is negative. The geometric factor \( \frac{a}{H} \) is typically \( \mathcal{O}(1) \) in our experiments and so are the logarithmic corrections arising from dislocations interactions.

B. Nucleation of pleats

In this section we estimate the threshold for the appearance of pleats in the limit that the spacing between dislocations in the pleat is very large and find that it agrees with the estimate made above in Eq. (27). In both cases there is an associated energetic cost that diverges logarithmically with the size of the system. We start by estimating the energy \( E_f \) of a frustrated crystal characterized by a bond angle rotation \( \Delta \theta \) as

\[
E_f \sim Y \Delta \theta^2 H^2 , \quad (28)
\]

where the angular strain \( \Delta \theta \) is equal to the integrated Gaussian curvature. The energy \( E_p \) of a dilute pleat, whose dislocation spacing is of order \( H \), can be estimated from the familiar expression for a low angle grain boundary, which to leading order reads [7]

\[
E_p \sim -Y b H \Delta \theta \ln (\Delta \theta) . \quad (29)
\]

In order to optimize the screening of the underlying curvature, the angle of the grain boundary \( \Delta \theta \) must be equal in magnitude to the angular deficit introduced by the curvature. When the angle \( \Delta \theta \) is small \( E_f \sim \Delta \theta^2 \) is lower than \( E_p \). Pleat nucleation becomes energetically favorable above a threshold \( \Delta \theta_c \) obtained by balancing the energies in Equations (28) and (29) with the result

\[
\Delta \theta_c \sim \frac{b}{H} \ln (\Delta \theta_c) . \quad (30)
\]

Upon solving Eq. (30) self consistently (neglecting doubly logarithmic corrections) one obtains

\[
\Delta \theta_c \sim \frac{b}{H} \ln \left( \frac{H}{b} \right) . \quad (31)
\]

Upon explicitly setting the bond angle rotation equal to the magnitude of the integrated Gaussian curvature and the Burgers vector to a length of order lattice spacing \( a \) we obtain

\[
\left| \int G \, dA \right| \sim \frac{a}{H} \ln \left( \frac{H}{a} \right) , \quad (32)
\]

which provides an estimate of the same order as Eq. (27).


[2] V. Vitelli, J.B. Lucks, and D.R. Nelson, PNAS 103, 12323,
[8] The interaction between the dislocations is not sufficient to displace them significantly from their equilibrium positions since $b \ll H, z_{min}$.
[9] Note that the total energy is positive definite because in both cases there is an energy of geometric frustration that depends only on the curvature and not on the defects and arises in Equations (18-19) from setting all $q_\alpha = 0$. 
FIG. 1: The counting of topological charge or defects in a hexagonal lattice and the associated constraints on total number of defects can be done in terms of the number of nearest neighbours of each particle and the counting that Euler, Descartes and Poincare taught us. For any triangulated surface \( V - E + F = \chi \), where \( V \), \( E \) and \( F \) are respectively the number of vertices, edges and faces and \( \chi \) is the Euler characteristic, an invariant for topologically equivalent surfaces. For a cube (or a sphere) \( \chi = 2 \), remove a face to get a surface with a boundary, e.g. a box without a lid, a dome or a disk and \( \chi = 1 \). Remove two faces to obtain a surface with two boundaries, such as a cylinder, and \( \chi = 0 \). \( \chi \) is related to topological charge as follows: for an \( n \)-coordinated vertex there are \( n \) edges shared with two neighbors and \( n \) faces shared with three neighbors. The contribution to the Euler sum of each vertex is therefore \( 1 - n/2 + n/3 = (6 - n)/6 \). Hexagons contribute nothing, pentagons \(+1/6\), heptagons \(-1/6\). A four-fold coordinated straight edge contributes 0, a three-fold has charge \( +1/6 \), a five-fold has charge \( -1/6 \). A disk and a dome which are topologically equivalent (since a continuous deformation takes one to the other) must have net charge 6 to give \( \chi = 1 \), but here the charges may reside entirely on the edge or entirely on the face or some on each. The bottom left panel above shows three relaxed bounded surfaces with \( \chi = 1 \), a flat patch with \( +1/6 \) disclination on the edge, a positively curved patch with \( 5 +1/6 \) s on the edges and one on the surface and a negatively curved surface with one \(-1/6\) on the surface and \( 7 +1/6\)s on the edge.
Supplementary Movies 1 and 2 - Crystal dynamics on a pulled capillary bridge

I. SUPPLEMENTARY MOVIE 1 - RAW CONFOCAL DATA

Raw confocal data for an experiment in which we pulled on a quasi-cylindrical capillary bridge. Since the boundary of the bridge is pinned and its volume conserved, its shape changes from almost cylindrical to highly curved as the top and bottom plates are pulled apart. The reconstructed data, showing the defect dynamics can be seen in Supplementary movie 2

II. SUPPLEMENTARY MOVIE 2 - RECONSTRUCTED DATA

This movie shows the reconstructed crystal structure from the confocal data of supplementary movie 1, in which we pulled on a quasi-cylindrical capillary bridge. Since the boundary of the bridge is pinned and its volume conserved, its shape changes from almost cylindrical to highly curved as the top and bottom plates are pulled apart. The reconstruction allows us to follow the introduction of defects as the curvature increases.

The crystal starts from a configuration with very few defects. As the glass plates are pulled apart and the crystal adjusts to the growing negative Gaussian curvature and to its new height, dislocations appear on the surface. We have introduced pauses in the movie to emphasize the appearance of dislocations polarized to screen the curvature of the system. As the (negative) curvature is increased dislocations, dominantly polarized with their - disclinations pointing toward the region of highest negative curvature, the waist of the capillary bridge, appear. As the integrated negative curvature is increased these polarized dislocations proliferate and align, forming ‘pleats’ (See Fig. 4 in the manuscript). Finally, as the curvature is increased, disclinations appear on the surface.

In addition to the dislocations polarized to interact with the curvature, we observe unpolarized dislocation pairs whose role is to bring about an increase in the number of rows as the capillary bridge is stretched.

In material terms, dislocations mark the point in which two semi-infinite rows of atoms terminate in bulk. Conventionally, dislocation glide, motion in the direction perpendicular to the line joining + and -, facilitates shear and deformation of the solid, adding rows of ‘atoms’. As the height of the capillary bridge is increased, the number of rows that fits vertically also increases, the creation of dislocation pairs which glide from top to bottom brings about this increase.
FIG. 1: Oppositely polarized dislocation pairs glide to change the number of vertical rows in the crystal.

The dislocations whose role is to screen Gaussian curvature (discussed above) are instead aligned along the cylindrical axis, do not glide or serve to increase the number of rows and are dominantly polarized with their ‘-’ disclinations pointing toward the region of highest negative curvature, the waist of the capillary bridge.