Shape bistability in 2D chromonic droplets

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Abstract

An extensive experimental study of the shapes of two-dimensional bipolar droplets of the chromonic nematic phase of disodium cromoglycate (DSCG) sandwiched between glass plates, by Kim et al was published in (2013 J. Phys.: Condens. Matter 25 404202). The paper includes a mathematical model of this system. We have extended this study by further theoretical modelling. Our results are in good, quantitative agreement with the experimental data. The model has produced what promises to be a more accurate estimate for the isotropic surface tension at the nematic/isotropic solution interface—and predicts a regime of shape bistability (which has not yet been observed) for larger droplets, where tactoids (pointed, zeppelin-shaped droplets) and smooth-edged discoids can coexist in equilibrium. The general method presented in this paper is also applied to the tactoids formed by F-actin filaments in solution, for which an estimate is given for the value of the isotropic surface tension at the nematic/isotropic interface.

Keywords: bistability, droplets, chromonic liquid crystals, lyotropic liquid crystals, surface tension, F-actin filaments

(Some figures may appear in colour only in the online journal)

1. Introduction

Chromonic liquid crystals (CLCs), also called chromonics, form a very peculiar class of liquid crystals (LCs). These latter are systems constituted by basic mesogens (either single molecules or molecular assemblies), which possess orientational ordering, and sometimes—under appropriate circumstances—also positional ordering. Basically, there exist two classes of LCs: thermotropic LCs, in which the ordering process is temperature-driven, and lyotropic LCs, in which the mesogenic units are molecular assemblies in a solvent, and concentration is the driving parameter for ordering. These latter are found in detergents and living organisms alike [1].

In particular, CLCs are lyotropics formed by certain dyes, drugs, and short nucleic-acid oligomers in aqueous solutions [2–8]. Since most biological processes take function normally in these types of solutions, it is no wonder that interest in CLCs has recently surged for possible applications in medical sciences. But this is not the only reason that makes them special (or rather unique). A number of informative, updated reviews are available on this topic [2, 9–12]; they are all highly recommended.

In conventional amphiphile/water systems, the temperature/composition phase diagrams are often complex—and can show a wide range of patterns of aggregation—spherical micelles, cylindrical columns, layered structures complex cubic phases—with the additional factor of there being both oil-in-water and water-in-oil inverse structures. What perhaps distinguishes CLCs is that aggregation starts at very low concentrations and that aggregates are columns, although with variants [2]. Some are stacks of single molecules, others have more than a molecule in their cross-sections [1]. These might seem to be minor details, but they may have momentous consequences at the macroscopic scale [13].

More specifically, CLC molecules are typically plank-shaped with aromatic cores and polar groups on their
peripheries [14]. They tend to stack face-to-face and their aggregates order into a fluid nematic phase or (for higher concentrations or lower temperatures) even in a solid-like medium phase, where columns are organised parallel to one another with their centres arranged in an hexagonal pattern [2].

It is the variability in size and shape of the supra-molecular columns that makes CLCs so unique. The aggregation process is isodesmic, as the energy gain in adding a unit to a preexisting column (typically between 5 and 10 kT) does not depend on the length of the column. The isodesmic nature of the process results in a broad length column distribution, which is prone to the action of temperature. When the temperature is increased, the concentration of longer assemblies decreases. This is reflected by the elastic properties of the phase, in a way that ordinary lyotropics do not exhibit [14]. Further increasing the temperature results into a first order nematic-isotropic transition with a wide coexistence region (5–10°C). Conversely, when the temperature is decreased, short, disordered columns in the isotropic phase tend to grow and aggregate, eventually separating from the parent isotropic solution to form islands of ordered phase.

As customary, the nematic phase is described by the extra director field \( n \), which represents at the macroscopic level the local orientation of the supra-molecular rod-like aggregates. At the nematic/isotropic interface, an orientation-dependent surface tension arises that favours the (degenerate) tangential orientation of \( n \), in agreement with the purely entropic argument of Onsager [15], according to which a tangential anchoring would enhance the local translational freedom of columns at the interface.

We shall only be concerned with a two-dimensional problem, inspired by the experimental setting explored by Kim et al. [16]. There, CLC droplets in the nematic phase appeared surrounded by the isotropic phase, their shape being the characteristic tactoids (spindle-like) that were first observed by Zocher [18] in aqueous solutions of monocrystals of vanadium pentaoxide. They were later also seen in aqueous dispersions of tobacco mosaic viruses by Bernal and Fankuchen [19].

Most droplets reported in [16] were bipolar, with point defects of \( n \) at the pointed poles. In this paper, we adapt to the two-dimensional setting our theory for the representation of tactoids recently presented in [20]. We shall achieve two main results.

First, we predict a shape bistability, which seems characteristic of the two-dimensional setting. We identify a range of droplet’s areas where two distinct shapes could be observed, one tactoidal, as expected, and the other discoidal (smooth), both bearing a bipolar arrangement of \( n \). A sort of shape coexistence thus parallels the phase coexistence observed in these materials. This regime manifests itself for droplets larger than those reported in [16]; to our knowledge, it has not yet been observed.

Second, we use the very detailed data of [16] to compare the observed shapes with those predicted by our theory. We extract an estimate for the isotropic component of the surface tension at the droplet’s interface, which turns out to be comparable in order of magnitude to the typical values measured for standard thermotropic LCs (~ 10 μN m\(^{-1}\), see [21, p 495]).

To put our theory to the test for another lyotropic system, we also consider the bipolar tactoids observed in solutions of F-actin filaments [22]. For the isotropic surface tension, we estimate a value three orders of magnitude less than for CLCs, but comparable to that measured for some phase-separated colloid-polymer dispersions [23].

The paper is organized as follows. In section 2, we recall from [20] our theory and adapt it to the present two-dimensional setting. The optimal shapes of bipolar droplets that minimize the total free-energy functional are derived and discussed in section 3, where we illustrate in detail the bistability scenario that we envision. We show, in particular, how the critical values of the droplet’s area that delimit the corresponding shape hysteresis depend on the elastic constants of the material. Section 4 is devoted to the comparison with experiments. We contrast our predicted shapes to the observed ones and, encouraged by their agreement, we estimate the isotropic component of the surface tension for both CLCs and F-actin filaments. Finally, in section 5, we summarize our conclusions and comment on possible further extensions of our study. The paper is closed by three mathematical appendices, where we collect computational details and auxiliary results needed in the main text, but inessential to its comprehension.

2. Two-dimensional setting

Here, we set our theoretical scene; we shall first recall the energetics of a CLC drop squeezed between two parallel plates and we shall then describe both its outer profile and inner direction field.

2.1. Energetics

In classical liquid crystal theory, the nematic director field \( n \) describes the average orientation of the molecules that constitute the material. The elastic distortions of \( n \) are locally measured by its gradient \( \nabla n \), which may become singular where the director exhibits defects, that is, discontinuities in the field \( n \). The bulk free energy is the following functional,

\[
\mathcal{F}_b[\mathcal{B}, n] := \int_{\mathcal{B}} f_{\text{OF}}(n, \nabla n) \, dV, \tag{1}
\]

where \( \mathcal{B} \) is the region occupied by the material, \( dV \) is the volume element, and \( f_{\text{OF}} \) is the Oseen–Frank free-energy density. Letting the latter be the most general frame-indifferent function, even in \( n \) and quadratic in \( \nabla n \), one arrives at (see, e.g., [24, chapter 3])

\[
f_{\text{OF}} := \frac{1}{2} K_{11} (\text{div} \, n)^2 + \frac{1}{2} K_{22} (n \cdot \text{curl} \, n)^2 + \frac{1}{2} K_{33} |n \times \text{curl} \, n|^2 + K_{24} \left(\text{tr}(\nabla n)^2 - (\text{div} \, n)^2\right), \tag{2}
\]
where $K_{11}, K_{22}, K_{33}$, and $K_{24}$ are the *splay, twist, bend,* and *saddle-splay* elastic constants, respectively. They are material moduli characteristic of each nematic liquid crystal, corresponding to four independent orientation fields, each igniting a single distinctive term in (2).²

The energy $f_{OF}$ is meant to penalize all distortions of $n$ away from the uniform alignment (in whatever direction); to this end, the elastic constants in (2) must satisfy Ericksen’s inequalities [27],

$$K_{11} \geq K_{22} \geq 0, \quad K_{22} \geq K_{24} \geq 0, \quad K_{33} \geq K_{24}. \quad (3)$$

CLCs in three-dimensional space exhibit a different behaviour: in cylindrical capillary tubes subject to degenerate tangential boundary conditions³, the director $n$ has been seen to steer away from the uniform alignment along the cylinder’s axis [28–32]. Two symmetric twisted configurations (left- and right-handed) have been observed in capillaries, each variant corresponding to four independent orientation fields, each igniting a single distinctive term (2).²

Despite the clear indication that CLCs’ ground state differs from the uniform alignment presumed in $f_{OF}$, the Oseen–Frank theory has been applied to rationalize the experiments with capillary tubes [28–31], at the cost of taking $K_{22} < K_{24}$, which violates one of Ericksen’s inequalities (3). Since such a violation would make $f_{OF}$ unbounded below, the legitimacy of these theoretical treatments is threatened by a number of mathematical issues that will be tackled elsewhere [33]. Here, we can spare this trouble, as our setting is two-dimensional and neither $K_{22}$ nor $K_{24}$ play a role. The region $\mathcal{B}$ will be a thin blob representing a droplet of CLC surrounded by the isotropic solution, squeezed between two parallel plates at the distance $h$ from one another.

Formally, $\mathcal{B} = \mathcal{R} \times [-\frac{h}{2}, \frac{h}{2}]$, where $\mathcal{R}$ is a region with piecewise smooth boundary $\partial \mathcal{R}$ in the $(x, y)$ plane of a Cartesian frame $(e_x, e_y, e_z)$ (see figure 1). The field $n$ lies in the same plane as $\mathcal{R}$ and it is thought of as being extended uniformly through the blob’s thickness. For a planar field of this sort, the twist term in (2) vanishes identically, as $\text{curl}(n \parallel e_z)$, as also does the saddle-splay term (see appendix A). Thus, leaving aside the possibly controversial issue arising in general from the violation of one of Ericksen’s inequality, we remain assured that in the present setting the Oseen–Frank energy-density $f_{OF}$ is bounded below and the free energy $\mathcal{F}_b$ can be safely minimized.

We assume that a given mass of material constitutes the blob $\mathcal{B}$, and so its volume $V_0$ is prescribed by the incompressibility constraint (which is satisfied to a large degree of approximation). Consequently, the area $A_0$ of $\mathcal{R}$ is prescribed, as $V_0 = A_0 h$. We further assume that the plates bounding both $\mathcal{B}$ and the surrounding isotropic solution exert a degenerate tangential anchoring on the nematic director $n$, so that, in light of the constraint on the area of $\mathcal{R}$, the additional anchoring energy can be treated as an inessential additive constant.

This is not the case for the surface energy $\mathcal{F}_s$ at the interface between $\mathcal{R}$ and the surrounding isotropic solution. Here we assume that this energy is represented by the Rapini–Papoular formula [34],

$$\mathcal{F}_s[\mathcal{R}, n] = h \int_{\partial \mathcal{R}} \gamma(1 + \omega (n \cdot \nu)^2) d\ell, \quad (4)$$

where $\gamma > 0$ is the isotropic surface tension of the liquid crystal in contact with the isotropic solution, $\omega$ is a dimensionless anchoring strength, which we take to satisfy $\omega \geq 0$, $\nu$ is the outer unit normal to $\partial \mathcal{R}$, and $d\ell$ is the length element. Thus $\mathcal{F}_s$ is minimized when $n$ lies tangent to $\partial \mathcal{R}$. Henceforth, we shall enforce this minimum requirement as a constraint on $n$,

$$n \cdot \nu = 0, \quad \text{on } \partial \mathcal{R}, \quad (5)$$

save checking ultimately this assumption with appropriate energy comparisons (see appendix B) which ensure that such a tangential anchoring is not broken. In short, the validity of (5) requires that the droplet is not too small in a sense that will be made precise below.

To sum up, the total free energy $\mathcal{F} := \mathcal{F}_b + \mathcal{F}_s$ of the system is given by the functional

$$\mathcal{F}[\mathcal{R}, n] := h \left( \int_{\mathcal{R}} f_{OF} \, dA + \gamma \ell(\partial \mathcal{R}) \right), \quad (6)$$

subject to (5) and to the isoperimetric constraint

$$A(\mathcal{R}) = A_0, \quad (7)$$

where $A$ and $\ell$ are the area and length measures, respectively.

2.2. Admissible shapes

No preferred direction is present on the substrates that bound the drop, and so, contemplating tactoids among the possible equilibrium shapes of $\mathcal{R}$ (as well as other smooth shapes), we assume that these are mirror symmetric about two orthogonal axes, one joining the possible sharp tips of the boundary $\partial \mathcal{R}$.

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²Recently, an equivalent modal decomposition has been put forward for $f_{OF}$ [25], which has also been given a graphical representation in terms of an octupolar tensor [26].

³That is, when $n$ is bound to be tangent to the boundary, but oriented in any direction.
obtained by mirror symmetry about the orthogonal field, and axis of the drop, $\nu$ everywhere tangent to the retracted curves; $\nu = -n_\perp$. The function $R$ represents half a drop; the other half is obtained by mirror symmetry about the $y$ axis.

We denote by $y$ the latter axis and by $x$ the orthogonal symmetry axis\(^4\). Thus, only half of the curve that bounds $R$ needs to be described, the other half being obtained by mirror symmetry. We take this curve to be represented as $x = R(y)$, where $y$ ranges in the interval $[-R_0, R_0]$, with $R_0$ to be determined, and $R$ is a smooth, even function such that

$$R(R_0) = 0 \quad \text{and} \quad R'(0) = 0,$$

where a prime $'$ denotes differentiation with respect to $y$ (see figure 2). The points where $R$ vanishes correspond to the poles of $R$. Whenever $R(R_0)$ is finite, $R$ represents a tactoid, as the outer unit normal $\nu$ to $\partial \mathcal{A}$ is discontinuous at the poles. Smooth shapes correspond to $R(R_0) = -\infty$.

We call $R_0$ the radius of the equivalent disc, which has area $A_0$, and we denote by $\mu$ the dimensionless length of the semi-axis of the drop,

$$\mu := \frac{R_0}{R_c}. \quad \text{(9)}$$

Hereafter, we shall rescale all lengths to $R_c$ (while keeping their names unchanged, to avoid typographical clutter). With this normalization, the area constraint (7) reads simply as

$$\int_{-\mu}^{\mu} R(y) \, dy = \frac{\pi}{2}, \quad \text{(10)}$$

\(^4\) Clearly, due to the absence of a privileged orientation on the bounding substrates, the $y$ axis could indeed be oriented in any direction. If several drops are present, their axes would be isotropically distributed.

2.3. **Director retraction**

The shape of $\mathcal{A}$ is unknown and needs to be determined. Since $n$ is tangent to $\partial \mathcal{A}$, following [20] we devise a method that also derives $n$ inside $\mathcal{A}$ from the knowledge of $\partial \mathcal{A}$, thus reducing the total free energy $F$ to a pure shape functional. This is achieved by retracting $\partial \mathcal{A}$ inside $\mathcal{A}$ with its tangent field $n$.

Formally, we define a function $R_t(y) := g(t)R(y)$, where $t \in [0, \, 1]$ and $g$ is an increasing monotonic function such that $g(0) = 0$ and $g(1) = 1$. The graph of $R_t$, shown in figure 2, represents the retraction of $\partial \mathcal{A}$ that borders an inner domain $\mathcal{A}_t \subseteq \mathcal{A}$. All domains $\mathcal{A}_t$ are nested one inside the other as $t$ decreases towards 0. For $t = 0$, $\mathcal{A}_t$ reduces to the $y$ axis. The advantage of this method is that it also affords to describe the inner director $n$ as the field everywhere tangent to the family of curves $\partial \mathcal{A}_t$. All director fields obtained by this geometric construction are bipolar, in that they have two point defects at the poles; in the language of Mermin [35], they are boojums with topological charge $m = +1$ (see also [21, p 501]).

It is shown in appendix A how to compute the area element $dA$ in the $(t, \, y)$ coordinates and how to express $\nu \mathbf{n}$ in the orthonormal frame $(\mathbf{n}, \, n_\perp)$, where $n_\perp := e_x \times n$, in terms of the functions $R(y)$ and $g(t)$.

In the rescaled variables $y$ and $R(y)$, an appropriate dimensionless form of $F$ in (6) is then given by

$$F[\mu; \, R] := \frac{F[\partial \mathcal{A}]}{K_{11} R_t^2}$$

$$= \int_{-\mu}^{\mu} \left\{ \frac{R'}{R} - \frac{R''}{R^2} + \frac{1}{8} \frac{RR''}{R^2} (3 + k_3) \right\} \times \arctan \left( \frac{R''}{R} + \frac{1}{1 + R^2} + \frac{1}{8} \frac{RR''}{R^2} (1 + R^2)^2 \right)$$

$$\times \left[ (k_3 - 5) - \frac{1}{R^2}(k_3 + 3) \right] \right\} + 2 \alpha \sqrt{1 + R^2} \, dy,$$

where

$$k_3 := \frac{K_{33}}{K_{11}} \quad \text{(12)}$$

is a reduced bend constant and

$$\alpha := \frac{\gamma R_c}{K_{11}} \quad \text{(13)}$$

is a reduced area. Equivalently, $\alpha = R_c/\xi_e$, where $\xi_e$ is the de Gennes–Kleman extrapolation length [21, p 159].\(^5\)

The reduced functional $F$ in (11) suffers from a typical pathology of two-dimensional director theory: it diverges logarithmically to $+\infty$ near defects. Here, the culprit is the integrand $\frac{1}{1 + R^2} \arctan R$, which is not integrable at $y = \pm \mu$. Following a well established practice (see, e.g., [36, p 171]), we imagine that the energy concentration near defects causes a localized transition to the isotropic phase, which constitutes a defect core (whose fine structure is better explored within

\(^5\) Thus, a drop is either small or large, whether $\alpha \ll 1$ or $\alpha \gg 1$, respectively.
out of scale, is defined by (14). A sensible value for the parameter \( \varepsilon \), which here is out of scale, is \( \varepsilon \approx 10^{-3} \). A similar construction also applies to a drop with smooth poles (not shown here).

Ericksen’s theory [37]). The energy associated with such a phase transition is proportional to the core’s area and will be taken as approximately fixed. Moreover, for simplicity, instead of considering a circular core, which in the most common choice, we take it in the shape of the tapering drop’s tip. Letting \( r_c \) denote the core’s size, we set \( r_c = \varepsilon R_c \) and restrict \( y \) to the interval \([-\eta, +\eta]\), where \( \eta \) is defined by

\[
R(\eta) = R(-\eta) = \varepsilon,
\]

and so depends indirectly on \( \varepsilon \) (see figure 3). For \( R_c \) of the order of 10 \( \mu \)m, it is reasonable to take \( \varepsilon \approx 10^{-3} \), as we shall do here, which corresponds to \( r_c \) of the order of 10 nm. The integral in (11) will hereafter be limited to the interval \([-\eta, +\eta]\), so that it will always converge. The extra energy stored in the defects, being approximately constant, will play no role in our quest for the equilibrium shape of squeezed drops.

2.4. Special family of shapes

Here, we follow closely [20], albeit in a two-dimensional setting, in an attempt to restrict the admissible shapes of drops to a special family amenable to a simple analytical treatment. The admissible drop profiles will be described by the function

\[
R(y) = ax^2 - y^2 + b \sqrt{\mu^2 - y^2},
\]

where \( a \) and \( b \) are real parameters that must be chosen subject to the requirements that \( R(y) \geq 0 \) for all \(-\mu \leq y \leq \mu \) and that (10) is satisfied. It is a simple matter to show (see also [20]) that \( a \) and \( b \) can be expressed in terms of the free parameters \((\phi, \mu)\) that span the configuration space \( S := \{(\phi, \mu) : 0 \leq \phi \leq \frac{\pi}{3}, \mu > 0\} \). Precisely,

\[
a = \frac{1}{\mu^3} \pi \cos \phi \left( \frac{1}{h(\phi)} \right), \quad b = \frac{1}{\mu^3} \sin \phi \left( \frac{1}{h(\phi)} \right)
\]

with

\[
h(\phi) := \frac{8}{3} \pi \cos \phi + \pi \sin \phi > 0, \quad 0 \leq \phi \leq \frac{3\pi}{4}.
\]

Shapes with different qualitative features correspond to different regions of \( S \), as illustrated in figure 4. Prolate shapes are characterized by

\[
\mu \geq \varpi(\phi) := \sqrt{\frac{\pi(\cos \phi + \sin \phi)}{h(\phi)}},
\]

whereas oblate shapes are characterized by \( \mu < \varpi(\phi) \). Moreover, shapes represented by (16) are convex for \( 0 \leq \phi \leq \phi_t \) and concave for \( \phi_t < \phi \leq \frac{\pi}{3} \). The latter are represented by the red strip in figure 4; we call them butterflies: their waist narrows as \( \phi \) approaches the boundary of \( S \) at \( \phi = \frac{\pi}{3} \), where it vanishes altogether and the droplet splits in two.

\( \mathcal{R} \) has pointed tips only whenever \( \mathcal{R}'(\mu) \) is finite; according to (16), the only value of \( \phi \) that makes \( b \) vanish is \( \phi = 0 \). We call genuine these tactoids. For small enough values of \( \phi \) the shape represented by (15) via (16) cannot be visually distinguished from pointed tactoids; we find the conventional barrier at \( \phi = \frac{\pi}{3} \) appropriate to delimit the realm of tactoids (genuine or not). Further increasing \( \phi \), \( \mathcal{R} \) has tips that look smoother, justifying our calling them discoids. A conventional barrier is set at \( \phi = \frac{\pi}{3} \) to mark where discoids evolve into little batons, for which we use the French word bâtonnet. The names used to identify different shapes of \( \mathcal{R} \) and the corresponding strips in \( S \) where they are found are recalled in table 1.

Figure 5 illustrates a gallery of shapes for \( \mathcal{R} \) obtained from (15) and (16) for \( \mu = 1 \) and a number of values of \( \phi \) falling in the different types listed in table 1.

3. Optimal shapes

This section is devoted to the study of the minimizers of the free-energy functional in (11). Our major result will be the prediction of a shape bistability, which has not yet been observed in this setting. Before describing this phenomenon, however, we need to make sure that the droplets we consider are not too small for the tangential anchoring condition in (5) to be valid.

3.1. Admissible drop sizes

It is known [39] that drops sufficiently small in three-dimensional space tend to break the director’s tangential anchoring on their boundary, favouring the uniform alignment of \( n \) in their bulk. A simple heuristic argument tells us that a similar breaking would also take place in the present two-dimensional setting.

The elastic cost of the bulk deformation scales as \( Kh \), where \( K \) is a typical elastic constant, while the surface anchoring

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\[\text{Figure 3. Isotropic defect core (in red) at the tip of a drop (only a quarter is shown). In our dimensionless units (rescaled to } R_c, \eta \text{ is defined by (14). A sensible value for the parameter } \varepsilon, \text{ which here is out of scale, is } \varepsilon \approx 10^{-3}. A similar construction also applies to a drop with smooth poles (not shown here).} \]
shapes falling on the left of this stripe are convex. The circular disc is represented by the point \((\pi, 0)\). We also call tactoids the shapes for \(0 \leq \phi \leq \frac{\pi}{16}\) (genuine tactoids, only those for \(\phi = 0\) — red line), discoids those for \(\frac{\pi}{16} \leq \phi \leq \frac{\pi}{4}\), and bâtonnet those for \(\frac{\pi}{4} \leq \phi \leq \phi_c\), see also table 1, and figure 5 for a fuller gallery of shapes. Vertical dashed lines represent the barriers marking transitions from one family of shapes to another.

| Table 1. | We identify four strips in configuration space \(S\), which correspond to four qualitatively different shapes for a region \(\mathcal{R}\) represented by (15) via (16). Here \(\phi_c = \arccot \left(\frac{1}{3}\right) \approx 2.03\). |
| --- | --- | --- | --- | --- |
| Tactoids | Discoids | Bâtonnet | Butterflies |
| \(0 \leq \phi < \frac{\pi}{16}\) | \(\frac{\pi}{16} < \phi < \frac{\pi}{4}\) | \(\frac{\pi}{4} < \phi < \phi_c\) | \(\phi_c < \phi \leq \frac{3\pi}{4}\) |

energy scales as \(\gamma \omega h R_c\), and so \(\alpha\) in (13) estimates the ratio of the latter to the former. Thus, when \(\alpha\) is sufficiently small, the bulk energy becomes dominant and it is minimized by the uniform alignment of \(n\), which breaks the tangential anchoring, undermining our analysis. In appendix B, we perform an energy comparison that provides an estimate for the safeguard vale \(\alpha_\text{s}\) of \(\alpha\), above which tangential anchoring is expected to remain unbroken. We obtained the following explicit formula for \(\alpha_\text{s}\),

\[
\alpha_\text{s} := \frac{2}{\pi} \left( k_3 - 1 - k_3 \ln \frac{2 - \ln \varepsilon}{2 - \varepsilon} \right),
\]

where \(j(\omega)\) is the function defined in (B2). For \(\omega = 5\), which is a choice supported by some evidence\(^7\), \(\alpha_s \approx 0.2k_3 - 0.5(1 + \ln \varepsilon)\). In particular, for \(k_3 = 1\) and \(\varepsilon = 10^{-3}\), \(\alpha_s \approx 3\), which will be our reference choice henceforth. Taking \(K \sim 1 - 10\) pN as typical value for all elastic constants\(^8\) and \(\gamma \sim 10\) \(\mu\)N m\(^{-1}\) as typical value for the surface tension of a chromonic liquid crystal in contact with the isotropic solution\(^9\), by (13) taking \(\alpha > 3\) means taking \(R_c \gtrsim 0.3 - 3\) \(\mu\)m, which provides a lower bound on the admissible size of the droplets that can be treated within our theory.

3.2. Shape bistability

Finding analytically the minima of the reduced free energy \(F_n(\phi, \mu)\), the function defined on the configuration space \(\mathcal{S}\) by computing the functional \(F[\mu; R]\) in (11) on the special family of shapes in (15), is simply impracticable. Thus, for a given choice of the elastic parameter \(k_3\), we evaluated numerically \(F_n\) and we sought its minimizers in \(\mathcal{S}\) for increasing \(\alpha > \alpha_\text{s}\).

We found out that there are two critical values of \(\alpha\), \(\alpha_1\) and \(\alpha_2 > \alpha_1\), such that for either \(\alpha < \alpha_1\) or \(\alpha > \alpha_2\), \(F_n\) attains a single (absolute) minimum in \(\mathcal{S}\), whereas it attains two (relative) minima for \(\alpha_1 \leq \alpha \lesssim \alpha_2\). There is a third critical value, \(\alpha_0 \in (\alpha_1, \alpha_2)\), such that for \(\alpha = \alpha_0\) the two minima of \(F_n\) are equal and its absolute minimizer abruptly shifts from one point in \(\mathcal{S}\) to another. Were the points of \(\mathcal{S}\) to represent the different phases of a condensed system, this scenario would be described as a (first-order) phase transition. In our setting, it

\(^7\) See, for example [16, 40].

\(^8\) This estimate is supported for example by [41] for material such as DSG, SSY, and PBG.

\(^9\) Discordant estimates of \(\gamma\) have been given in the literature [16, 42, 43]. Here we take the average order of magnitude found in these works (see also section 4.1 below for an independent justification of this choice).
more simply describes the (local) stability of two equilibrium shapes for a squeezed drop: for \( \alpha_1 < \alpha < \alpha_2 \), both a tactoid and a discoid are local energy minimizers, the global minimum shifting from the former to the latter at \( \alpha = \alpha_b \). For \( \alpha < \alpha_1 \), the only equilibrium shape is a tactoid, whereas it is a discoid for \( \alpha > \alpha_2 \). The shape bistability exhibited by this two-dimensional system will now be documented in more details.

We start by representing the equilibrium landscape in the language of bifurcation theory. Taking \( \alpha \) as a bifurcation parameter and \( \phi \) as an equilibrium shape representative, in figure 6 we illustrate the minima of \( F_\alpha \) for \( k_3 = 1 \): one falls in \( 0 < \phi < \frac{\pi}{16} \) (blue line), and so it is a tactoid, while the other falls in \( \frac{\pi}{16} < \phi < \frac{\pi}{2} \) (red line) and is a discoid. Solid lines represent global minima, while broken lines represent local minima. Two separate local minima are also global minima for \( \alpha = \alpha_b \), where a perfect bistability is established between the two equilibrium branches. The tectoidal branch can be further continued, as it is locally stable, until \( \alpha \) reaches the critical value \( \alpha_2 \), where it ceases to exist altogether. Similarly, as soon as \( \alpha \) exceeds \( \alpha_1 \), the discoidal branch comes first into life as a locally stable equilibrium, which then becomes globally stable for \( \alpha > \alpha_b \). For \( \alpha \in [\alpha_1, \alpha_2] \), tactoids and discoids coexist as optimal shapes; both are metastable, one or the other is globally stable, according to whether \( \alpha < \alpha_b \) or \( \alpha > \alpha_b \).

The dimensionless parameter \( \alpha \) is ultimately related through (13) to the amount of material trapped in the drop. So the coexistence interval \([\alpha_1, \alpha_2]\) corresponds to a window of areas \( A_0 \) for which two different shapes could be observed, more likely (and frequently) the one corresponding to the global minimum. If, ideally, one could gently pump material into a tectoidal drop, so as to follow the blue branch in figure 6, the drop would continue to grow as a tectoid until the critical volume corresponds to \( \alpha_2 \), where a dynamical instability would presumably prompt the transition towards a discoid. Conversely, if material could be gently removed from the latter, this would keep its discoidal shape until the critical volume corresponds to \( \alpha_1 \), where it would dynamically evolve into a tactoid. The green lines in figure 6 delimit such a hysteresis loop.

Figure 7 illustrates the energy landscape for \( \alpha < \alpha_1 \); \( F_\alpha \) is convex, and so it attains a single minimum, which falls in \( 0 < \phi < \frac{\pi}{16} \), corresponding to a tactoid. The scene changes in figure 8, where \( \alpha = \alpha_b \) and \( F_\alpha \) attains two equal minima, corresponding to a tactoid and a discoid. For \( \alpha > \alpha_2 \), \( F_\alpha \) is again convex, with a single minimum on a discoid, which gets closer and closer to the round disc, represented by the point \((\frac{\pi}{2}, 1)\) in \( S \), as \( \alpha \) grows indefinitely (figure 9).

We have computed the three critical values, \( \alpha_1, \alpha_b, \) and \( \alpha_2 \) for several values of \( k_3 \). Figure 10 shows that, to within a good approximation, they all grow linearly with \( k_3 \). The best linear fit is provided by the following functions,

\[
\begin{align*}
\alpha_1 & \approx 157 + 15k_3, \\
\alpha_b & \approx 205 + 16k_3, \\
\alpha_2 & \approx 289 + 19k_3.
\end{align*}
\]

4. Comparison with experiments

A major motivation for this paper was offered by the experiments conducted in [16] with water solutions of disodium cromoglycate (DSCG). Here we apply our theory to interpret
Figure 7. For $\alpha < \alpha_1$, the reduced free energy $F_\alpha$ is convex on the configuration space $S$ and attains a single minimum in $0 < \phi < \frac{\pi}{16}$. Here, $k_3 = 1$, $\alpha = 170$, $\alpha_1 = 172$.

Figure 8. For $\alpha = \alpha_b$, the reduced free energy $F_\alpha$ has two (equal) global minima, one for $0 < \phi < \frac{\pi}{16}$ (tactoid) and the other for $\frac{\pi}{16} < \phi < \frac{\pi}{2}$ (discoid). Here, $k_3 = 1$ and $\alpha_b = 221$.

those experiments and show how to extract from them an estimate for the isotropic surface tension $\gamma$ at the interface between the nematic phase of a DSCG solution at a given concentration and its isotropic liquid phase.

To illustrate the generality of our method, we shall also apply it to estimate $\gamma$ at the interface between the tactoids formed in an F-actin solution at equilibrium with the isotropic phase [22].

4.1. DSCG solution

Among many other things, Kim et al [16] explored water solution of DSCG at concentration $c = 16\text{wt}\%$ confined between two parallel glass plates at a distance ranging in the interval 1–5 $\mu$m; the plates were spin-coated with a polystyrene layer, SE-7511, which exerts a degenerate tangential anchoring on the nematic director. This system can be treated as two-dimensional since the bounding plates suppress the out-of-plane distortions in the observed samples, as required by our theory.

Upon quenching the system from the isotropic phase into the coexistence regime, tactoidal droplets were observed, surrounded by the parent isotropic phase. In particular (see, figure 6(a) of [16]), a tactoid with bipolar nematic orientation was observed more closely, for which the area $A_0 = 200 \mu m^2$ and the aspect ratio $\delta = 1.3 \pm 1$ were measured at the temperature $T = 37.5^\circ\text{C}$, which we read off from figure 2(c) of [16]. We wish to make use of these data to validate our theory. To this end, we need to estimate the elastic constants $K_{11}$ and $K_{33}$ at $T = 37.5^\circ\text{C}$.

Interpolating the curves representing in [41] the temperature dependence of the elastic constants of the nematic phase of DSCG at $c = 16\text{wt}\%$, we readily arrived at

$$K_{11} = 4 \text{ pN}, \quad K_{33} = 9 \text{ pN},$$

at $T = 37.5^\circ\text{C}$ and $c = 16 \text{ wt}\%$, (20)
Figure 9. For $\alpha > \alpha_2$, the reduced free energy $F_\alpha$ is gain convex on the configuration space $S$ and attains a single minimum for $\frac{\pi}{16} < \phi < \frac{\pi}{2}$. Here, $k_3 = 1$, $\alpha = 307$, $\alpha_2 = 305$.

Figure 10. Critical values of $\alpha$ for different values of $k_3$. To within a good approximation, they all depend linearly on $k_3$. Red and blue ranges refer to discoids and tactoids, respectively, according to the same colour coding adopted in figure 6. The broken lines represent the best linear fits in (19).

and so we take $k_3 \approx 2.25$. We can also obtain $\alpha$ from the measured aspect ratio $\delta$. It follows from (15) that in our theory $\delta$ can be given the form

$$
\delta := \frac{\mu}{R(0)} = \frac{\mu^2 \left( \frac{3}{4} \cos \phi + \pi \sin \phi \right)}{\pi (\cos \phi + \sin \phi)},
$$

(21)

where use has also been made of (16). The plot of $\delta$ computed on the minima of $F_\alpha$ over $S$ is drawn against $\alpha$ in figure 11 for $k_3 = 2.25$.

It shows the typical behaviour also found in other theoretical studies [44, 45]: the aspect ratio of a bipolar droplet decreases as its linear size increases. We see that the value $\delta = 1.3$ is attained by the tactoidal branch (as expected) for $\alpha \approx 17.2$, the corresponding coordinates in $S$ of the minimum of $F_\alpha$ are $\phi \approx 0$ and $\mu \approx 1.24$, and so the equilibrium shape is a genuine tactoid (with pointed tips). For $k_3 = 2.25$, equilibrium tactoids cease to be genuine at $\alpha \approx 137$.

Since the equivalent radius $R_e$ is known from the isoperimetric constraint (7), $R_e \approx 8 \mu m$, we readily obtain for the long axis $l$ of the droplet $l = 2R_0 = 2\mu R_e \approx 20 \mu m$. The equilibrium tactoid predicted by our theory is plotted in figure 12 against the observed shape.

Our expected value for $l$ also agrees quite well with the measurement performed in [16] on the reconstructed shape, as does the cusp angle $\tau_N$ marked in figure 12. In our formalism, for a genuine tactoid, the latter can be expressed as

$$
\tau_N = -2 \arctan(R'(\mu)) = 2 \arctan \left( \frac{3\pi}{4\mu^2} \right),
$$

(22)

which for $\mu = 1.24$ delivers $\tau_N \approx 1.99$. The value of $\tau_N$ reported in [16] for the shape in figure 12 is $\tau_N = 1.05 \pm 0.05$, whereas a direct measurement on figure 12 would suggest that this value is indeed $\tau_N/2$, in good agreement with our theoretical prediction. Moreover, by use of equation (13), we
can give the following estimate for the surface tension at the nematic/isotropic interface of an aqueous DSCG solution at 16 wt%:

\[ \gamma \approx 8.9 \, \mu \text{N m}^{-1} . \] (23)

Different, discordant estimates have been given in the literature for the order of magnitude of \( \gamma \). For example, in [42] they estimate \( \gamma \sim 1 \, \mu \text{N m}^{-1} \), whereas in [16] they give \( \gamma \sim 10^2 \, \mu \text{N m}^{-1} \), an estimate obtained by applying the pendant drop technique [46, 47]. We trust that the measurements based on the theory presented in this paper might be more accurate. The value in (23) is closer to that found in [48] at the nematic-isotropic interface of 5CB (see also [49, 50]).

The critical values of \( \alpha \) corresponding to \( k_3 = 2.25 \) are \( \alpha_1 = 192 \), \( \alpha_2 = 330 \), and \( \alpha_0 = 243 \), while the observed droplet shown in figure 12 corresponds to \( \alpha = 17.2 \). According to our theory, one would then expect coexistence of tactoids and discoids for \( R_e \) in the range 90 \( \mu \text{m} \leq R_e \leq 153 \, \mu \text{m} \), that is, for an area \( A_0 \) in the range 0.3 mm\(^2\) \( \leq A_0 \leq 0.7 \, \text{mm}^2 \), a regime of large drops, for which no data are available in [16].

4.2. F-actin solution

Highly concentrated solutions of short (\( \lesssim 2 \, \mu \text{m} \)) F-actin filaments form liquid crystal phases which may coexist with their isotropic liquid phase [22].\(^\text{10}\) The two-dimensional islands of ordered phase are bipolar tactoids, just like those considered in this paper. Although it is suggestive to use liquid crystal theory to interpret the spindle shape exhibited in the process of cellular mitosis [52], ‘the existence of liquid crystal-like phases at physiological conditions\(^\text{11}\) is uncertain’, as appropriately pointed out in [53]. There, typical 3D tactoidal shapes were observed in solutions of F-actin filaments at equilibrium with the isotropic phase in the presence of physiological cross-linkings (filamin) at concentration 100-fold lower than in the case of free filaments. However, apart from being 3D, these tactoids contradict another hypothesis of our theory; they present a homogeneous alignment of the director. This makes them unsuitable for a comparison with our theory, which is instead a good fit for the experimental observations of 2D bipolar tactoids in [22].

To apply our theory to the F-actin solutions studied in [22], we need to estimate the elastic constants \( K_{11} \) and \( K_{33} \) at the appropriate concentration; unlike chromonics, direct experimental determinations are lacking for them. Here we shall resort to rigid-rod theory. For filament lengths less than 2 \( \mu \text{m} \), as in this case, good agreement with rigid-rod theory is found in [51], and as highlighted in [53], short actin filaments (average length between 180 nm and 2 \( \mu \text{m} \)) behave as rigid rods because they are appreciably shorter than the persistence length of F-actin polymers, which is approximately 15–18 \( \mu \text{m} \) [54, 55].

The bare excluded volume theory, which treats filaments as rigid cylinders of diameter \( d \) and height \( \ell \), predicts the following expressions for the elastic constants [56],

\[ K_{11} = \frac{7}{8\pi} \frac{kT}{d^3} q, \quad K_{33} = \frac{4}{3\pi^2} \frac{kT}{d^3} q, \] (24)

where \( k \) is the Boltzmann constant, \( T \) is the absolute temperature and \( q \) is the dimensionless concentration, defined as

\[ q := b c_\ell = \frac{\ell}{\eta d}. \] (25)

where \( c_\ell \) and \( \eta \) are the filaments’ number density and volume fraction, respectively, and \( b := \pi \ell^2 d/4 \) is the average isotropic excluded volume.

F-actin filaments are charged; the extra mutual repulsion that they feel affects the elastic constants. As already observed by Onsager [15], this effects can be taken into account in a purely steric theory by replacing \( d \) with an effective diameter \( d_e > d \). Thus, in (24) \( d \) needs to be replaced by \( d_e \) and \( q \) by \( q_e := q b_e/b \), where \( b_e := b d_e/d \) is the effective average excluded volume. It is easily seen that these corrections leave

\(^{10}\) For longer filaments, the ordering transition is continuous and no coexistence has been observed [51].

\(^{11}\) Meaning, in living cells.
unchanged, while increasing $K_{33}$:

$$K_{11} = \frac{7}{8\pi} \frac{KT}{d} \left( \frac{\eta}{\rho} \right), \quad K_{33} = \frac{32}{21\pi} \frac{K_{11}q_e^2}{\eta\ell}$$

with $q_e = \frac{\ell d_e}{\eta d}$,

so that

$$k_3 = \frac{K_{33}}{K_{11}} = \frac{32}{21\pi} \frac{q_e^2}{\eta\ell}$$

As first pointed out in [57], the electrostatic repulsion among filaments also affects the critical concentration $q^*$ at which the isotropic phase becomes unstable (which is a good estimate of the coexistence region), in a way that depends on the ratio $\kappa$ of the Debye screening length to the effective diameter. Thus, from $q^* \approx 4$ for $\kappa = 0$ [58], $q^*$ may increase up to $q^* \approx 6$ for $\kappa = 0.5$ [57]. Taking as in [59] $q_e \approx 6$ in (27), we estimate $k_3 \approx 17.5$ for F-actin filaments.

To obtain $K_{11}$, we need to estimate $\eta$, $\ell$, and $d$. From [60], we get $d \approx 8$ nm. As for $\eta$ and $\ell$, we rely on the data of [22]: they estimate $\ell \approx 1.2 \mu$m and produce solutions with a mass density $\rho \approx 1.08 \text{ mg m}^{-1} = 10.8 \text{ Kg m}^{-3}$. Since the molecular mass of a monomer of actin is $M = 4.2 \text{ Kg mol}^{-1}$ and its volume is $v_m = 50.1 \text{ nm}^3$ [60], the molar concentration of the solution is $c_M = \rho M \approx 259 \times 10^{-3} \text{ mol m}^{-3}$, which gives $\eta = c_M N_A v_m \approx 7.8 \times 10^{-3}$ (where $N_A \approx 6.02 \times 10^{23}$ is the Avogadro number) and, by (26), $d_0/d \approx 5$. Thus, again by (26), we obtain that at room temperature $K_{11} \approx 0.17 \text{ pN}$ and $K_{33} \approx 2.93 \text{ pN}$.

With these data we are now in a position to interpret within our theory the observations of [22]. Their figure 7 collects a number of tactoids by their aspect ratio $\delta$ and axial length $l$. We select as a representative observation one around which most data seem to be clustered, namely $d = 2.7$ and $l = 50 \mu$m. Following the same steps as in section 4.1, for the estimated $k_3$ we find that $\alpha \approx 3.9$. To extract finally $\gamma$ from this, we only need to estimate the equivalent radius $R_e$ of the representative tactoid. Taking this as the region bounded by two symmetric arcs of circle, equation (C2) from appendix C delivers $A_0 \approx 634 \text{ m}^2$ for its area. Thus, $R_e \approx 14.2 \mu$m and

$$\gamma_{\text{actin}} = \frac{\alpha K_{11}}{R_e} \approx 46 \text{ nN m}^{-1}$$

which is an order of magnitude less than the value ($\gamma_{\text{actin}} \approx 300 \text{ nN m}^{-1}$) estimated experimentally in [53] with a dynamical relaxation method for tactoids formed at a concentration 100-fold lower (in the presence of a cross-linking physiological ligand), but consistent with the measurements at the interfaces of some phase-separated colloid-polymer dispersions [23]. For a further comparison, we note that our estimate for $\gamma_{\text{actin}}$ is appreciably less than the surface tension reported for other protein-based liquid droplets, which ranges from $\gamma \sim 1 \mu\text{N m}^{-1}$ [61] to $\gamma \approx 100 \mu\text{N m}^{-1}$ [62].

As for the regime of coexistence between tactoids and discoids as equilibrium shapes, which is the main object of this paper, for $k_3 = 17.5$ equation (19) delivers $\alpha_1 \approx 420$ and $\alpha_2 \approx 622$ as delimiters of the coexistence window (with perfect bistability at $\alpha_3 \approx 485$). They correspond to the following values of the equivalent radius, $R_e \approx 1528 \mu$m and $R_e \approx 1768 \mu$m, respectively, more than two orders of magnitude larger than for the observed tactoids. According to our theory, this makes it highly unlikely the observation of equilibrium shapes of condensates of F-actin filaments other than tactoids, at least at the given concentration.

5. Conclusion

We studied chromonic droplets in two space dimensions. Our major motivation was the thorough experimental investigation in [16] on thin cells with degenerate tangential anchoring for the nematic director. In particular, a solution of DSCG in water was quenched in the temperature regime where nematic and isotropic phases coexist in equilibrium, the former forming islands with a tactoidal shape.

We introduced a wide class of two-dimensional shapes for the equilibrium droplets, which includes tactoids, discoids, båtonnet, and butterflies, the latter of which are concave, whereas the former three are convex. Our analysis revealed that upon increasing the droplet’s area a tactoidal equilibrium branch gives way to a discoid branch, while neither båtonnet nor butterflies can ever be equilibrium shapes. Moreover, there is a regime of shape coexistence, where a tactoid and a discoid are both local minima of the free energy, the global minimum shifting from one to the other at a critical value of the droplet’s area, where perfect bistability is established. This shows another, possibly less usual incarnation of a phenomenon broadly present in physics (think of optical bistability, for example). A typical bifurcation diagram with hysteresis describes the situation, reminiscent of a first-order phase transition with super-heating and super-cooling temperatures (replaced here by corresponding values of the area).

We put our theory to the test by interpreting experimental data provided by [16]. We found a fairly good quantitative agreement between experiment and theory, although further data should be collected to establish on firmer grounds the degree of confidence of the theory. In particular, we could extract from the data available in [16] an estimate for the isotropic component $\gamma$ of the surface tension at the interface between coexisting nematic and isotropic phases of DSCG in 16%wt aqueous sol. This estimate seems to promise more accuracy than the rough evaluation of order of magnitude available in the literature for this material and its chromonic siblings. We hope that the theory proposed here could be used for a systematic determination of $\gamma$ for different temperatures and concentrations.

To illustrate the generality of our method, we also used it to estimate the isotropic surface tension $\gamma_{\text{actin}}$ for tactoids shown in solutions of F-actin filaments [22]. We estimated the elastic constants for the nematic phase of F-actin filaments taking them as charged, rigid cylinders; we obtained a value of $\gamma_{\text{actin}}$ comparable to the one measured for some phase-separated colloid-polymer dispersions [23].

The data available for both systems do not cover the range of predicted bistability. We estimated the area that a droplet
should reach to display an abrupt transition from tactoid to discl.
It remains to be seen whether a controlled growth in the
droplet’s size can be realized to observe neatly this transition.

We have shown that the critical values of the area that
delimit the shape hysteresis are (increasing) linear functions of the ratio
$K_{33}/K_{11}$ between bend and splay elastic constants.
If this critical phenomenon could be explored experi-
mentally, our theory would also offer an independent way to
measure $K_{33}/K_{11}$. This might be especially welcome for non
conventional new lyotropic phases.

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broader contribution.

Data availability statement

All data that support the findings of this study are included
in the article (and any supplementary files).

Appendix A. Retracted tangential field

In this appendix, we justify the expression for the free-energy
functional (11) associated with the bipolar director field $n$
defined as the unit vector field tangent to the lines with given
t and varying $y$ (see figure 2). A generic curve in that family is
represented by the position vector

$$p(t) := g(t)R(y)e_x + ye_y - R_0 \leq y \leq R_0,$$  \hspace{1cm} (A1)

where $g$ is any function of class $C^1$ strictly increasing on $[0, 1]$ and
such that $g(0) = 0$ and $g(1) = 1$. For given $t \in [0, 1]$, the
tangent vector field $n$ is given by

$$n = \frac{gR'e_x + e_y}{\sqrt{1 + (gR')^2}},$$  \hspace{1cm} (A2)

where a prime denotes differentiation. The unit vector field

$$n_\perp := e_z \times n = \frac{-e_y + gR'e_x}{\sqrt{1 + (gR')^2}},$$  \hspace{1cm} (A3)

is everywhere orthogonal to $n$ and such that $n_\perp|_{e = \perp} = -\nu$,
where $\nu$ is the outer unit normal to $\partial R$ (see again figure 2).

Imagine now a smooth curve $p$ in $R$ parameterized as $\xi \mapsto (t(\xi), y(\xi))$; it follows from (A1) and (A2) that

$$\dot{p} = g'R'e_x + \dot{y}(gR'e_x + e_y) = g'R'e_x + \sqrt{1 + (gR')^2} \dot{n},$$  \hspace{1cm} (A4)

where a superimposed dot denotes specifically differentiation
with respect to $\xi$. Thus, the elementary area $dA$ is

$$dA = dt \ dy g'R\sqrt{1 + (gR')^2} e_x \times n \cdot e_z = g'R \ dt \ dy,$$  \hspace{1cm} (A5)

and the elementary length $dl$ on the retracted curve $\partial R$, for
given $t$, is

$$dl = \sqrt{1 + (gR')^2} \ dy.$$  \hspace{1cm} (A6)

In particular, the area of $R$ and the length of its boundary are
given by

$$A(R) = 2 \int_0^1 g' \ dt \int_{-R_0}^{R_0} R \ dy = 2 \int_{-R_0}^{R_0} R \ dy$$  \hspace{1cm} (A7)

and

$$\ell(\partial R) = 2 \int_{-R_0}^{R_0} \sqrt{1 + R'^2} \ dy.$$  \hspace{1cm} (A8)

Differentiating $n$ in (A2) along the smooth curve $\xi \mapsto (t(\xi), y(\xi))$, we find that

$$\dot{n} = \frac{g'R'i + gR'\dot{y}}{1 + (gR')^2} n_\perp.$$  \hspace{1cm} (A9)

Assuming that $n$ is differentiable in $R$, $\dot{n}$ and $\dot{\nu}$ must be related
through

$$\dot{n} = (\nabla n) \dot{\nu}.$$  \hspace{1cm} (A10)

Since $n$ is a unit vector field, $(\nabla n)^T$ annihilates $n$, and so there exists a vector $a = a_n n + a_\perp n_\perp$ such that

$$\nabla n = n_\perp \otimes a.$$  \hspace{1cm} (A11)

To determine the scalar components $a_i$ of $a$, we observe that, by (A11), equation (A10) also reads as

$$\dot{n} = (a \cdot \dot{\nu}) n_\perp.$$  \hspace{1cm} (A12)

Making use of (A4) and both (A2) and (A3), we readily see that

$$a_1 = \frac{gR''}{[1 + (gR')^2]^{3/2}},$$  \hspace{1cm} (A13)

and inserting this into (A12) alongside with (A9), we obtain
an identity for arbitrary $(i, \dot{y})$ only if

$$a_1 = \frac{gR''}{[1 + (gR')^2]^{3/2}},$$  \hspace{1cm} (A14)

which leads us to

$$\div n = -\frac{gR''}{[1 + (gR')^2]^{3/2}} n_\perp \otimes n$$

$$\div \nabla n = \left( \frac{R'}{R} \sqrt{1 + (gR')^2} - \frac{g^2 R'^2}{[1 + (gR')^2]^{3/2}} \right) n_{\perp \otimes n_{\perp}}.$$  \hspace{1cm} (A15)

The following expressions for the traditional measures of
distortion then follow from (A15),

$$\div n = -\frac{gR''}{[1 + (gR')^2]^{3/2}} e_z,$$  \hspace{1cm} (A16a)

$$\curl n = -\frac{gR''}{[1 + (gR')^2]^{3/2}} e_z.$$  \hspace{1cm} (A16b)
The prescribed boundary conditions lead to a specific functional \( F \) which we arrive at the following reduced functional,

\[
F[\mu; R] : = \frac{\mathcal{F}[\mathcal{B}]}{K_{11} h} \\
= \int_\mu^\rho dy \int_0^1 \frac{g^4RR'^2R''^2}{(1+(gR')^2)^3} + \frac{R^2}{(1+(gR')^2)} + \frac{1}{(1+(gR')^2)} \\
- 2 \frac{g^2R'^2R''^2}{(1+(gR')^2)^2} + k_3 - \frac{g^2RR''^2}{(1+(gR')^2)^2} \right] dt \\
+ 2\alpha \int_\mu^\rho \sqrt{1+R^2} \, dy, \tag{A16e}
\]

where \( k_3 \) and \( \alpha \) are as in (12) and in (13). The integration in \( t \), which delivers (11) in the main text, is independent of the specific function \( g \), provided it is monotonic and obeys the prescribed boundary conditions.

**Appendix B. Preventing anchoring breaking**

Here, we perform an energy comparison to identify the safeguard value \( \alpha_s \) of \( \alpha \), that is, the lower bound that should be exceeded for a drop to be bipolar at equilibrium. It is known that for \( \alpha \) sufficiently small the tangential anchoring favoured by the interfacial energy (4) is bound to be broken, so that the nematic alignment becomes uniform throughout the droplet. Uniform and bipolar alignments will indeed be the terms of comparison for our estimate of \( \alpha_s \).

For drops with uniform alignment, the total free energy reduces to (4), subject to the area constraint (10). The optimal shape is delivered by the classical Wulff’s construction [63] (see also [21, p 490]). Assuming that \( n = e_\gamma \) and that \( \mathcal{B} \) is mirror-symmetric with respect to both axes \( e_x \) and \( e_y \), one needs only determine the equilibrium shape of \( \mathcal{B} \) in the positive \((x, y)\) quadrant. We let \( y = y_w(x) \geq 0 \) represent the profile of \( \mathcal{B} \) and define \( \lambda > 0 \) by setting \( y_w(\lambda) = 0 \). Here, as in the main text, all lengths are scaled to \( R_0 \). The method illustrated in [24, chapter 5] and [44] delivers Wulff’s shape through the following explicit function

\[
y_w(x) = \frac{\lambda}{v} = \frac{1 + \omega(1 - \xi^2) - \xi x \lambda^{-1}}{\sqrt{1 - \xi^2}}, \tag{B1a}
\]

where \( \xi \) is given in terms of \( x \) by solving the algebraic equation

\[
\omega \xi^3 + (1 - \omega) \xi - x \lambda^{-1} = 0 \tag{B1b}
\]

and \( \lambda \) is determined by the isoperimetric constraint (10), which here reads as

\[
4 \int_0^\lambda y_w(x) \, dx = 4 \lambda^2 \int_0^1 y_w(x \lambda^{-1}) \lambda^{-1} \, d(x \lambda^{-1}) = \pi. \tag{B1c}
\]

Correspondingly, by computing the free energy in (4) on Wulff’s shape, we obtain that

\[
\mathcal{F}_w = 4K_{11} h \alpha j(\omega), \tag{B2}
\]

where the function \( j \), defined by

\[
j(\omega) := \frac{1}{2} \sqrt{\frac{\pi}{\int_0^1 \sqrt{1 + y_w^2} \, dx}} \\
+ \omega \int_0^1 \frac{1}{\sqrt{1 + y_w^2}} \, dx, \tag{B3}
\]

is computed on the solution \( y_w \) of (B1), renormalized so that \( y_w(1) = 0 \). Figure 13 illustrates the Wulffian shape obtained with this method for \( \omega = 5 \) and the graph of \( j \) for \( 0 \leq \omega \leq 10 \).

We now compare this energy to that of a disc with an in-plane bipolar director field whose integral lines are Apollonian circles passing through the poles (see, for example, section 2 of [64]); their radius increases to \(+\infty\) upon approaching the \( y \)-axis, as shown in figure 14, which represents a quadrant of the disc. Adapting the computations in bipolar coordinates \((\xi, \eta)\) of Williams [65] to the present two-dimensional setting, we arrive at

\[
\mathcal{F}_{\text{bip}} := K_{11} h \left\{ 4 \int_0^{\frac{\pi}{2}} d\xi \int_0^{\frac{\pi}{2}} \frac{1}{\sin \xi} \left[ \frac{1}{\tan \xi} - \frac{1}{\sin \xi} \right] \right. \\
+ \frac{k_3}{2} \frac{1}{(1 + \sin \xi \cos \eta)^2} \right\} \right] + 2\alpha \pi \right) \\
= 4hK_{11} \left\{ \frac{\pi}{4} (k_1 - k_3 \ln 2 - \ln \varepsilon) + \varepsilon + \alpha \left( \frac{\pi}{2} - \varepsilon \right) \right\} \frac{\alpha \omega \xi^3}{2}, \tag{B4}
\]

where the defect core has been identified with the region \( 0 \leq \xi \leq \varepsilon \). The estimate in (18) follows from requiring that \( \mathcal{F}_{\text{bip}} < \mathcal{F}_w \).

**Appendix C. Spindle area**

In the bipolar coordinates \((\xi, \eta)\) adopted by Williams [65], a *spindle* is the region between two Apollonian circles symmetrically located relative to the axis joining the poles (figure 14 shows quarters of the members in a single family of such Apollonian circles). Half a spindle is described by letting \( \xi \) range in \([0, \pi]\) and \( \eta \) in \([0, \eta_0]\), where \( \eta_0 \) is related to the aspect ratio \( \delta > 1 \) by \( \eta_0 = 2 \arctan(1/\delta) \). Denoting by \( l \) the distance between the poles, the area element is given by
Figure 13. Shape and (scaled) energy for the fully aligned droplet.

\[ \text{Area of a spindle} = \frac{1}{2} \int_0^{\pi/2} \int_0^{\pi/2} \sin \xi (1 + \sin \xi \cos \eta)^2 \, d\xi \, d\eta. \]  
\[ \text{(C1)} \]

The area of a spindle will then be expressed as

\[ A_0 = \frac{1}{2} \left( \frac{1}{2} \right)^2 \int_0^{\pi/2} \int_0^{\pi/2} \frac{\sin \xi}{(1 + \sin \xi \cos \eta)^2} \, d\xi \, d\eta \]

\[ = \left( \frac{1}{2} \right)^2 \frac{(1 + \delta^{-2})^2 \arctan \delta + \delta^{-3} - \delta^{-1}}{\delta^{-2}}. \]  
\[ \text{(C2)} \]

which is the formula used in section 4.2 to estimate the area of a tactoid representing a cluster of observations in [22].

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