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Geometry and Thermodynamics of Filament Bundles

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GEOMETRY AND THERMODYNAMICS OF FILAMENT BUNDLES

A Dissertation Presented

by

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ABSTRACT

GEOMETRY AND THERMODYNAMICS OF FILAMENT BUNDLES

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In this dissertation I present a study of the geometry and energetics of bundles composed of flexible cohesive filaments. This is a general class of materials, both biological and artificial, existing across many length scales. The aim of this thesis is to investigate the interdependence between the 2D organization of filaments in a bundle’s cross section, and the 3D structure, with an emphasis on the twisting mode of deformation. First we present a model of filament contacts and interactions, which we employ in numerical simulations to study the connection between the ground state energies of constant-pitch bundles and their interior packing topology. We then focus on exterior features, and construct a continuum model of the surface energy and its twist-dependence. Finally, we employ a fully 3D model of filament bundles with a fixed packing topology to examine the connection between filament organization and the resulting 3D structures, be they twisting, writhing, bending, undulating, or other modes of deformation.
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CHAPTER 1
MOTIVATION

1.1 Introduction to columnar materials

Cohesive assemblies of filaments are a common structural motif found in diverse contexts, ranging from biological materials such as collagen [1, 2, 3], fibrin [4, 5], sickle hemoglobin macrofibers [6, 7], and condensed DNA [8, 9, 10]; to artificial materials such as carbon nanotube ropes [11, 12, 13, 14], mesoporous silica [15, 16, 17], and micropatterned filament arrays [18]. The forces involved in the cohesion of these columnar materials include Van der Waals interaction, electrostatics, depletion forces, and capillary interactions. Throughout this dissertation we attempt to answer the following question with a variety of approaches: What is the most energetically favorable way to organize a collection of long flexible cohesive filament, independent of the specific materials or interactions? A trivial answer would be to packing straight filaments straight into a hexagonal lattice; this could potentially maximize the contact between filaments and yield a very dense packing. However, under certain conditions this is not always the case, such as when the filaments possess chirality.

An object is said to be chiral if it cannot be superimposed onto its mirror image [19]. A simple example is a screw, which has a right-handed helical thread. Biological filaments (such as proteins and DNA) are generally chiral in shape, and commonly express their chirality through interactions that promote skewed orientations, a feature that leads to twisted assemblies with self-limiting sizes [20, 21, 22, 23]. Finding the most efficient way of organizing any collection of objects in three dimensions is already a difficult task, with chirality and twist only complicating the matter [24].
However, the goal is still set, as self-assembled helical structures are an emerging class of new and exciting functional materials [25].

The problem of packing chiral columnar materials is a commonly found in liquid crystals. Liquid crystals are a form of matter where the molecules are organized in such a way as to possess both liquid and crystalline-like properties, depending on the orientations and length scales being observed [26]. The average orientation of the molecules is characterized by a nematic director, which allows for a numerical description of the crystalline nature of the material. Liquid crystals composed of chiral molecules are able to organize into phases that incorporate their chirality [27]. They have been studied a great deal, revealing a multitude of possible phases with various degrees of symmetry [26, 28]. Two common examples are the chiral nematic (a.k.a. cholesteric) phase [29], and the double-twisted blue phase [30, 31]. The organization of molecules within the chiral nematic phase incorporates the chirality by skewing (i.e. twisting) along one direction perpendicular to the nematic director, while the blue phase is a more complex structure that contains regions where the molecules twist along both orthogonal directions perpendicular to the nematic director; thus it is said to be “double-twisted”. Both of these structures are shown in Fig. 1.1. Double stranded DNA is a well-studied molecules that, under the correct conditions, produces many of these textures when in condensed liquid crystalline form [33], as well as other alternative configurations, such as toroidal condensates [34]. Using liquid crystals as an analogy, we may suppose that densely packed flexible filaments may also organize into similar twist-incorporating structures, especially if they are chiral. A trivial guess would be to pack them parallel into a bundle, and then twist them into a rope- or cable-like structure [20].
Figure 1.1. Schematic representations and experimental images of the (a) nematic, (b) cholesteric, and (c) blue phases of liquid crystals. Bold black arrows show the direction along which the molecular orientation twists. Experimental images are from [32]

Unusually enough, it has been shown that intrinsic chirality is in fact not a requirement for realizing symmetry-breaking textures in a large variety of materials.¹ To start, liquid crystals composed of achiral molecules are able to achieve chiral twisted phases under strong confinement, such as within a cylinder [35] or tactoid spindle [36], as a method to reduce splay. Additionally, bent-core (a.k.a. banana) molecules can readily form twisted nematic phases [37]. Plate-like twisted structures are possible too, as demonstrated with both chiral and achiral rod-like viruses forming monolayered membranes with edges that twist as a means to reduce the surface energy [38, 39]. Moving to the microscale, patterned arrays of plastic fibers aggregated by capillary forces of water are able to form twisted bundle-like structures (interestingly,

¹Though with no chirality of the molecule to select a handedness of the phase, it will be equally likely to be both
in a hierarchical manner), as a result of the competition between the capillary forces and intra-filament elasticity [18, 40, 41]. Not even continuous filaments are required, as simulations have shown achiral disc-like particles stacking into segmented fibers and then self-assembling into structures resembling twisted bundles [42, 43]. Another example is mesoporous silica, which is able to form hexagonally-packed arrays of pores that spontaneously break symmetry and twist, despite being produced from only achiral amphiphiles [44, 45, 46, 17]. This ability has been attributed to a number of possible effects, such as twist’s role in reducing the surface energy [47], staggered amphiphile packings [44], or other entropic arguments [48]). Lastly, DNA, a well-studied chiral molecule, form twisted toroidal configurations when in a condensed phase [34]; however recent studies have shown that chirality is not a requirement for twist; instead, the driving force is a reduction of bending energy [49]. And DNA inside virus capsids equivalently form twisted packings when under strong confinement, even when modeled excluding chirality [50, 51, 9, 52, 53, 54]. In light of this large body of work, we similarly model twisted filament bundles without explicitly considering intrinsic chirality. And in chapter 3, we propose that bundles composed of highly flexible filaments can in fact lower their energy by twisting due to a reduction in surface energy.

1.2 Geometric frustration and twist

In order to fully describe filament bundles, we must first overview some key geometric principles underlying our study. To start, twisted filament bundles fall into the category of geometrically frustrated materials. Geometric frustration, in general, is a phenomenon where local interactions promote a certain state that cannot be repeated within the material due to the global geometry [55]. Some of the earliest examples of geometric frustration were found in magnets. Even a simple 2D spin glass model captures the effect of geometric frustration for a triangular lattice of spin sites with
antiferromagnetic bonds. In this example, a triangular section of the lattice consisting of three spin sites will have one site spin up, one site spin down, and a third site with equal propensity to be up or down, hence it is frustrated [56]. Globally, this results in a large amount of degenerate states for the system. The same effect has been shown in buckled monolayers of hexagonally pack colloids, where individual particles have a tendency to be displaced up or down, oppositely of their neighbors, thus leading to patterns much like the frustrated magnets [57]. Similarly, geometric frustration in materials with non-fixed lattices occurs when molecules are subject to conflicting forces, and relieve themselves by aligning into non-trivial positions and complex arrangements [55]. Here the conflicting forces are generally between intermolecular interactions that attempt to enforce constant spacing between particles, and the particular geometric details involved in packing the particles. In this case, the geometry effectively disrupt the ability to achieve constant particle spacing everywhere. Common examples are small 3D colloidal clusters [58], large binary hard-sphere mixtures [59], and 2D packings of particles on curved surfaces [60]. In the latter case, curvature functions to disrupts the ability to retain constant spacing between particles, necessitating defects in the crystalline texture. Analogously, throughout this dissertation we will show that in in columnar materials, variable tilt is a form of geometric frustration that disallows ordered filament packing.

The non-linear influence of twist is quickly apparent with small numbers of filaments. Twisting just two fibers together (known as a 2-ply) will wind them around each other, each forming a helix with a radius equal to the filament diameter, $d$. However, this only happens up until the point where the pitch, $P = \pi d$. In order to further decrease the pitch beyond this point, the helical radius of the filaments must increase and a gap will open in the center of the 2-ply. Similarly formed $n$-plys of arbitrary numbers of fibers undergo the same transition, but at different critical values of the pitch [61, 62, 63]. A simple measure of this twist-induced limitation,
but with an eye towards packing larger numbers of filaments, is the *kissing number* [64, 65], \( Z \), of a central filament in a twisted bundle, like the one shown in Fig. 1.2. We define \( Z \) as the maximum number of non-overlapping filaments allowed to simultaneously contact the central filament. The helical shape of the finite-diameter filaments surrounding the central filament implies that contacting neighbors occupy greater than \( 2\pi/6 \) of the planar angle surrounding the central (straight) filament.

A similar phenomenon occurs when packing discs on curved 2D surfaces. Gaussian curvature effectively alters the metric spacing between discs, upsetting the hexagonal packing that is allowed on a flat surface [66]. In this case, \( Z < 6 \) for positively curved surfaces. The connection is more than just qualitative, as a full mapping between twist and Gaussian curvature is fully explained in chapter 2.

Before we proceed, let us quickly address the mechanical benefits of twist [67], a fact that has been known to humans for at least tens of thousands of years, in our making of ropes [68, 69]. Crucial for yarns spun from fibers of a finite length, twist

---

**Figure 1.2.** Kissing number, \( Z \), for the central (blue) filament, twist angle \( \theta \). Examples show the structures with the largest \( \theta \) for a given \( Z \). The left figure shows a longer bundle with two cutting planes used to produce the short segment (in this case, \( Z = 5 \)) shown within the plot.
transforms tension along the yarn’s length into compression of the fibers, locking them together via friction [70]. Second, twist relaxes the variable stretching/compression from bending the cable, conferring superior flexibility [71]. Given these advantages, it is perhaps unsurprising that Nature incorporates this design into many of its biological structures. For instance, helically wound cellulose microfibrils provide mechanical reinforcement for walls of wood cells [72], while helically twisted fibrils of extracellular protein filaments, like collagen [3], and fibrin [5], play crucial mechanical roles in animal tissue. The spontaneous twist of these biofilaments must be a consequence of their self-assembly, and in this case the molecular scale chirality of biomolecules is broadly implicated as the driving force for twisted filament packings [73, 6, 4, 74, 75]. However, throughout this dissertation we will discuss the geometric and energetic effects of twist, regardless of any intrinsic chirality. Our purpose is to isolate the structure and energy of filament bundles from a dependence on twist.

1.3 Topological defects in crystalline materials

A common reference state for columnar materials, is an unperturbed crystalline lattice of straight filaments. Because all planes perpendicular to the filaments are identical up to a translation along the the length direction, we are effectively left with a 2D crystalline lattice, with lattice sites located at the filament-plane intersections. The densest packing of a single particle in two dimensions is to surround it with six neighboring particles, where each neighbor occupies $2\pi/6$ of the planar angle, as seen when $\theta = 0$ and the kissing number, $Z = 6$, in Fig. 1.2. Coincidentally, this sort of arrangement is repeatable in all directions, forming a hexagonal lattice [24]. However, if twist is present, $Z < 6$, and the perfect six-fold hexagonal close-packing is no longer possible.

It is possible to have an imperfect hexagonal structure, where the majority of the material retains it’s crystalline nature, and is only occasionally interrupted by
defects. The two types of topological defects most important in this study are dislocations and disclinations. Dislocations are formed by adding or removing a crystal plane of atoms from part of the material.\(^2\) Alternatively, disclinations are formed by adding or removing an entire wedge of the crystalline material [79]. Both are shown for a hexagonal lattice in (Fig. 1.3(b)). On the other hand, disclinations are characterized by a topological charge, \(s\), corresponding to the quantity of angle that was added/removed. In a hexagonal lattice, \(s\) is confined to discrete values compatible with the six-fold nature of the crystal, yielding \(s = 2\pi n/6\), where \(n\) is a positive or negative integer. The two most common type of disclinations in a hexagonal lattice are, 5-fold where \(s = +\pi/3\), and 7-fold where \(s = -\pi/3\). In this case, the disclination atom itself does not have the usual six nearest neighbors.

\(^2\)formally known as the Volterra construction [76, 77]
An intriguing characteristic of dislocations is that they are composed of two disclinations of opposite charge (i.e. value of \( s \)). Historically, this feature was important in the discovery of the hexatic phase, by Halperin and Nelson [80]. According to the scenario predicted in ref. [81], at non-zero temperatures a 2D crystal will incorporate dislocations naturally, preserving the long-range orientational order while disrupting the long-range positional order. Upon further heating, the dislocations unbind into individual disclinations, additionally disrupting the long-range orientational order. Still further heating will fully destroy the lattice, and yield a liquid with only short-range positional and orientational order.

The existence of single disclinations in real materials are rare, due to the long-range stresses they impose; however, they can be found in the ground state configurations of sufficiently geometrically frustrated systems. In chapter 2, we will show that twisted filament bundles fall into this class of materials, which also includes curved crystalline membranes [82]. Gaussian curvature, defined as the product of the two principle directions of curvature, \( K_G = k_1 k_2 \), produces a non-Euclidean geometry (shown in Fig. 1.4(a)). A flat membrane can only achieve Gaussian curvature by stretching, as famously implied in Gauss’ *Theorem Egregium* [86]. However, this stretching effectively disrupts the hexagonal order of the crystal, necessitating topological defects in the ground state structures. This phenomenon has been observed in a number of systems, such as: colloidal crystals on curved oil-glycerol interfaces, shown in Fig. 1.4(b) [83]; Virus capsids, where identical subunit capsomer proteins must form defects in order to achieve a fully enclosed shell, shown in Fig. 1.4(c) [87]; And the large class of materials belonging to the generalized Thomson problem, where particles packed on a spherical surface require at minimum twelve 5-fold disclinations, shown in Fig. 1.4(e) [88, 89, 85, 82]. The two biggest conclusions from these studies are that: first, the net number of 5-fold disclinations is largely dependent only on the integrated Gaussian curvature.
Figure 1.4. (a) Positive and negatively curved surfaces with the principle curvatures shown as black lines. (b) A colloidal crystal with defects on an oil-glycerol interface [83]. (c) Cowpea mosaic virus capsid composed of 60 subunit capsomer proteins [82]. (d) A egg-carton shaped membrane with disclinations located at points of maximum curvature [84]. (d) A solution to the Thomson problem with 732 particles [85].

curvature of the surface\(^3\); and second, the total number of defects increases with the number of particles for a constant integrated Gaussian curvature, mainly through the addition of dislocations [82].

Conveniently, any theories about elasticity and defects within 2D crystals can easily be converted and employed towards columnar materials by taking any extrinsic quantity and multiplying it by the length of the filaments in the \( z \) direction. While the crystal structure itself is generated by considering the intersections of filaments with a perpendicular \( z \) plane. Additionally, in chapter 2.5, we will introduce a new geometry-based conversion between surface curvature and bundle twist, that will conveniently allow us map our bundle problem onto the already well established membrane one. We have already confirmed that the densest packing of straight filaments is a hexagonal lattice, shown in Fig. 1.5(a). But when the filaments within

\(^3\)The "net number" of 5-fold disclinations is defined as the number of 5-fold minus the number of 7-fold disclinations, and is commonly referred to as the topological charge.
Figure 1.5. (a) A section of a bundle of straight filaments, exhibiting hexagonal packing. Twisting this structure yields (b), where yellow regions highlight overlap between filaments. (c) A bundle of straight filaments with a single centered 5-fold disclination shown in red, and open gaps between filaments highlighted in purple. Twisting this structure yields (d), where the azimuthal compression from twist is conveniently (though only partially) screened by the disclination.

this structures tilt non-uniformly with respect to each other, as they do in a twisted bundle, it becomes difficult to retain the original crystalline hexagonal pattern (Fig. 1.5(b)). Twist effectively imposes a compressive stress in the azimuthal direction, leading to an overlapping of nearby filaments, which can be seen as a form of geometric frustration.

As we now might expect, introducing defects into the crystalline packing can remedy the twist-induced stresses. Concisely put, whereas twist azimuthally compresses the filaments, the inclusion of a 5-fold disclination azimuthally decompresses them (Fig. 1.5(c)); therefore one might expect, from this purely geometrical argument, that the two may exist together in cohesive filament bundles (Fig. 1.5(d)). This implies that in sufficiently large and twisted bundles, an excess of these 5-fold disclinations are necessary components of the minimal-energy lattice packing [90, 91]. Hence, a quantitative analysis of the cohesive energy (performed in section 2.6) requires careful accounting for the number and organization of the lattice defects (performed in section 2.4).
1.4 Continuum elasticity of columnar materials

Here we will present some insight into the expected results of twisted filament bundles based on continuum elasticity arguments. This is necessary to understand some of the predictions and phenomena already known to exist. We will simplify the derivations originally found in references [90, 91], and additionally explore the stress-response of columnar materials. In this calculation we derive the equations of bundle stresses in the limit of infinitely small filaments.

We begin with the assumption that our material shears freely in the direction along the filaments axes (the $z$ axis in the reference state), and we ignore any complications arising at filament ends. The general energy density for a 2D plane of such material governed by continuum elasticity is

$$
E_S = \frac{1}{2} \int \lambda u_{kk}^2 + 2\mu u_{ij}u_{ij} dV,
$$

where $\lambda$ and $\mu$ are the Lamé constants, $u_{ij}$ is the strain tensor, and the subscripts $i, j, k$, each denote the two principle directions in the 2D cross-sectional plane, $\hat{x}, \hat{y}$. We employ a nonlinear form of the strain that retains the geometrical coupling between out-of-plane tilt and in-plane strain

$$
u_{ij} \simeq \frac{1}{2} \left( \frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i} - t_i t_j \right),
$$

where $u_i$ is the displacement, and $t_i \simeq \partial u_i/\partial z$ is the filament tangent. The cost of bending filaments is dependent on the curvature of the backbone, and following the standards used in liquid crystals [77], is designated as

$$
E_B = \frac{K_3}{2} \int (d_z t)^2 dV.
$$

From this basis, we can explore the mechanical stability of columnar material to applied strains, a key feature of filamentous assemblies that is unique to their
One such example is the Helfrich-Hurault effect, where filaments will buckle under a transversely applied strain [92]. This can be posed as a simple boundary value problem, that we will now summarize from reference [93]. Beginning with filaments that are aligned along the $z$ axis and confined between two plates at $x = 0$ and $x = D$, visualized in Fig. 1.6. We apply a stress along the $x$ axis, that will induce a homogenous strain response of $u_0(\mathbf{r}) = \gamma x \hat{x}$, where $\mathbf{r}$ is the coordinate position. It can be shown that in the limit of zero splay cost, the system is unstable to a small perturbation of $\mathbf{u}' = \mathbf{u} - u_0$, where we consider

$$u' = \epsilon \sin(\pi x / D) \sin(kz) \hat{x}.$$  

(1.4)

Solving for the free energy and minimizing over the wave vector, $k$, we find that

$$k \approx \sqrt{\gamma(\lambda + 2\mu)}.$$  

(1.5)
This yields a free energy in the lowest powers of $\epsilon$ of

$$F \approx \text{const} + \frac{\epsilon^2}{8} \left( (\lambda + 2\mu)(\pi/D)^2 - \frac{\gamma(\lambda + 2\mu)}{4K_3} \right) + O(\epsilon^4). \quad (1.6)$$

If the coefficient of $\epsilon^2$ is negative, then the perturbation will grow. This threshold strain occurs at

$$\gamma^* \approx \left( \frac{2\pi}{D} \right) \left( \frac{K_3}{\lambda + 2\mu} \right)^{1/2}. \quad (1.7)$$

In conclusion, his shows that columnar materials are unstable to tensional strains applied in a direction perpendicular to the filaments.

As one may have surmised from section 1.3, topological defects can effectively introduce exactly these tensile strains required for mechanical instability. A 5-fold disclination introduces displacements linear in $r$, resulting in an azimuthal stress that is tensional everywhere

$$\sigma^\text{disc}_{\phi\phi} \approx sK_0(1 + \log r/R), \quad (1.8)$$

where $K_0 = 4\mu(\lambda + \mu)/(\lambda + 2\mu)$, is the 2D Young’s modulus, and $R$ is the outer radius of the bundle.

Alternatively, twist has the opposite effect of a 5-fold disclination, by introducing azimuthal compression as a result of filaments tilting into each other. When twisting filaments along the $z$ axis, with a rate $\Omega$, the in-plane component of our tangent is

$$\hat{t}_\perp(r) = \Omega \hat{r} \hat{\phi},$$

where $r \hat{\phi} = -y' \hat{x} + x' \hat{y}$. Solving for the twist-induced stress yields

$$\sigma^\text{twist}_{\phi\phi} \approx K_0(R^2 - 3r^2)\Omega^2. \quad (1.9)$$

It is important to note that twist does not produce a stress (eqn (1.9)) that is exactly opposite that of the disclination (eqn (1.9)). Indeed eqn (1.9) shows that only the outer region exhibits compressive stress, while the inner region is still tensile. Thus it can be said that twist and 5-fold disclinations screen each other, but don’t cancel each other out.
Combining the full solutions of eqn (1.8) and eqn (1.9), we can derive the strain energy of a filament bundle with both a centered 5-fold disclination and twist

\[
E_S = \frac{\pi L}{K_0} \int drr (\sigma^{\text{disc}}_{kk} + \sigma^{\text{twist}}_{kk})^2
= VK_0 \left( \frac{3(\Omega R)^4}{128} + \frac{s^2}{32\pi^2} - \frac{3(\Omega R)^2 s}{64\pi} \right).
\] (1.10)

The first term represents the nonlinear energetic cost of the unscreened twist; the second term represents the self-energy of a single disclination, which is always positive even for \( s < 0 \); and the third term represents the interaction between twist and disclinations, which is negative when \( s = +2\pi/6 \) for a 5-fold disclination.

By including defect-defect interactions, one can derive the full energy landscape for the relation between twist and disclinations. This is summarized in Fig. 1.7, from reference [90], where is it shown that the strain energy approaches a finite maximum value for large values of twist, due to the screening power of the disclinations.

A further consideration is the bending cost of twist, which resists twist with an energy of

\[
E_B = VK_3 \frac{3(\Omega R)^4}{4}.
\] (1.11)

By minimizing the total energy, \( E_T = E_S + E_B \), we find that an optimal degree of twist to screen a centered 5-fold disclination is

\[
\Omega^* R = \sqrt{3 + \frac{32K_3}{R^2K_0}}^{-1},
\] (1.12)

which gives the surprising results that, for a bundle of flexible filaments \( (K_3/R^2K_0 \to 0) \), it is independent of the bundle size.

The preceding continuum elasticity theory yields intriguing results, but is limited in its ability to fully understand the connection between twist and packing topology. The model is valid only in the limit of small strain, and hence is unreliable for large
Figure 1.7. Plot of strain energy vs bundle twist from reference [90]. Each new colored segment represents a discrete increase in the number of 5-fold disclinations.

twist and/or large numbers of defects. Additionally, this model only addresses packing topology in a continuous manner, something which is inherently a very discrete problem for bundles composed of finite numbers of filaments, and is not able to derive the exact locations of defects. With a nod to these nuances, in chapter 2 we will introduce a discrete model of twisted adhesive filament bundles, with a focus on analyzing the cross-sectional packing structure for defects.

1.5 Conclusions

The main purpose of this dissertation is to explain the connections between the 2D packing topology and the 3D structure, in equilibrium states of cohesive filament bundles. The questions we pose are generally geometric in nature, and are influenced strongly by the non-linear effects of tilt on interfilament spacings. The models presented here are intended to be of the simplest form, and yet we will quickly find complex and non-intuitional behavior. Much of this work motivated by the large
body of work connecting topological defects to curvature of surfaces, summarized in
section 1.3. The most intriguing connection is the very recent discovery of a method
for mapping between Gaussian curvature and filament tilt, explained in section 2.5.

This dissertation is divided into three chapters. In the first chapter we assume
that a bundle of filaments has an imposed twist, and search for the ground state
filament packings that use defects to minimize the twist-induced stresses (chapter 2).
The challenge here, as is common to many geometrically frustrated systems, is that it
becomes non-trivial to determine the optimal locations and number of these defects for
any non-zero amount of twist. Comparatively, it is laborious to even find the ground
state for a simple collection of under 10 interacting spheres in 3D [94, 95]. In this
chapter, we will also explicitly explain the non-linear effects of tilt on the distance of
closest contact between filaments. Our main conclusion is that the packing topology
is only dependent on the bundle’s pitch, and not its size. However, the total number
of defects, primarily dislocations, is dependent on the bundle’s radius. The findings of
this chapter are strongly correlated to similar phenomena found in curved crystalline
membranes [90, 91, 96].

Once the optimal packing structures have been established we thoroughly analyze
the thermodynamics of the equilibrium states in chapter 3. Here we differentiate
between the energetic contributions of defects upsetting the crystalline structure (as
they screen the twist-induced stresses), from the surface energy contribution derived
from rearrangement of filaments at the bundle’s boundary. The conclusion here be-
ing that for bundle composed of sufficiently long and flexible filaments, one can in
fact lower the total energy by twisting, despite the necessary inclusion of topological
defects within the packing.

In the final chapter 4, we will take an alternate view, where we will instead assume
a filament bundle to have a fixed topology (hexagonally packed with defects), and
search for the ground state 3D structures that minimize the defect-induced stresses.
This investigation is done in light of similar defect-induced buckling patterns found to exist in flexible crystalline membranes. To do this analysis, we will construct two cooperative models: a fully 3D coarse-grained bead-spring model used to determine the final buckled structures, and a continuum elasticity model used to explore the linear stability of various buckling modes. We find that 5-fold disclinations promote azimuthal patterns of tilt, resulting in twisting motifs. We also introduce and analyze a new mode of deformation, where the twist-handedness alternates directions along the bundle, and suggest that it may have a role in focusing the defect-induced stresses into the neighborhood of the defect. Off-centered defects are examined for their effect of shifting the central helical axis of the twisting filaments. And finally, 7-fold disclinations are revealed to promote radial patterns of filament tilt with inherently different behavior from their 5-fold cousins, largely due to the non-periodic nature of the tilt direction.
CHAPTER 2
INTERIOR PACKING OF FILAMENT BUNDLES

In the following chapter, we will develop a discrete model describing the interactions of cohesive filaments. This model was initially established in two previous papers [97, 98]. The purpose of this model is to allow us to study the complex dependency on twist of the structure and energy of a filamentous bundle. We are motivated to understand the structures of the numerous systems that form twist, such as those found in nature: collagen [1, 2, 3], fibrin [4, 5], sickle hemoglobin macrofibers [6, 7], and condensed DNA [8, 9, 10]; as well as artificial materials: carbon nanotube ropes [11, 12, 13, 14] and micropatterned filament arrays [18]. In this context, our model does not consider the origin of the twist (chiral interactions) but instead makes predictions for the optimal packing and energy for a given twist. In chapter 3, this model will be used to understand the surface energy’s impact on the thermodynamics of twisted ground state bundles; and in chapter 4, it will be adapted to a fully 3D model and applied to assemblies of achiral filaments to consider the possibility of spontaneous twist for bundles of fixed topology.

First, in section 2.1 we will define a working concept of interfilament contact and cohesive interactions. Then in section 2.2 we establish the geometry of a twisted bundle and its implications on interfilament contact. In section 2.3, this model will be applied to numerical simulations as a means to study the dependence of the filament packing topology on twist. In section 2.4 we lay out the framework for studying packing defects found within the cross section as a stepping stone to describing the role of defects on the total cohesive energy. In section 2.5 we will derive a mapping
between this problem, and the problem of packing particles on a curved surface, where the source of geometric frustration has been replaced by the Gaussian curvature of the surface. Finally, in section 2.6, we will analyze the energy of the filament bundles as it depends on twist, with a focus on the interior component that is most influenced by the packing structure.

2.1 Model of filament contact and cohesive interactions

A single filament is easily modeled using the Frenet-Serret apparatus for describing the positions $X(s)$, of a curve in space as a function of an arc length, $s$ [99]. For a continuous smooth curve, the tangent can be defined at any point as $T(s) = X'(s)$. An osculating circle that describes how the tangent changes with arc length can be locally fitted to the curve at any point, with a radius of inverse curvature, $\kappa(s) = \| T'(s) \|$. The direction of this curvature at any point is defined as the normal vector, $N$, where $N(s)\kappa(s) = T'(s)$. To complete the full set of three orthogonal vectors that define the local geometry of a curve we define the binormal, $B(s) = T(s) \times N(s)$. Only one more quantity is required to fully define a unique curve, one that measures the rate of change of the osculating circle’s plane, known as the torsion, $\tau$, where $B'(s) = -\tau(s)N(s)$. This mathematical representation of a curve is used throughout this thesis to describe the centerline positions of filaments, and thus the definition of curvature will allow us to ascribe an energy cost to bending filaments.

Here, we develop a model of pair-wise interactions between portions of filaments (arc-length elements) and their cohesive energy expressed in terms of the local geometry of inter-filament contact. In this proposal, we consider bundles of homogenous and mutually-attractive filaments, where the interactions and mechanics can be described purely in terms of the shape of the filament center line, $X_i(s_i)$, which describes the position of filament $i$ at arc-position $s_i$ along the backbone. We assume that the filament pairs interact via the summation of short-ranged, pairwise interactions between
arc-length elements, such that the interaction between filaments $i$ and $j$ has the form,

$$E_{ij} = \int ds_i \int ds_j V(|X_i(s_i) - X_j(s_j)|),$$  \hspace{1cm} (2.1)$$

where $V(r)$ is an isotropic, finite-range potential describing interactions between length elements.

While a given length element at $s_i$ on the filament $i$ interacts with the entire length on $j$, the finite range of segment interactions generically implies that interactions of filament $i$ at $s_i$ are dominated by the region of filament $j$ closest to $X_i(s_i)$, which we call the contact region. Sufficiently, far from the filament ends, we may describe the contact geometry of $s_i$ with filament $j$, in terms of $s^*_j(s_i)$, a function that maps the arc-position on $i$ to the position on $j$ closest to $X_i(s_i)$, which we call the point of contact. Notably, this allows for a generic expansion of the shape of the contacting filament around the point of contact, in terms of the local geometry of $j$ and distance along $j$ from this point, $\delta s_j = s_j - s^*_j(s_i)$,

$$X_j(s_j) = X_j(s^*_j) + \delta s_j T_j + (\delta s_j)^2 \frac{\kappa_j}{2} N_j + O[(\delta s_j)^3],$$  \hspace{1cm} (2.2)$$

where $T_j, N_j$ and $\kappa_j$, are the tangent, normal and curvature of filament $j$ at $s^*_j(s_i)$ [99]. From this expression we find the interfilament distance, $\Delta \equiv X_j(s_j) - X_i(s_i)$, from

$$|\Delta(\delta s_j)|^2 = |\Delta_j|^2 + (\delta s_j)^2(1 + \kappa_j \Delta_{ij} \cdot N_j) + O[(\delta s_j)^3],$$  \hspace{1cm} (2.3)$$

where $\Delta_{ij} = X_j(s^*_j) - X_i(s_i)$ is the distance of closest contact from $s_i$ to filament $j$, such that $\Delta_{ij} \cdot T_j = 0$. These parameters are shown schematically in Fig. 2.1.

Assuming that $V(r)$ is sufficiently short-ranged compared to filament length and curvature, we may use eqn (2.3) to derive the cohesive energy contribution of the length element at $s_i$. 

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Figure 2.1. (a) One and a half pitch lengths of a contacting helical filament pair, \( i \) and \( j \). The helical radius and helical angle for filament \( j \) are \( \rho_j \) and \( \theta_j(\rho) \) respectively.

(b) A view of a region of contact, showing the filament parameters defined in eqns (2.2) and (2.3).

\[
dE_{ij} = ds_i \int_{-\infty}^{+\infty} d(\delta s_j)V(|\Delta(\delta s_j)|) = \frac{\gamma(\Delta_{ij})}{\sqrt{1 + \kappa_j \Delta_{ij} \cdot N_j}} ds_i, \quad (2.4)
\]

where

\[
\gamma(\Delta_{ij}) = \int_{-\infty}^{+\infty} du \, V(\sqrt{\Delta^2_{ij} + u^2}), \quad (2.5)
\]

is a local contact potential, describing the cohesive energy per unit length of filament \( i \) in contact with filament \( j \). This potential is a function of the local distance of closest contact between the filaments. The derivation of this local inter-filament cohesive energy is fully general for any short-ranged potential. In the remainder of this proposal we study the case of a Lennard-Jones interaction between length elements, \( V(\Delta) = \epsilon[(\sigma/\Delta)^{12} - 2(\sigma/\Delta)^6] \), for which the contact potential becomes,
\[
\gamma(\Delta_{ij}) = \gamma_0 \left[ \frac{5}{6} \left( \frac{d}{\Delta_{ij}} \right)^{11} - \frac{11}{6} \left( \frac{d}{\Delta_{ij}} \right)^5 \right],
\]  
(2.6)

which, similar to the Lennard-Jones, has stiff repulsion at short-range and soft attraction at long range. This potential is characterized by an attractive well of depth \( \gamma_0 = 1.6862 \epsilon \sigma \), at a preferred spacing \( d = 0.9471 \sigma \), which we denote as the filament diameter.

Eqn (2.4) describes how short-ranged, pair-wise interactions between all length elements of a filament pair can be formulated in terms of functions of the local contact geometry (e.g. \( \Delta_{ij} \), \( \kappa_j \), and \( N_j \)) integrated over the contacting length of a single filament (in this case, filament \( i \)). In the following section, we derive this contact geometry for filament pairs in twisted bundles.

### 2.2 Filament geometry in twisted bundles

In this section, we derive the geometry of inter-filament contact in bundles twisted around the central \( \hat{z} \) axis at a uniform rate of rotation \( \Omega \), as shown in Fig. 2.2. In this manner, all filaments within a bundle have the identical pitch, \( P \), where \( P = 2\pi/\Omega \). This constant pitch assumption is valid for most materials, as it maximizes the amount of contact between filaments. That is to say, if two filaments are in contact at one location of the bundle, they are in contact everywhere. This assumption reduces the problem of filament assembly to one of packing filaments in the 2D plane of the bundle’s cross section perpendicular to \( \hat{z} \), where the packing structure between any two cross sections differ only by the rigid rotation about the \( \hat{z} \) axis. In this manner the geometric frustration derives from the in-plane tilt of the filaments.

Defining the position of filament \( i \) at a common plane \( z = 0 \) (arc-position \( s_i = 0 \)), by the polar coordinates \( (\rho_i, \phi_i) \), the shape of the filament backbone follows the helical curve,

\[
X_i(z_i) = \rho_i \cos (\phi_i + \Omega z_i) \hat{x} + \rho_i \sin (\phi_i + \Omega z_i) \hat{y} + z_i \hat{z},
\]  
(2.7)
Figure 2.2. Examples of three cohesive bundles comprised of 145 filaments of a fixed length, $L = 40d$. From left to right, $\theta = 0^\circ, 31.2^\circ$, and $61.2^\circ$. Additional parameters are shown: the filament contour length, $L$, the bundle radius, $R$, and the twist angle, $\theta$, defined as the twist angle, and is the angle of the tangent of the outermost filaments with the central $\hat{z}$ axis. Coloring is simply used to highlight radial depth of filament position.

where it is convenient to express position in terms of vertical height $z_i = s_i \cos \theta(\rho_i)$, where $\theta(\rho) = \arctan(\Omega \rho)$ is the helical angle of filament $i$ with respect to $\hat{z}$, shown in Fig. 2.1(a).\(^1\) The unit normal and curvature are easily calculated from the second-derivative of $X_j$ with respect to arc-length,

$$\kappa_j = \frac{\Omega^2 \rho_j}{1 + (\Omega \rho_j)^2} \quad (2.8)$$

$$N_j = -\cos (\phi_j + \Omega z_j) \hat{x} - \sin (\phi_j + \Omega z_j) \hat{y}. \quad (2.9)$$

\(^1\)Note, that $\theta(\rho)$ refers to the local tilt angle of filaments at a radius $\rho$ in the bundle, where as, in our notation, when tilt angle appears without an explicit radius, as $\theta$, it refers to helical angle at the outer radius of the bundle, which we call the twist angle of the bundle.
For a pair of filaments, \(i\) and \(j\), it is convenient to express the separation between curves in terms of a vertical offset, \(z_{ij} = z_i - z_j\), and the angular separation in the plane, \(\phi_{ij} = \phi_i - \phi_j\),

\[
\Delta^2(z_{ij}) = \rho_i^2 + \rho_j^2 - 2\rho_i\rho_j \cos(\phi_{ij} + \Omega z_{ij}) + z_{ij}^2. \tag{2.10}
\]

The distance of closest contact between \(i\) and \(j\) is determined by the minimization of \(\Delta^2(z_{ij})\), with respect to the vertical offset between contacting points, to find the height separation at the distance of closest contact, \(z^*_{ij}\). From \(d\Delta^2(z_{ij})/dz_{ij} = 0\), we find a transcendental equation satisfied by \(z^*_{ij}\),

\[
\Omega z^*_{ij} = -\Omega^2 \rho_i \rho_j \sin(\phi_{ij} + \Omega z^*_{ij}). \tag{2.11}
\]

To make practical use of this condition, we examine the solutions in the limiting cases where \(\Omega^2 \rho_i \rho_j\) is either small or asymptotically large. The former case corresponds to either small twist, or filament positions sufficiently close to the center of the bundle, in which case the Taylor series expansion of eqn (2.11) yields,

\[
\lim_{\Omega^2 \rho_i \rho_j \ll 1} \Omega z^*_{ij} = -\frac{\Omega^2 \rho_i \rho_j \sin \phi_{ij}}{1 + \Omega^2 \rho_i \rho_j \cos \phi_{ij}} + O[(\Omega^2 \rho_i \rho_j)^3], \tag{2.12}
\]

which represents a modest tipping out-of-plane of the distance of closest contact between azimuthally-separated filaments.

In the opposite limit, filaments are far from the core of the bundle in comparison to their helical pitch. Since \(z^*_{ij}\) is always strictly less than the pitch, the left-hand side of eqn (2.11) is never larger than \(2\pi\) in magnitude, hence, in the limit \(\Omega^2 \rho_i \rho_j \gg 1\), the solution for \(z_{ij}\) must satisfy \(\sin(\phi_{ij} + z^*_{ij}) \propto (\Omega^2 \rho_i \rho_j)\) so that in the asymptotic limit of large helical angles, we have

\[
\lim_{\Omega^2 \rho_i \rho_j \gg 1} \Omega z^*_{ij} = -\phi_{ij}. \tag{2.13}
\]
which shows that the inter-filament distance of closest contact is nearly vertical far from the core of bundle.

These simple results can be incorporated into an approximate formula for vertical contact separation that interpolates between the two asymptotic limits,

$$\Omega z_{ij}^* \simeq -\arctan\left(\frac{\Omega^2 \rho_i \rho_j \sin \phi_{ij}}{1 + \Omega^2 \rho_i \rho_j \cos \phi_{ij}}\right), \quad (2.14)$$

We employ this form of $z_{ij}^*$ to approximate distance of closest contact in our numerical studies via the relation $\Delta_{ij} = \Delta(z_{ij}^*)$. Notably, this approximation of $\Delta_{ij}$ becomes poor only for filament pairs distant from the center (large $\rho$) and on opposing sides of the bundle ($\phi_{ij} \approx \pi$), thus providing an accurate description for filament interactions in twisted bundles incorporating sufficiently short-ranged potentials ($|\Omega d| \ll 1$).

### 2.3 Simulation methods

To study the twist-dependence of the ground-state packing and cohesive energy of filament bundles, we numerically minimize the sum of pair-wise interactions described by eqns (2.4) and (2.6), based on the approximation of $\Delta_{ij} = \Delta(z_{ij}^*)$, described previously in eqn (2.14). Since the interfilament contact geometry is constant along the bundle height, the cohesive energy for the $i$ and $j$ pair becomes,

$$E_{ij} = \frac{\gamma(\Delta_{ij}) L_{ij}}{\sqrt{1 + \kappa_j \Delta_{ij} \cdot \mathbf{N}_j}}, \quad (2.15)$$

where $L_{ij}$ is defined as the length of filament $i$ in contact with filament $j$. In this section, we consider only the case of infinite length, where filament contact is maintained along the full contour $L_{ij} = L$, provided that we take $i$ to be the “outer” filament ($\rho_i \gtrsim \rho_j$). In this way we may neglect any explicit change of filament contact length at the ends of the bundle in the $L \to \infty$ limit.
We numerically optimize the total cohesive energy for bundles of fixed $N$ and $\Omega$. Our approach is to generate 2000 random initial configurations of in-plane filament positions, $\{\mathbf{X}_i(s_i = 0)\}$, and then seek a minimal-energy configuration of in-plane filament positions using the method of steepest descent. For the purposes of efficiency, inter-filament forces are truncated beyond a cutoff distance of $4d$. This is implemented via a Verlet neighbor list, which is completely regenerated after any filament has moved a distance of $1d$. Once a suitable minima is reached, the interaction cutoff is removed for the final relaxation step. From this, we have found minimum energy packings for bundles with $N$ between 16 and 196, and a multitude of twist angles between $0^\circ$ and $\sim 80^\circ$.

The energy landscape of twisted bundles is particularly complex and possesses a large number of nearly degenerate minima, especially for large bundles where $N \gtrsim 50$.

We find the number of local energy minima appears to grow exponentially with the system size. This is a feature seemingly common among systems that include defects in their lowest energy packings, such as the Thomson problem where repulsive particles are packed on the surface of a sphere [89]. The complexity of the energy landscape can be seen in Fig. 2.3, which shows the distribution of local energy minima resolved by our algorithm for a particular bundle size and twist. Given the huge number of the minima in the complex energy landscape of a large twisted bundle, it is possible that even the large number of initial configurations used here is insufficient to resolve absolute lowest global energy states (especially for large $N$). Notwithstanding the unavoidable roughness of the ground state manifold of frustrated crystals [85], we find that all states within a neighborhood of the the global minima are characterized by equal, or nearly equal, geometry and total energy. Additional details on the accuracy of our method with regarding energy and geometry are shown in Fig. 2.9 and Fig. 3.1 respectively.
Figure 2.3. Each data point is the result of a simulated quenches with $N = 80$, $\Omega = 0.15$, and random initial starting filament locations. Numbers in the legend represent the topological charge, $Q$ of the interior packing (introduced next in section 2.4). There are 1,000 simulations total, of which just six achieve the ground state energy. Only the lowest energy result is retained for full analysis.
2.4 Characterizing the interior packing of twisted bundles

The resultant packings are analyzed in terms of the network of nearest-neighbor contacts. The bond network is formally defined through a specialized Delaunay triangulation that takes into account the true (out-of-plane) distance of closest contact between filament pairs. The full details of this triangulation are given in appendix A. This procedure produces a unique network of non-overlapping bonds, whose “nearest neighbor” connections serve as a measure of the local packing geometry. In the case of an untwisted bundle, the network universally describes the 6-fold, hexagonal packing; however, for sufficiently large twist uniform 6-fold coordination is not maintained and the packing becomes interrupted by topological defects, as suggested previously in section 1.2. It will be shown in section 2.6 that the presence of these defects greatly influences the total cohesive energy, therefore they will be discussed in more detail here.

A primary type of topological defect in 2D crystalline packings is a disclination, which describes the breakdown of long-range $n$-fold orientational symmetry in the lattice at a singular point [80]. In a hexagonal crystal, low-energy disclinations are typically of two types: 5-fold and 7-fold. At the core of a 5-fold disclination, is a lattice site (corresponding to a vertex in the bond network) with five nearest neighbors, one fewer than the six neighbors of the perfect lattice. While the large elastic strains generated by the defect throughout the crystal generically make even single disclinations prohibitively expensive in most types of (planar) 2D crystals, our simulations reveal that disclinations are necessary components in the minimal-energy packings of twisted filament bundles.

Following the topological characterization of disclinations in crystalline solids [80], we define the total topological charge of disclinations as

$$Q \equiv \sum_{n} (6 - n)V(n), \quad (2.16)$$
Figure 2.4. Example cross sections of for three example bundle sizes and seven example twist angles, $\theta$, showing two trends: 1) the topological charge, $Q$, increases with twist angle, and 2) the total number of defects, $N_{\text{Disclination}}$, increases with bundle size. 5 and 7-fold disclinations are colored red and blue respectively. The bottom left two cross sections show their triangulated nearest neighbor bond network. The roman numeral labels match specific bundles to their locations within Fig. 2.5.

where $n$ is a coordination number of nearest neighbors belonging to a filament, and $V(n)$ is the number of *internal* filaments that possess $n$ nearest neighbors. Non-internal, or *boundary* filaments, are distinguished by having at least one neighbor bond on the outer edge of the bond network.

Throughout this chapter, we consider the structure and energetics of three discrete bundle sizes in details: small ($N = 34$); intermediate ($N = 82$); and large ($N = 184$). In Fig. 2.4 we show the evolution of ground-state packings these three bundle sizes. The packing of each of these bundles exhibits a common trend with increasing twist. Packings evolve from defect-free $Q = 0$ at zero twist, to an increasing value of topological charge—characterized a *universal excess 5-fold defects* with increasing twist. As the intermediate and large bundle packings illustrate, while the ground-state packing may include 7-fold disclinations, *negatively-charged* defects are always sufficiently outnumbered such that the net charge *increases* with twist.
Figure 2.5. (a) Phase diagram of net disclination charge, \( Q \), with \( \theta \) and \( N \). (b) Phase diagram of \( N_{\text{Disclinations}}/Q \), with twist angle \( \theta \), and number of filaments, \( N \). Black lines are shown to roughly delineate regions of qualitatively distinct ground-state packing. The roman numeral labels correspond to bundle cross sections seen in Fig. 2.4 and part (d). (c) A zoomed-in view of a seven disclination long \( Q = 1 \) scar, with bond network shown, for a bundle with \( N = 196 \) and \( \theta = 25.7^\circ \). In (d) we show five examples of bundles that exist in the variable-size defect region. 5, 7, and 8-fold disclinations are colored red, blue, and purple respectively.

By analyzing the topological charge, we construct a phase diagram bundle ground states in terms of twist angle, \( \theta \), and number of filaments, \( N \), shown if Fig. 2.5a. Notwithstanding some obvious limitation of small bundle sizes to reach large values of \( Q \), it becomes clear that the optimal value of topological charge is predominantly determined by \( \theta \), and largely independent of, \( N \). \( Q \) reaches a maximum of six at \( \theta \gtrsim 70^\circ \). Further simulations show that \( Q \) does not increased above 6, even for twist angles of up to \( 87^\circ \). We discuss the geometric origin of this universal dependence of \( Q \) on \( \theta \) in the next section.
While the net charge is largely fixed at a given twist angle, there are many ways to achieve a particular value of $Q$. Specifically, 5 and 7-fold disclinations that appear in “neutral” pairs do not adjust this value. A commonly observed example occurs for $Q = 1$ bundles, which have $n$ 5-fold disclinations and $(n - 1)$ 7-fold disclinations. For large bundles this feature becomes important, as shown in the 2nd column of Fig. 2.4 (bundles $Ila$, $IIb$, and $IIC$). While these three examples have the same twist angles and hence maintain $Q = 1$, the total number of disclinations, $N_{\text{Disclination}}$, increases from 1 to 3 to 5 for small, intermediate and large bundles, respectively.

A second phase portrait of the number of disclinations, $N_{\text{Disclination}}$, per charge, $Q$, is show in Fig. 2.5b. Unlike the net disclination charge, the optimal value of $N_{\text{Disclination}}/Q$ in ground-state bundles varies with both $N$, as well as $\theta$. In Fig. 2.5b we have roughly delineated the $Q \neq 0$ behavior into four regions. In the small-$N$ red region, $N_{\text{Disclination}} = Q$, and bundles contain primarily only 5-fold disclinations, without excess 5-7 pairs. For larger filament number, shown as a light blue region, $N_{\text{Disclination}} = 3Q$. Ground states within this region contain structures such as $IIm$, where there are one 7-fold and two 5-fold disclinations per topological charge. At even larger bundle sizes is the pink region that continues this trend, i.e. there are two 7-fold and three 5-fold disclinations per topological charge, as seen in $IIC$. This trend continues with increasing $N$, reaching $N_{\text{Disclination}} = 7$ (as seen in Fig. 2.5c) and higher for simulations not shown. A final region is loosely defined at the upper limits of twists, where packings identified in our ground-state search belong to a more complex taxonomy, possessing in general, non-integer values of $N_{\text{Disclination}}/Q$. This is achieved using solely 5 and 7-fold disclinations, like in $Xb$, $Xc$, and $Xd$, or with higher-charge disclinations like the 8-fold disclination shown in $Xa$ (Fig. 2.5d).
2.5 Mapping to the bundle-equivalent dome

In this section we briefly review the connection between twisted bundle packing and packing on spherically-curved surfaces. This mapping provides additional intuition regarding the existence of defects within the packing, as well as a more natural way of describing the bond network. Furthermore, it directly relates this research with the already well studied problem of particle packing on curved surfaces [88, 24, 89, 100, 82]. Here the in-plane tilt of filaments acting as the source of geometric frustration has been replaced by the Gaussian curvature of the surface itself.

The universal twist dependence of $Q$ in ground-state bundles derives from a formal mapping of the inter-filament distance and the geodesic distances between points on a dome-like surface that encodes the metric properties of bundles. The geometry of the bundle-equivalent dome can be constructed from simple considerations of the space available for packing filaments at a radial distance, $\rho$, from the center of a twisted bundle, characterized the length, $\ell(\rho)$, of a span between points of “self-contact” on a helical curve. In terms of the dual surface representation, $\ell(\rho)$ corresponds to the circumference of the surface an arc-distance $\rho$ from its pole (shown in Fig. 2.6). This length is determined by considering a helical curve at $\rho$ and the length of the shortest, constant-radius span that connects the curve to itself at another point along its distance. The finite length of the span between “self-contacts” of a helical filament at $\rho$ implies that the number of finite-diameter filaments that may be packed at a given radius is limited, as is the space available for packing finite-diameter discs at a given radius from the center of an axi-symmetric surface. Fig. 2.6 shows the geometry of this proposed surface, and the span $\ell(\rho)$, defined to be

$$\ell(\rho) = P \sin \theta(\rho) = \frac{2\pi \rho}{\sqrt{1 + (\Omega \rho)^2}}.$$  \hspace{1cm} (2.17)
Figure 2.6. Helically twisted filaments (upper row) and their corresponding discs on the mapped bundle-equivalent dome (lower row). The radial distance from the center of twist, $\rho$, is shown. Also the length, $\ell(\rho)$, is shown for the orange filament (disc), along which additional filaments (discs) can fit. Twist a filament ensures that $\ell(\rho) < 2\pi \rho$, similar to the effect of Gaussian curvature. The helical pitch, $P$, is shown for the middle image.

The circumference of the surface grows with $\rho$ more slowly than $2\pi \rho$, imply a spherically-curved geometry, characterized by a positive Gaussian curvature. Following the full derivation from appendix B, the equivalent Gaussian curvature is

$$K_G(\rho) = \frac{3\Omega^2}{[1 + (\Omega \rho)^2]^2},$$

(2.18)
showing that curvature (and hence geometric frustration) is concentrated at the pole of that surface (i.e. $\Omega \rho \ll 1$), corresponding to a region near the center of twist in a bundle. An example of bundle packings mapped in this way are shown in Fig. 2.7.

Figure 2.7. A sequence of bundles with $N = 34$ filaments, and the corresponding mapped bundle-equivalent dome, which shares the packing geometry and topology with the twisted bundle. From left-to-right: $Q = 0, \theta = 8^\circ$; $Q = 1, \theta = 28^\circ$; $Q = 2, \theta = 41^\circ$; $Q = 3, \theta = 49^\circ$. 5 and 7-fold coordinated elements are colored red and blue respectively.

We now consider the relationship between the geometry of the dual surface and the topology of the triangular network of nearest-neighbor bonds of particles packed on this surface (see e.g. Fig. 2.7(b)), whose constraints carry over to the packing geometry of twisted bundles. For a triangular element, connecting three vertices of the packing, the Gauss-Bonnet theorem relates the integrated Gaussian curvature of the surface patch within the element to the internal angles, $\theta_i$, at vertices,

$$\int_{\text{patch}} dA \ K_G + \pi = \sum_{i=1}^{3} \theta_i, \quad (2.19)$$

where we have taken the edges of the patch to be geodesics. Summing this over the entire mesh of the packing, we have
\[ \int_{\text{mesh}} dA \ K_G + \pi F = 2\pi \sum_n V(n) + \sum_b \theta_b, \] (2.20)

where \( F \) is the total number of faces in the mesh, \( V(n) \) is the number of \( n \)-fold vertices not at the boundary of the triangulation (each of contributing \( 2\pi \) from the sum of interior angles), and \( \theta_b \) are the interior angles of vertices on the boundary of the triangulation. Defining \( V_b(n) \) as the number of \( n \)-fold vertices at the boundary and using \( 3F = \sum_n \left[nV(n) + (n-1)V_b(n)\right] \), we may rewrite eqn (2.20) as

\[ \int_{\text{mesh}} dA \ K_G = 2\pi \sum_n \left(1 - \frac{n}{6}\right)V(n) + \sum_b \left(\theta_b - \frac{\pi}{3}\right), \] (2.21)

where we note that an \( n \)-fold boundary vertex possesses \((n-1)\) interior angles. Dividing by \( 2\pi \) and making use of the definition of topological charge in eqn (2.16), we have

\[ 6\chi - Q = \frac{1}{2\pi} \sum_b \left(\theta_b - \frac{\pi}{3}\right), \] (2.22)

where we have defined

\[ \chi = \frac{1}{2\pi} \int_{\text{mesh}} dA \ K_G, \] (2.23)

which plays the role of the Euler characteristic for a boundary-free surface domain, notably increasing as the lateral size of the patch grows large in comparison the curvature radius of the surface (proportional to \( P \)). The right-hand side of eqn (2.22) represents distortion of the nearest-neighbor packing from an equilateral geometry (\( \theta_b \neq \pi/3 \)) at the free boundary of cluster, such that the deficit between the topological charge of the interior packing and \( 6\chi \) must be distributed as boundary distortion of the packing. Approximating the boundary geometry of the packing as circular and integrating the Gaussian curvature within a packing of arc-radius \( R \) we find,

\[ \chi(R) = 1 - \frac{1}{\left[1 + (\Omega R)^2\right]^{3/2}}, \] (2.24)
which increases as $3(\Omega R)^2/2$ from 0 at small twists, to a maximum of 1 in the limit $|\Omega R| \to \infty$.

Based on the assumption that large deviations from equilateral packing at the boundary are energetically expensive, and therefore, unlikely in ground-state packings, we exploit this theorem to derive an expression for $Q_{id}$, the ideal value of topological charge of the interior packing, which requires no distortion from equilateral bond-order at the patch edge (and equivalently, the bundle surface). We define $Q_{id} = 6\chi(R)$ as the ideal disclination charge that perfectly neutralizes the integrated Gaussian curvature in eqn (2.22). Specifically, when the actual topological charge can achieve the ideal value ($Q = Q_{id}$), the distortion of the packing at the outer boundary from equilateral geometry ($\theta_b = \pi/3$) vanishes. Assuming an axisymmetric shape for the boundary of the packing, evaluating the integrated curvature on the dual surface gives the ideal charge purely in terms of twist angle,

$$Q_{id} = 6\left(1 - \cos^3 \theta\right). \quad (2.25)$$

Importantly, the $\theta$-dependence of $Q_{id}$ encodes the increase of integrated Gaussian curvature as the patch size grows large compared to the curvature radius of the dual surface (proportional to the pitch). As the surface domain grows to cover a larger portion of the curved surface, the preferred topological charge becomes non-zero. Though the presence of the free boundary of the bundle allows the topological charge to adjust based on purely energetic considerations, the positive curvature of the bundle-equivalent surface suggests a connection between the optimal packings of highly twisted bundles and the better-known packings of particles on closed, spherical surfaces, studied in the context of the generalized Thomson problem [88, 24, 89]. In the language of the continuum theory of curved 2D crystals, excess 5-fold disclinations screen the elastic stresses generated by geometric frustration, such that increasing cur-
vature (or twist in the case of bundles) requires an increasing number of “neutralizing”
disclinations.

Though $Q_{id}$ increases continuously from 0 at $\theta = 0^\circ$ to 6 at $\theta = 90^\circ$, the actual
topological charge of the packing may only take on integer values, the simple assump-
tion that the integer value of $Q$ in ground-state packings is determined by the closest
integer value to $Q_{id}(\theta)$ is remarkably consistent with our numerical simulations of
twisted bundles. In Fig. 2.8 we plot the $Q$ for bundle simulations vs. twist angle and
compare this to the continuously increasing value of $Q_{id}(\theta)$. While the agreement is

![Figure 2.8](image.png)

**Figure 2.8.** The ideal charge, $Q_{id}$, from eqn (2.25) (solid black line) vs twist angle $\theta$. The data points are the $Q$ values for an accumulation of 5000 simulation results for the various twist angles and bundle sizes introduced in section 2.3. The number of filaments range from $N = 16$ (red data points) to $N = 196$ (blue data points).

between $Q$ and $Q_{id}$ is imperfect, this above argument highlights the fundamentally
geometric nature of packing frustration in twisted bundles, and importantly, provides
a natural explanation for the observation that optimal values of $Q$ are independent
of $N$, determined only by $\theta$, which controls the value of the integrated curvature on the bundle-equivalent surface.

### 2.6 Twist-dependence of cohesive energy

Having analyzed the twist-dependence of the structure of minimal-energy bundles in terms of the topological charge of the packing, we now consider the twist-dependence of cohesive energy, with the aim of discerning the influence of the universal evolution of $Q$ on the cohesive energy. We find that in the large-$N$ limit, bundle energetics converge to a common behavior. Fig. 2.9 shows the change in mean cohesive energy per filament length for three bundle sizes. Twisting initially increases the

**Figure 2.9.** Mean filament cohesive energy per unit length vs twist angle, $\theta$, for three selected bundle sizes: small $N = 34$ (red), medium $N = 82$ (blue), and large $N = 184$ (green). The roughness is a consequence of sudden rearrangements in the packings to accommodate the twist-induced geometric frustration. In this and all following plots, the energy change is defined relative to the *untwisted* case. Error bars are shown for each $N$ at three select values of $\theta$. These error estimates derive from the standard deviation taken from 100 implementations of the numerical ground-state search algorithm (each of which samples $\sim 10^3$ initial configurations). These estimates suggest our sampling yields bundle energies to within less than 1% of the true ground-state energy.
energy of untwisted bundles, until reaching a rough plateau region at intermediate
twist. However, further twist lowers the cohesive energy, ultimately driving it below the energy of the untwisted state.

In chapter 3, we carefully analyze the surface geometry of twisted bundles to show that the tendency to decrease the cohesive energy with twist is driven by a decrease of non-contacting filament lengths at the boundary of long bundles. In this section, we show that removing the effects of changes in filaments at the boundary of the bundle reveals a universal dependence of the bulk packing on twist. To perform this analysis, we subtract the surface energy of the bundle from the total energy of the discrete model to define the bulk cohesion energy, $E_{\text{bulk}} = E_{\text{tot}} - E_{\text{surf}}$. The surface energy, $E_{\text{surf}}$, which is considered fully in section 3.2, accounts for the loss of favorable cohesive interactions due to the exposure of non-contacting filament lengths at a surface. More formally, the surface energy attributes a loss of cohesive energy of $\gamma_0/2$, per unit length of lost neighbor contact, relative to the hexagonal packing of the bulk. The contributions to $E_{\text{surf}}$ are a combination of the surface energy from bundle sides (calculated from eqn (3.10), and ends (calculated from eqn (3.12). Fig. 2.10 shows the change in mean $E_{\text{bulk}}$ per filament length for large bundles in the range of $N = 166 - 193$, revealing a common increase of energy relative to the untwisted state. For small twist, the bulk energy increases smoothly with $\theta$, due to the increasing frustration of inter-filament spacing in defect free bundles. The small-$\theta$ dependence of large-$N$ bundles is consistent with results of elasticity theory calculations which show that $E_{\text{bulk}} \sim \theta^4$ in defect-free bundles [90, 91]. Also consistent with elasticity theory results, is the appearance of cusps in $E_{\text{bulk}}$ at transitions in topological charge, such as the transition from $Q = 0$ to $Q = +1$ at $\theta \simeq 27^\circ$. Just beyond the transition, energy decreases with twist, highlighting the key ability of excess disclinations to mitigate the twist-induced frustration, taming the rapid growth in bundle energy. The five
local minima mark values of twist that are locally stable due the optimal screening provided by discrete values of $Q$.

To summarize, this analysis of the bulk cohesive energy of ground-state packings of our discrete filament model reveals two key influences of twist on the cohesive energy of bundles. First, twist frustrates the uniform inter-filament packing allowed in straight bundles, leading to a necessary increase in energy with twist angle. Second, we find that increasing twist triggers the stability of excess 5-fold disclinations in the cross-sectional packing that mitigates the growth of bundle energy associated with twist-induced frustration of nearest-neighbor spacing. Additional description of the importance of defects on the cohesive energy and how suppressing them leads to higher energy highly stressed states is given in the appendix C.
2.7 Conclusions

In this chapter we introduced a model for interfilament contact and cohesive energy. We then discussed the interior cross-sectional packing topology of a filamentous bundle. Next we derived the method of mapping the positions of filaments in the cross section of a twisted bundle to the positions of particles on the bundle-equivalent dome surface, and illustrated the connection between the topology of the two geometrically frustrated systems. Finally, we explained the topology dependence of the sharp transitions in the interior cohesive energy’s dependence on twist. In the following chapter we will further build upon this model by discussing the thermodynamic trends of ground-state bundles, and the strong influence of exterior filaments.
CHAPTER 3
SURFACE ENERGY OF FILAMENT BUNDLES

In the previous chapter we discussed a bundle’s topology and general energy dependence on twist. In this chapter we provide the remaining details pertaining to a bundle’s exterior geometry and energy. A filament bundle is a finite structure (in number of filaments and length), and therefore its surface can account for a significant contribution to the total energy. The exterior geometry has a non-linear dependence on twist, and must be understood to discern the full relation between twist and cohesive energy.

Beginning in section 3.1, we derive a continuum model that describes the twist-dependent global geometry of a bundle’s shape, first introduced in reference [97]. Then in section 3.2, we further this model by establishing a surface energy cost that derives from an exterior filament’s deficit from the ideal cohesive energy gained by existing in the bulk. Next, in section 3.4, we add the energy cost of bending filaments to our continuum model. Finally in section 3.3, we give some of the results of this model and make a few comparisons to the discrete model from chapter 2.

3.1 Surface geometry

In this section, we analyze the surface geometry of twisted filament bundles in the continuum limit where filament diameter is small compared to both filament length and the lateral size of the bundle, with the goal of developing an analytical formula for the dependence of surface energy on twist. Our model supposes a fixed constraint on the total volume of the bundle, and a fixed length on every filament. This model
is appropriate to systems such as artificially manufactured filament bundles [18]. Alternatively, many filamentous biological materials, such as collagen [1], are able to adjust their length through the addition/subtraction of subunits. However, our main conclusions of this chapter are largely unaffected by the exact model chosen (at least qualitatively), especially in the limit of very long bundles when the amount of exposed filaments at the ends are insignificant compared to those on the outer hull of the bundle. We make the additional approximations that: 1) the filament packing is locally hexagonally-close packed, with a density that is independent of twist; and 2) the shape of the bundle is axisymmetric, with an outer cylindrical radius, $R$. The first approximation is clearly violated in the neighborhood of defects that enter the packing at finite twist. Though, for large bundles $N \gg 1$, the local packing is non-hexagonal for only a relative minority of filaments. Hence, we assume that the local filament spacing and occupied volume fraction change only modestly in the bulk of twisted bundles, which is consistent with the density of maximally-compact bundles studied in chapter 2. For straight filament bundles, the second approximation (cylindrical bundle symmetry) clearly fails to capture the hexagonal faceting of the bundle sides. However, our simulations show that bundles become more axisymmetric at high twist as the packing trades high-energy corners at the bundle surface for excess disclinations in the bundle interior.$^1$

Based on these assumptions, we now consider the change in the surface shape with twist (shown in Fig. 3.1). As twist increases, the helical tilt of filaments away from the center of rotation increases as $\theta(\rho) = \arctan(\Omega \rho)$. Since the contour length of filaments are fixed to $L$, twist requires a change of the height, $H$, the extent of a filament along the pitch axis, according to

---

$^1$This follows from the fact that the sum of topological charges interior to the bundle and the net deficit of nearest neighbor contacts at the boundary are constrained to obey, $Q + \sum_n (4-n)V_6(n) = 6$. See, e.g. [82].
Figure 3.1. Filament bundle radius, $R/R_0$ vs twist angle, $\theta$ from numerically simulated ground states. The black line is the continuum model prediction of eqn (3.2), while the discrete model is represented with three bundle sizes: small $N = 34$ (red), medium $N = 82$ (blue), and large $N = 184$ (green). Both the continuum approximation (opaque pink surface, radius is $R + d$ to account for the filament diameter), and the discrete model representations are shown together for three example twist angles for a bundle of $N = 46$. The height of the outermost filaments is shown for example C. Error bars are shown for each $N$ at three select values of $\theta$, these represent the standard deviation (for 100 trials) of the low energy state found via the ground-state search algorithm.

$$H(\rho) = L \cos \theta(\rho) = \frac{L}{\sqrt{1 + (\Omega \rho)^2}}. \quad (3.1)$$

Hence, for non-zero $\Omega$, the vertical profile of the ends varies with radius. Assuming filaments distribute the taper equally over both of the free ends of the bundle, the shape of this tapered profile is described by $H(\rho)/2$, as seen in the example bundles in Fig. 3.1. It is important to point out that the curved shape of this profile is unrelated to the geometry of the bundle-equivalent surface which encodes the metric properties of interfilament spacing (see section 2.5).
Local decreases in the height of the bundle with twist imply that the lateral radius must necessarily increase in order to preserve a constant volume and density. Given a rate of twist $\Omega$, and an outer bundle radius $R$, the volume within a bundle is easily computed as $V(\Omega, R) = 2\pi\Omega^{-2}L\left(\sqrt{1 + (\Omega R)^2} - 1\right)$. Assuming an untwisted bundle radius of $R_0$, the $\Omega$-dependence of $R$ is determined from the solution to $V(\Omega, R) = \pi R_0^2 L$,

$$R = R_0\sqrt{1 + (\Omega R_0/2)^2}. \quad (3.2)$$

This formula, while derived from global considerations of volume conservation, implicitly encodes the same constraints of lateral filament-packing in twisted bundles described by the mapping to the dome-like surface: twisting a bundle reduces the number of filaments that can be packed at a given radius $\rho$ by $\cos \theta(\rho)$, implying that filaments must be redistributed to larger radii, and notably compares quantitatively with simulated bundle radii, as shown in Fig. 3.1.

### 3.2 Continuum-limit surface energy

We now proceed to analyze the contribution to the cohesive energy deriving from the twist-induced changes of filament contact at the surface of the bundle. The surface energy per unit area, $\Sigma$, accounts for the loss of favorable cohesive interactions due to exposure of non-contacting filament lengths at the surface. Simply put, the surface energy attributes a loss of cohesive energy, $\gamma_0/2$, per unit length of lost neighbor contact, relative to the locally-hexagonal packing of the bulk.\(^2\) Note that even for fixed $N$, twist leads to non-trivial variations of filament contact at the surface; hence, our continuum model is twist-dependent. Additionally, because packing defects tend to be located near the bundle’s center, while the outer filaments remain hexagonally packed, we can fairly compare this continuum model with the results from our discrete

\(^2\)Following standard arguments for surface energy, the factor of $1/2$ follows from the fact that the separation of one contacting pair creates two non-contacting filaments [101].
simulation, by assuming the lost neighbor contacts at the surface are a continuation of the hexagonal packing.

The surface of a bundle is composed of exposed filaments that lack the full complement of neighbors, relative to an ideal hexagonal bulk packing that achieves the maximal cohesive energy density. In a twisted filament bundle, non-contacting filament lengths arise in two ways. First, filaments at the radial sides expose lengths of non-contact along the entire outer contour. Second, due to the finite contour length of filaments, twist leads to “slip” of filament pairs at the ends of bundles. The cohesive energy cost in both cases may be derived by considering the creation of non-contacting filament length by introducing a planar “cut” through a bulk hexagonal array of filaments, as shown in Fig. 3.2(a). The per area exposure of non-contacting filament length is determined by the unit normal \( n \) to the planar cut (the normal of the free surface element) and the local orientation of filament tangents, \( T \). Consider, for example, the loss of contact, i.e. the slip length, \( \ell_s \), for the neighbor pair shown in Fig. 3.2(c), where the tilt of the cut direction is along the neighbor separation. In this case, it is straightforward to relate the length of the surface separating filament ends, \( ds \), to the slip length, \( \ell_s = ds |\sin \Theta| \), where \( |\sin \Theta| = |T \times n| \). In general, summing over the slip of nearest-neighbor contacts yields a surface energy per unit area of the form,

\[
\Sigma = \alpha \frac{\Sigma_0}{2} |T \times n|, \tag{3.3}
\]

where \( \Sigma_0 = \gamma_0/d \), and \( \alpha \) is a numerical coefficient deriving from the orientation of neighbor directions with respect to the surface element. For the low-energy sides of the bundle where filaments are perpendicular to the exposed surface (\( |T \times n| = 1 \)), it is straightforward to show that \( \alpha = 2 \), due to the two fewer neighbors for filaments at the surface relative to the bulk. A more detailed calculation (given in Appendix D) that averages the slip-cost of a cut hexagonal array with respect to all cutting directions yields \( \alpha = 4\sqrt{3}/\pi \approx 2.2 \). For the remainder of the chapter, we neglect the
Figure 3.2. (a) A bulk collection of filaments with a cutting surface. (b) Zoomed in view. Oblique (c), and side (d) views of the surface cut. The orange segments represent the lengths of filaments that are now no longer interacting with the neighbors in front of them. This length, $\ell_s$, is dependent on the angle, $\Theta$, between the surface normal, $\mathbf{n}$, and the filament tangent vector, $\mathbf{T}$. This cut corresponds to an end surface area section, such as the example shown in (e).

variation in relative geometry of neighbor directions and cut directions and simply take $\alpha = 2$ for all bundle surfaces.

For long filaments, the radial sides of the bundle carry most of the surface energy as filaments are normal to the free surface along their lengths and $|\mathbf{T} \times \mathbf{n}| = 1$ is maximal. We define $E_{side}$ as the change in surface energy relative to the untwisted bundle, which we calculate using eqns (3.1) and (3.2),

$$E_{side} = \Sigma_0 A_0 \left( \frac{R/R_0}{\sqrt{1 + (\Omega R)^2}} - 1 \right),$$  \hspace{1cm} (3.4)
where $A_0 = 2\pi R_0 L$ is the side area of the untwisted bundle. Since the height of bundle side, $H(R)$, decreases more rapidly with twist than the lateral growth in radius, $E_{\text{side}}$ is a decreasing function of twist. And since the length of non-contacting filaments at the boundary is fixed to $L$, this change must derive from a change in the number of surface filaments. This demonstrates that a twisted bundle contains a larger proportion of its filaments in the interior than does an untwisted bundle.

While twist reduces $E_{\text{side}}$, this reduction comes at the expense of increasing surface exposure of non-contacting length at the ends of the bundle. We evaluate the surface energy contribution from one of the ends of the bundle, $E_{\text{end}}$, beginning with eqn (3.3). The normal and tangent vectors are defined as

$$n = \frac{\hat{z} + H'/2\hat{\rho}}{\sqrt{1 + (H'/2)^2}}$$

$$T(\rho) = \cos \theta(\rho) \hat{z} + \sin \theta(\rho) \hat{\phi}.$$  

Integration over the surface area of ends, for which $dA = 2\pi d\rho \rho \sqrt{1 + (H'/2)^2}$, and $H' = \partial_\rho H$, yields

$$E_{\text{end}} = \Sigma_0 \int_{\text{end}} dA |T \times n|$$

$$= 2\pi \Sigma_0 \int_0^R \frac{d\rho |\rho|}{\sqrt{1 + (\Omega \rho)^2}} \left( 1 + \frac{(\Omega L/2)^2}{[1 + (\Omega \rho)^2]^2} \right)^{1/2}. \quad (3.7)$$

Analysis of the integrand of eqn (3.7) reveals that $E_{\text{end}}$ has two analytically tractable limits whose form depend on the relative magnitude of $\Omega L$, which is proportional to the number of helical turns of a bundle, and $\sec^2 \theta = 1 + (\Omega R)^2$. In the limit of infinite length (and finite twist) the surface energy per end becomes,

$$\lim_{\Omega L \gg \sec^2 \theta} E_{\text{end}} = \pi L \Omega^{-1} \Sigma_0 [\arcsinh(\tan \theta) - \sin \theta]. \quad (3.8)$$
In this limit, the surface energy of ends derives predominantly from radial slip of neighbor filament pairs extending to different heights, for small twist, \( \ell_s(\text{rad}) \sim d|H'| \sim d\Omega^2 RL \), per pair (see Fig. 3.3(a)). In the opposite limit of vanishing length the surface energy takes the form,

\[
\lim_{\Omega L \ll \sec^2 \theta} E_{\text{end}} = \pi R \Omega^{-1} \Sigma_0 \left[ \cot \theta - \frac{\arcsinh(\tan \theta)\tan \theta}{\tan \theta} \right]
\]  \hspace{1cm} (3.9)

The end surface energy cost in this limit (few helical turns per bundle) is dominated by interfilament slip between azimuthally-separated neighbors, for which \( \ell_s(\text{azi}) \approx |\mathbf{T} \cdot \hat{\phi}|d \sim d|\Omega R| \) at small twist (see Fig. 3.3(b)).

### 3.3 Thermodynamics of exterior surface energy

Notably, the ratio of the surface energy contributions captured in eqns (3.8) and (3.9), which derive from the two distinct modes of slip occurring at the ends of twisted bundles, are consistent with the relative magnitudes of radial vs. azimuthal slip in weakly twisted bundles, \( \ell_s(\text{rad})/\ell_s(\text{azi}) \sim |\theta|(L/R_0) \). This implies that the
aspect ratio of the bundle, $L/R_0$, is a key parameter governing the twist-dependence of surface energy. In Fig. 3.4 we plot the total surface energy, $E_{surf} = E_{side} + 2E_{end}$, as a function of twist angle $\theta$ for aspect ratios ranging from $L/R_0 \to 0$ to the infinite length limit, $L/R_0 \to \infty$.

![Figure 3.4](image)

**Figure 3.4.** Filament bundle surface energy vs twist angle for various aspect ratios of $L/R_0$.

We may assess the quality of the continuum-limit surface energy analysis by direct comparison to the numerical simulations of the discrete filament model. Simulations of bundle cross sections in chapter 2 are carried out in the $L/R_0 \to \infty$ limit so that surface energy changes with twist are derive only from $E_{side}$ and the radial-slip contributions to $E_{ends}$, which are both proportional to $L$. To extract the surface energy of bundle sides, we calculate the excess energy of surface filaments due to fewer filament neighbors than the predominantly six-fold packing in the bulk,

$$
E_{side} = \frac{L}{2} \sum_{i \in b} \left[ \sum_{j \neq i} \gamma(\Delta_{ij}) - 6\gamma(d) \right],
$$

(3.10)
where \( i \in b \) refers to filaments at the surface of the bundle. Fig. 3.5 compares the relative change in surface energy at the sides of small, intermediate and large bundles in our discrete model, to the continuum expression for \( E_{side} \) as eqn (3.10) \(^3\). While the

![Figure 3.5](image)

**Figure 3.5.** External filament cohesive energy vs twist angle for three selected bundle sizes: small \( N = 34 \) (red), medium \( N = 82 \) (blue), and large \( N = 184 \) (green). Black line is the continuum model expression eqn (3.4). Inset shows number of external filaments, \( N_{ext} \) vs twist angle.

circular approximation of the faceted boundary shape for straight bundles leads to an underestimation of the surface energy change, we find that the continuum expression for \( E_{side} \) effectively captures the shape and magnitude of surface energy decrease as the bundle is twisted.

Though not considered explicitly, the discrete model simulations of the previous section do implicitly count the cohesive energy loss due to radial slip at the filament boundaries, deriving from the curvature dependence of the cohesive energy. In the continuum limit where \( \kappa_j d \ll 1 \) we may approximate the curvature-dependent

---

\(^3\)For the discrete model, the value of 6.70\(\gamma_0\) was used in place of the \(6\gamma(d)\) in eqn (3.10), corresponding to the cohesive energy for nearest and next nearest neighbors of a bulk filament in a hexagonal packing of spacing, \(d\).
prefactor in eqn (2.15) as

$$L \left( 1 + \kappa_j \Delta_{ij} \cdot N_j \right)^{-1/2} \simeq L - \ell_s(ij)/2,$$  \hspace{1cm} (3.11)

where $\ell_s(ij) \simeq \kappa_j \Delta_{ij} \cdot N_j = L_{ij} - L_{ji}$ is the difference in contacting length of $i$ with $j$ and contacting length of $j$ with $i$ \(^4\). Note that the implicit loss of contact from radial slip deriving from the curvature dependence does not account for the additional twist-dependent slip between azimuthally separated pairs (e.g. Fig. 3.3b), which enters explicitly into $L_{ij}$. In the following section we generalize our discrete model to include these additional costs. We derive the surface contribution from loss of filament contact at the ends of twisted bundles of infinite length as

$$E_{\text{ends}}(L \rightarrow \infty)/L = \sum_{ij} \gamma(\Delta_{ij}) \left( \frac{1}{\sqrt{1 + \kappa_j \Delta_{ij} \cdot N_j}} - 1 \right),$$  \hspace{1cm} (3.12)

where again we take $i$ to be the outer filament of the pair so that $L_{ij} = L$. Fig. 3.6 compares this surface energy formulation applied to our small, intermediate, and large discrete model bundles, to the continuum model prediction of eqn (3.8). This shows strong agreement over a large range of twist angles as the number of filaments per bundles grows sufficiently large. As described in section 2.6, we calculate the bulk cohesive energy shown in Fig. 2.10, by subtracting the surface contributions given in eqn (3.10) from the total discrete model energy, $E_{\text{bulk}} = E_{\text{tot}} - E_{\text{sides}} - 2E_{\text{ends}}$.

### 3.4 Continuum model of filament bending

Along with the constraints and costs of packing frustration at the bundle core, the additional mechanical cost of filament bending competes with the surface energy prefactor.

\(^4\)This identity derives from the mapping of curve $i$ to the point of contact on $j$, $R_j(s_i) = R_i(s_j) + \Delta_{ij}(s_i)$. Since $d\Delta_{ij}/ds_j \cdot T_j = -\kappa_j (N_j \cdot \Delta_{ij})$, we have $|dR_j/ds_i| = (1 + \kappa_j \Delta_{ij} \cdot N_j)^{-1}$ and $L_{ji} \simeq L_{ij}(1 - \kappa_j \Delta_{ij} \cdot N_j)$.  

53
Figure 3.6. End surface energy for three selected bundle sizes: small $N = 34$ (red), medium $N = 82$ (blue), and large $N = 184$ (green), in the infinite length limit. Black line is the continuum model expression eqn (3.12).

The mechanical cost to bend a straight filament into a helical shape is simply $B\kappa^2 L/2$, where $B$ is the bending modulus, or stiffness, of the filament. In the continuum limit, we compute the total bending energy of the filaments in a twisted bundle by integrating over the cross-sectional area of the bundle

$$E_{bend} = \frac{BL}{2} \int dA \left( \frac{dN}{dA} \right) \kappa(\rho)^2,$$

(3.13)
where $dN/dA$ is the areal density of filaments in the horizontal cross section of the bundle. Following volume-conservation considerations similar to section 3.1, assuming a volume fraction of filaments in the bulk of the bundle, the density at an area element located at $\rho$ is reduced by twist, according to,

$$
\frac{dN}{dA} = \frac{n_0}{\sqrt{1 + (\Omega \rho)^2}},
$$

(3.14)

where $n_0^{-1} = (\sqrt{3}/2)d^2$ is the cross-sectional area per filament in a dense hexagonal packing. Combining both eqns (3.13) and (3.14) to perform our integral in polar coordinates yields,

$$
E_{\text{bend}} = \frac{\pi Bn_0 L}{3} \left( 2 - \frac{2 + 3(\Omega R)^2}{(1 + (\Omega R)^2)^{3/2}} \right). \tag{3.15}
$$

Note that the form of $E_{\text{bend}}$ is not an explicit function of filament number or $R_0$, but instead depends only on twist angle $\theta = \arctan(\Omega R)$. In the limit of weakly twisted bundles, bending energy exhibits a soft dependence on twist, $\lim_{\Omega R \ll 1} E_{\text{bend}} = (\pi Bn_0 L/4)(\Omega R)^4$, while in the limit of large twist, the bending cost asymptotically approaches a constant value $\lim_{\Omega R \gg 1} E_{\text{bend}} = (\pi Bn_0 L/3)[2 - 3/(\Omega R)]$.

We define the total continuum energy as the sum of surface and bending energy contributions

$$
E_{\text{cont}} = E_{\text{surf}} + E_{\text{bend}}, \tag{3.16}
$$

where again $E_{\text{surf}} = E_{\text{side}} + 2E_{\text{ends}}$, as defined by the continuum expressions eqns (3.4) and (3.7). To compare the two types of energy, we define a length scale,

$$
\lambda = n_0 B/\Sigma_0, \tag{3.17}
$$

which parametrizes the relative costs of bending to cohesive energies in filament assemblies. Optimizing $E_{\text{cont}}$ with respect to the twist angle for fixed $R_0$, $L$, and $\lambda$, we compute the diagram of state, shown in Fig. 3.7.
Figure 3.7. Twist diagram of state of for the continuum energy, $E_{cont}$, showing three regions of optimal twist behavior: Untwisted (left of white line), bending energy dominated (upper right “Low Twist” blue region), and cohesive energy dominated (lower right “High Twist” red region). The color represents the preferred value of $\theta$, ranging from $0^\circ$ (blue) to $90^\circ$ (red).

The phase diagram divides into three principle regions: untwisted ($L/R_0 \ll 1$), bending energy dominated ($\lambda/R_0 \gg 1; L/R_0 \gg 1$), and cohesive energy dominated ($\lambda/R_0 \gg 1; L/R_0 \ll 1$). In the untwisted region, twisting a bundle is always unfavorable, due to the combined cost of slip at the ends of short bundles and bending. In the limit of highly flexible filaments, where $\lambda/R_0 \ll 1$, the transition between untwisted and twisted bundles occurs at $L/R_0 \approx 5.07$; and as filament length grows, the balance of side and end surface energy yields an optimal twist that diverges with aspect ratio as $\Omega R \sim (L/R_0)^{1/3}$. For larger stiffness, $\lambda/R_0 \gg 1$, the more significant cost of filament bending shifts the boundary between untwisted and twisted states to larger aspect ratio as $L/R_0 \sim (\lambda/R_0)^{1/2}$. In this bending energy dominated region, there is a significant mechanical cost for bending filaments, however, for sufficiently large
\( L/R_0 \), \( E_{cont} \) is minimized by a modest twist of a few degrees. This arises from the fact that for small twist the cost of bending grows as \( \sim B(\Omega R)^4 \), while the surface energy decreases as \( \sim \Sigma_0(\Omega R)^2 \), leading to an optimal twist of \( (\Omega R) \sim (R_0/\lambda)^{1/2} \). In this regime, the lowest energy state is nearly independent of the bundle length because the bending cost dominates the end effects of the surface energy term. Conversely, the amount of twist in the cohesively-dominated region \( (L/R_0 \gg 1; \lambda/R_0 \ll 1) \) is largely only dependent on \( L/R_0 \). These two regions are separated by a first order transition for bundle lengths beyond a critical size \( L/R_0 \geq 9.9 \). In the infinite length limit, the optimal twist angle jumps from \( \theta \rightarrow 90^\circ \) to \( 14.0^\circ \), at \( \lambda/R_0 = 2.996 \). For lower aspect ratios, this first order transition disappears, and the high and low twist energy minima merge into one. This critical point is shown as the blue dot in Fig. 3.7 at \( L/R_0 = 9.9 \), and \( \lambda/R_0 = 0.425 \).

To summarize, we find that the balance of cohesive energy at the surface of sufficiently long bundles \( (L > 5.07R_0) \) and flexibility favors large bundle twist. For bundles of rigid filaments, surface energy drives a more modest degree of spontaneous twists in minimal energy bundles above a critical aspect ratio that grows with filament stiffness.

### 3.5 Optimal twist of ground-state bundles: Finite stiffness and length

The previous sections have identified two competing effects of twisted filament bundle geometry: packing frustration of filaments in the bulk, and the surface energy cost of non-contacting filaments. In this section, we reexamine the energetics of our discrete filament bundle simulations, including the full costs associated with filament bending and loss of contact in bundles of finite length filaments. As described in section ??, cohesive energy between filament pairs derives from the local contact geometry in twisted bundles. However, for the case of finite length filament bundles,
the contact length of filament $i$ to $j$, $L_{ij}$ used in eqn (2.15), must be calculated explicitly to account for azimuthal slip at the ends of the bundle (see Fig. 3.3b). Defining the ends of filaments to be at positions $s_j \pm L/2$, $L_{ij}$ is calculated for any given length in terms of the contact function defined in section ?? as

$$L_{ij} = s_i(s_j^* = +L/2) - s_i(s_j^* = -L/2),$$

(3.18)

where we follow our original notation that $s_j^*$ is the arc length coordinate of filament $j$ that is the point of contact with $s_i$. It can easily be shown that explicitly determining contact length for a filament pair along with the curvature-dependent correction to cohesive energy, properly accounts for the cost of both azimuthal and radial slip of filaments at the bundle ends.

Evaluating the total energy characterized by an aspect ratio $L/R_0$ and a finite stiffness corresponding to $\lambda/R_0$, we determine the optimal (energy-minimizing) value of twist. The phase boundaries separating untwisted and twisted ground states for small, intermediate, and large bundles are shown in Fig. 3.8. Importantly, we find that the cost of bulk packing, excluded from the continuum analysis of the previous section (see Fig. 3.7), in combination with the bending cost, eliminates the regime of stable, weakly-twisted bundles in the bend-dominated region, $\lambda/R_0 \gg 1$, that is predicted by in the continuum model. The discrete model of cohesive filament bundles exhibits only two well-defined phases: untwisted bundles for sufficiently short or rigid filaments, or highly twisted with a twist angle of $\theta \gtrsim 75^\circ$ for long and flexible filaments. The bundles in this highly twisted state have the maximum topological charge of $Q = 6$.

In the limit of infinitely flexible filaments ($\lambda/R_0 \ll 1$), we find that the inclusion of bulk energy shifts the predicted critical aspect ratio required for twist from $L/R_0 \geq 5.07$, to $L/R_0 \gtrsim 8.9$. In the other limit of infinitely long filaments ($L/R_0 \gg 1$), the critical value of $\lambda/R_0$, above which the filaments are stiff enough to resist the surface
Figure 3.8. Boundaries of the preferred state of filament bundles for three select sizes: small $N = 34$ (red), medium $N = 82$ (blue), and large $N = 184$ (green); overlaid on the continuum model results from Fig. 3.7. Above these boundaries bundles prefer an untwisted state, while below, the bundles can lower their energy by adopting a highly twisted state ($\theta \gtrsim 75^\circ$).

The energy drive to twist, is $\lambda/R_0 \geq 0.63$, $\lambda/R_0 \geq 1.41$, and $\lambda/R_0 \geq 1.78$, for $N = 34$, $N = 82$, and $N = 184$, respectively.

These results show that the additional costs of packing frustration in the bulk significantly offsets the gain in cohesive energy derived from the surface of long bundles. We find that increasing the number of filaments generically increases the range over which minimal energy bundles are twisted, substantially raising the threshold stiffness for the boundary between twisted and untwisted states. As a final comment, we note the appearance of highly-twisted ground states of our discrete simulation model, above the first order line separating highly-twisted from weakly-twisted bundles in the oversimplified continuum model predicted by optimizing $E_{cont}$ alone, which derives specifically from the underestimation of $E_{side}$ (and the driving force for twist) in the continuum approximation (see Fig. 3.7).
3.6 Implications for cohesive filaments assemblies

We now consider how the predictions of our model apply to cohesive assemblies of filaments from a range of systems in materials and biology, whose properties vary in terms of size, stiffness and cohesive interactions. While we opted to use a Lennard-Jones potential to describe interfilament forces in our discrete model, we find ultimately that the predominant thermodynamic sensitivity of bundles to twist depends only on two primary quantities characterizing the interaction: \( \gamma_0 \), the depth of the cohesive interactions (per unit length); and \( d \), the preferred local spacing between bound neighbors. Thus, it is natural to extend the predictions of the current study to filament systems whose finite-range cohesive forces are not explicitly modeled by our “Lennard-Jones thread” model, provided appropriate values of \( \gamma_0 \) and \( d \). Indeed, it is a key finding of the present study that the ground-state packing, characterized in terms of topological charge \( Q \), is entirely insensitive to even these features of the inter-filament potential, either its depth or preferred separation.

Assuming that the assembly kinetics of bundles accommodate the appropriate number and distribution of disclinations, we find that regimes of thermodynamically preferred bundle twist are separated from untwisted bundles by characteristic measures of bundle size. As shown in section 3.5, our discrete model calculations suggest that equilibrium bundles are spontaneously twisted when \( L \gtrsim 10R_0 \) and \( R_0 \gtrsim \lambda \).

Unlike the aspect ratio, which is a purely geometric parameter, \( \lambda \) varies with intrinsic properties of filament stiffness and cohesive forces. We present a brief consideration of the value of \( \lambda \) for three distinct filamentous systems, with the goal of assessing the thermodynamic stability of each to bundle twist: (i) capillary-condensed arrays

---

\[ \text{In principle, the relative cost of the surface exposure to twist-induced frustration is also sensitive to the “stiffness”, or second derivative, of the potential, which controls the elastic properties of the array. The stiffness of the “LJ thread” potential in eqn (2.6), is of order } \sim \gamma_0/d^2, \text{ hence, we expect any system interacting via a similarly soft potential to be well described by the large-N results of the present model.} \]
of micro- and nano-fabricated pillars; (ii) carbon nanotube ropes; and (iii) DNA bundles condensed in the presence of polyvalent counterions. We quantify cohesive tendency for twist in terms of $N_c \equiv (\lambda/d)^2$, roughly the number of filaments needed to stabilize surface-driven twist.

(i) Capillary condensed filaments - On the upper end of filament diameters, we find filament arrays held together by capillary forces, as occurs when filament arrays are drawn from a wetting into to non-wetting fluid medium [102]. In such cases, inter-filament cohesion is mediated by liquid bridges spanning neighboring filaments in the array. As such, we expect the surface energy of the bundle, $\Sigma_0$, to be proportional to the surface tension between wetting and non-wetting fluids, of order $\sim 10 \text{ mN/m}^2$ [103]. Since stiffness is a strong function of filament diameter $B \sim d^4$, bundles of large diameter filaments, such as hair [104] and similarly sized glass or polystyrene fibers [105], with diameters $d \gtrsim 10 \mu\text{m}$ and bending stiffness in the range $B \sim 3 \text{ mN mm}^2$ to $3000 \text{ mN mm}^2$, are relatively stable to twist, only twisting for filament numbers larger than $N_c \approx 3000$. On the other hand, arrays of more slender nano-fabricated pillars $d \approx 300 \text{ nm}$ are relatively easy to twist by capillary forces even for bundles of just a few filaments, as $N_c \approx 1$, consistent with observations of ref. [18].

(ii) Nanotube ropes - On the opposite end of the size spectrum are ropes of single-walled carbon nanotubes, with diameters typically in the range of $d \approx 8 \text{ Å}$ (for (6,6) SWNTs) and $d \approx 27 \text{ Å}$ (for (20,20) SWNTs). Ropes of carbon nanotubes are prepared by a variety of methods, from the electric-arc discharge of graphite [11] to nanotextiles spun from grown nanotube mats [12]. Nanotube ropes are typically formed in the limit of extreme aspect ratio, $L/R_0 \gg 100$. While the influences of tube chirality, metallic/semi-conducting and polydispersity properties of nanotubes complicates the simplistic treatment of inter-filament cohesion considered here, twisted structures have been reported in bundles containing at least tens of the SWNT [13, 14]. Depending on the nanotube diameter estimates for stiffness vary considerably, ranging
from $B \approx 30$ nN nm$^2$ for (6,6) tubes to 4000 nN nm$^2$ for (20,20) tubes, while van der Walls attraction between nanotubes in vacuum suggestion a cohesive energy per length of $\gamma_0 \approx 800$ pN relatively independent of diameter [106, 107], from which we estimate $\lambda \approx 80$ nm and 2 $\mu$m for small and large diameter tubes, respectively. The large value of $\lambda/d$ implies that nanotubes are fairly rigid despite their small diameter, presumably due to intrinsic stiffness afforded by covalent bonding within tubes.

These estimates suggest a very modest tendency for nanotube ropes to twist, which varies considerably with tube diameter: $N_c$ of 5,000 and 500,000 nanotubes for single tube diameters of 8 Å and 27 Å, respectively.

(iii) Condensed DNA bundles - dsDNA condenses in solutions of multivalent counterions into tightly packed toroids [108, 34, 9] and bundles (sometimes referred to in the literature as “rods”) [8, 9, 10]. Given a bending rigidity of $B \approx 0.24$ nN nm$^2$ [109, 110], an interaction energy per unit length of $\gamma_0 \approx 6$ pN, and a center-to-center spacing of condensed DNA, $d \approx 3$ nm [111] in the presence of trivalent cations, we can estimate $\lambda \approx 13$ nm. This sets a critical number of cross-sectional DNA strands to stabilize twist as $N_c \approx 18$. This result implies that the relative flexibility of dsDNA (in comparison to, say, carbon nanotubes) is overwhelmed by inter-strand cohesion in nominally sized-bundles, and cohesive forces alone may be sufficient to stabilize twist in toroidal bundles of dimensions typical for encapsulated bacteriophage genomes [112, 9, 53, 54].

The simple model estimates above neglect many key aspects of inter-filament forces that may further stabilize or inhibit twist in cohesive bundles. Notably, the present model does not account for the twist dependence of interactions between chiral filaments, a feature well-known to bias the handedness and drive the twist of interfilament packings in condensed phases of helical molecules from DNA to collagen [113, 114]. Surprisingly, the broad range of filament sizes and (achiral) cohesive forces considered here suggest that even in the absence of intrinsic or external driving
torques, thermodynamically preferred twist is the rule rather than the exception in cohesive bundles of long and flexible filaments. Furthermore, this feature persists despite the inclusion of defects within the cross-sectional packing of sufficiently twisted bundles.

3.7 Conclusions

In this chapter we constructed a continuum model of twisted bundles composed of finite-length filaments. By ignoring the changes of filament packing in the bulk, we were able to generalize the dependence on twist of the global-scale geometry of a bundle. From this framework we introduced the concept of a surface energy derived from filaments at the exterior possessing less than the ideal number of contacting neighbors. We then proceeded to analyze the thermodynamics of our continuum model, and even included an energetic cost for bending filaments. We find that in the limit of very long filaments \( L \gg R \) and small twist \( \Omega R \ll 1 \), the energy terms resisting twist (bulk frustration and filament bending) both scale as \( + (\Omega R)^4 \), while the side surface energy energy term promoting twist scales as \( - (\Omega R)^2 \). \(^6\) This reveals the surprising result that there is a regime in the available space of parameters \( (\lambda/R_0 \ll 1 \text{ and } L/R_0 \gg 1) \) where filament bundles are unstable to twist.

Thus far we have demonstrated how a bundle’s structure behaves with twist. In chapter 2, the local and interior bulk effects were explored with a discrete model. And in chapter 3, the global and exterior effects were considered utilizing a continuum model. In the next chapter we will, in a way, reverse this question and ask: how does a bundle’s twist—or more generally how does its 3D structure—depend on the filament organization in the cross section?

\(^6\)Strictly speaking, the twist-resisting term of radial slip at the ends also scales as \( + (\Omega R)^2 \), but with a magnitude less than the side surface energy term. Also, the azimuthal slip scales linearly with \(|\Omega R|\), but this is dropped in our prescribed limit of infinite length filaments.
Cohesive filament bundles are a specific class of columnar materials, where large aspect ratio flexible filaments self-assemble into a cable-like structure via attractive interactions. Such examples include biological materials such as collagen [1, 2, 3], and fibrin [4, 5]; as well as artificial materials such as carbon nanotube ropes [11, 12, 13, 14], and micropatterned filament arrays [18].

As a columnar material, they are mechanically unstable to tensions applied transverse to the filament backbone. An example of this deformation is the Helfrich-Hurault instability [28, 92]. Posed as a simple fixed-boundary problem by Selinger and Bruinsma [93]: consider infinitely-long flexible chains, packed in a hexagonal lattice and aligned along the $z$ axis, with two parallel rigid plates bounding the material in the $yz$ plane. A displacement of the plates along the $x$ axis results in an instability of the chains to form longitudinal waves along $z$, as a means to reduces the strain energy. In the limit of infinite spacing between the plates, the critical strain at which this deformation occurs drops to zero. Building upon this concept, in the following chapter we propose a similar instability, but instead of the tensional strain originating from receding rigid plates, it emerges from topological packing defects.

In 2D crystalline materials, the two most basic types of topological defects are dislocations and disclinations. The former are characterized by the insertion/removal of an extra crystalline plane material, while the latter involve the insertion/removal of an extra wedge of material [77]. These defects are linked to unique singular patterns
of strain, and are therefore generally not observed at temperatures well below the melting temperature of the crystalline lattice. This is especially true of disclinations, whose strain energy grows with the area of the lattice [80]. However, both dislocations and disclinations are prevalent in geometrically frustrated materials, where perfect packing cannot be attained. For filament bundles, recent advances in the continuum elasticity theory of ordered filaments, has uncovered an interdependence between defects and a globally twisted filament pattern. This exists for both disclinations [90, 91] and correctly oriented dislocations [96]. A significant finding was that the net numbers of 5-fold disclinations increases with the bundle’s inverse pitch, but is independent of the bundle’s radius. This was justified through an equivalence between bundle twist and a positive (spherical) Gaussian curvature of a 2D membrane [98, 97, 115]. The geometric mapping between bundle twist and membrane curvature arrises from the fact that tilt upsets the in-plane\(^1\) distance between filaments, that instead wish to preserve their distance of closest contact, i.e. the smallest separation between neighboring filaments. This is similar to the effect of Gaussian curvature on the metric spacing between two points on a membrane.

Whereas, flexible filaments in a cohesive bundle may develop a pattern of tilt when topological defects are present, a 2D crystalline lattice is said to ”buckle” under the same condition [78]. The similarities in behavior, and the fact that the curvature of a buckled plate can now be mapped one-to-one to a pattern of filament tilt, will allow us to freely apply the large library of knowledge already documented for curved defective membranes [83, 60, 82, 88, 100, 116, 117, 118]. The most important parameter in determining the buckling behavior of such materials, is the unitless Föppl-von Kármán number [78, 87, 119, 120], here defined as \(\gamma_m = Y R^2 / B_m\), where \(Y\) is the 2D Young’s modulus, \(R\) is the membrane radius, and \(B_m\) is the bending rigidity of the membrane.

\(^1\)The specific plane referenced here, and for the rest of this document, is the one that intersects the bundle of filaments and is perpendicular to the average filament orientations.
Based on the original analysis by Seung and Nelson [78], a membrane containing a disclination, and sufficiently flexible such that $\gamma_m \ll 1$, strongly resist buckling by remaining flat like in Fig. 4.1(a); However, for $\gamma_m \gg 1$, the membrane is highly flexible and therefore unstable to a buckled configuration. These are the cone-like (for a 5-fold), and saddle-like (for a 7-fold disclination) structures shown in Fig. 4.1(b). Furthermore, it can be shown that because the energetic contributions of the bending rigidity, as well as the disclination-induced screening of curvature in the strain energy, both exhibit a lowest-order squared curvature dependence, there exists a critical $\gamma_m^*$, only above which the membrane is unstable to buckling. The value of $\gamma_m^*$ was numerically estimated using a bead-spring model, and is shown in Fig. 4.1 [78].

![Figure 4.1.](image)

**Figure 4.1.** Unbuckled (a), and buckled (b), states of a crystalline membrane containing centered 5-fold and 7-fold disclinations. (c) Plot of membrane energy versus radius, for various values of $\gamma_m \sim R^2 K_0 / \tilde{\kappa}$, for a 5-fold disclination. Note that the energy increases with the radius squared, matching the energy of a flat sheet (limit of $\gamma_m \to 0$), until a particular $R$. This point of turnoff is plotted in (d), showing the critical $\gamma_m^*$ required to buckle the membrane. All images borrowed from Seung and Nelson [78].

Here we propose an equivalent Föppl-von Kármán number for cohesive filament bundles, defined as $\gamma = Y R^2 / K_3$, where $Y$ is the 2D Young’s modulus of interfilament elasticity, $R$ is the radius of the bundle, and $K_3$ is the bending rigidity of the
filaments. Previous work with a continuum elasticity model has already established the optimal amount of twist for a bundle with a centered 5-fold disclination to be \( \theta = \arctan(\sqrt{3 + 32/\gamma^{-1}}) \), where \( \theta \) is the angle of tilt for the outermost filaments with respect to the vertical \( z \) axis [91]. Therefore, unlike the disclination-induced buckling of membranes, a straight bundle is unstable to filament tilt at any value of \( \gamma \). Building upon this knowledge, our current study will develop a linear instability analysis of defect-induced buckling in bundles, with an emphasis on never-before-considered crumpled patterns of deformation, and a focus on the previously unstudied cases of 7-fold and off-centered disclinations.

First, it is worth highlighting some recent work that compares the similarities in geometry between bundles and membranes, in order to obtain some expectations for the defect-induced buckling behavior. Following the recent work of Grason [115], we can model the effects of tilt on a bundle in the limit of infinitesimal filament spacing. We define the squared distance of closest contact between two filaments \( i \) and \( j \), as

\[
\Delta_*^2 = g_{ij}(x)dx_idx_j, \tag{4.1}
\]

where \( g_{ij} \) is a metric tensor that corrects for the discrepancy in distances measured between the cross-sectional plane, and the plane of interfilament contact

\[
g_{ij}(x) = \delta_{ij} - t_i(x)t_j(x). \tag{4.2}
\]

Eqn (4.1) is identical to the description of the metric geometry of 2D surfaces. The unique case of \( g_{ij} = \delta_{ij} \), corresponds to a straight bundle or a flat (zero Gaussian curvature) surface. In the limit of small deviations away from this state, we uncover a unique conversion between patterns of tilt, and an equivalent Gaussian curvature.
Logically following from this result, if 5-fold disclinations in flexible membranes promote positive Gaussian curvature, then $K_{eq}$ can be made positive for bundles by engaging the first two terms in eqn (4.3); specifically, by possessing a twisted texture. Alternatively, 7-fold disclinations promote negative curvature, which can be easily accessed with a pattern of splay, producing a non-zero value in the third term.

The general goal of the following chapter is to study how the 2D cross-sectional packing of filaments effects the 3D structure of the bundle. Specifically, how does the type and location of defects within the crystalline packing lead to twisting, undulations, writhing, bending, or other such modes of deformation. In section 4.1, we describe a fully 3D discrete model of cohesive filament bundles. Then in section 4.2, we introduce a comparable continuum elasticity model. Using these two models, we will examine the various buckling instabilities for centered 5-fold disclinations in section 4.3, with a nod to the comparable behavior of 2D crystalline membranes. In section 4.4, we investigate a newly discovered mode of deformation, termed "torsional crumpling", available to sufficiently long and flexible bundles with 5-fold disclinations. Next, in section 4.5, we perform a similar analysis, but for 7-fold disclinations, and explain how the incompatibility of radial splay with patterns of tilt along the $z$ axis result in qualitatively different behavior from their 5-fold cousins. Finally, in section 4.6, we use our coarse-grained model to analyze bundles with off-centered defects, which give rise writhing structures.

### 4.1 Fixed-lattice model of discrete 3D filaments

Here we introduce a coarse-grained bead-spring model for interacting filaments. The end goal is a working model of flexible and cohesive “featureless” tubes that
incur minimal cost for sliding along their lengths. A bundle contains $N_f$ filaments, indexed with the letters $i$ and $j$, with each filament containing $N_b$ beads, indexed by the letter $n$. Bead positions along a single filament are located at $x_n$, and we define $\ell_n$ as the length of the line segment between beads $n$ and $n + 1$. From here the tangent at bead $n$ is defined as $\hat{T}_n = (x_{n+1} - x_n)/\ell_n$. This is schematically shown in Fig. 4.2. From this we define a cost for filament bending as

$$E_b^{(i)} = B \sum_{n=1}^{N_b-1} \frac{1 - \hat{T}_n \cdot \hat{T}_{n+1}}{\ell_n}. \quad (4.4)$$

Interactions between neighboring filaments are modeled as generic Hookean springs,
\[ E_{c}^{(i,j)} = \frac{\epsilon}{2} \sum_{n=1}^{N_b} (\Delta_{n,j} - \Delta_0)^2, \tag{4.5} \]

where \(\Delta_{n,j}\) represents the distance of closest contact from bead \(n\) on filament \(i\), to a point along the backbone of the neighboring filament \(j\). By definition, this distance intersects filament \(j\) at a right angle. Because our filaments are composed of line segments anchored to jointed beads, our distance of closest contact is calculated not between beads, but rather between a bead and a neighboring segments (represented as the yellow line between filaments, in Fig. 4.2). Although eqn (4.5) is a decidedly simplified representation of filament interactions, it is still enough to capture the qualitative features we wish to describe, and its generic form easily lends itself to more complex models down the line, that may be specifically tuned to individual materials.

Assembling eqns (4.4) and (4.5), the final energy of our bead-spring model is

\[ E_{\text{total}} = \sum_{i=1}^{N_f} E_b^{(i)} + \frac{1}{2} \sum_{i=1}^{N_f} \sum_{\langle i,j \rangle} E_{c}^{(i,j)}, \tag{4.6} \]

where the third sum is over all the nearest neighbor filaments \(j\), to filament \(i\).

This free energy is used in subsequent sections to determine the low energy structures of filamentous bundles. Energy minimization simulations are performed by starting with an initial state (generally straight filaments aligned along the \(z\) direction) consisting of up to \(N_F = 400\) filaments, and up to \(N_b = 800\) beads per filament. During minimization, the \(z\) coordinates of beads are fixed as a means to relieve the stretching at bundle ends caused by slipping. Although strictly speaking, the volume of the bundle and the length of the filaments are no longer conserved, this model is reminiscent of biological materials that self-assemble into equilibrium structures by adjusting length and radius simultaneously, such as collagen [1]. Qualitatively, the results we find are generic to all buckled filament bundles, and not specific to this
particular method, but also found when the length of filaments are fixed. The most significant difference arises at the ends of the bundle, where filaments slip past each other when tilting. However these end effects lessen as $L/R \to \infty$.

Naturally, a perfect hexagonal lattice of straight filaments is already in its mechanical equilibrium ground state, with $E_{\text{total}} = 0$. However, when topological disclinations are present in the cross-sectional packing, the distance of closest contact between filaments can no longer be $\Delta_0$ everywhere. As we will see, filaments must tilt to minimize their cohesive energy, leading to a competition between the cohesive and bending energy terms.

### 4.2 Continuum elasticity of filament bundles

Now we'll employ a continuum elasticity model to analyze the stability of longitudinally periodic and axisymmetric deformation patterns of bundles of radius $R$, with a centered disclination of charge $s$. A general model includes two types of elasticity,

$$E = \int (f_{el} + f_{Frank})dV. \quad (4.7)$$

The first term describes the elastic strain energy density of columnar materials

$$f_{el} = \frac{1}{2}u_{ij}\sigma_{ij}, \quad (4.8)$$

where the stress and strain are respectively defined as

$$\sigma_{ij} = df_{el}/du_{ij} = \lambda u_{kk}\delta_{ij} + 2\mu u_{ij} \quad (4.9)$$

$$u_{ij} = \frac{1}{2}(\partial_i u_j + \partial_j u_i - \tau_it_j). \quad (4.10)$$
The second term in eqn (4.7), describes the Frank elastic energy cost for non-uniform orientations of the filaments

\[ f_{Frank} = \frac{1}{2} \left( K_1 (\nabla \cdot \hat{t})^2 + K_2 [\hat{t} \cdot (\nabla \times \hat{t})]^2 + K_3 [(\hat{t} \cdot \nabla) \hat{t}]^2 \right), \]  

where filament orientation is derived from the 2D displacement field, \( u(x) \)

\[ \hat{t} = \frac{\hat{z} + \partial_z u}{\sqrt{1 + |\partial_z u|^2}} \simeq \hat{z} + t, \]  

and \( t \) refers to only the in-plane component of orientation. Here we have assumed only small in-plane tilt, \( t \simeq \partial_z u \), and expanded the energy to lowest nontrivial order.

To determine the equations of equilibrium, we begin with an initial displacement field, \( u(x) \), subject to a small perturbation, \( \delta u(x) \), and consider the variation of the energy, \( \delta E = E[u(x) + \delta u(x)] - E[u(x)] \). We specialize to the case of vanishing elastic constants for splay, \( K_1 = 0 \), and twist, \( K_2 = 0 \), as in-plane elasticity in columnar materials suppress these modes at long wavelength, and because there is no equivalent term in the 3D discrete model, eqn (4.6). Solving for the equations of equilibrium, we arrive at

\[ \partial_j \sigma_{ij} - \partial_z (t_j \sigma_{ij}) - K_3 \partial_z^2 t_i = 0 \quad \text{(force balance)} \]  
\[ dS_i \sigma_{ij} = 0 \quad \text{(stress free sides)} \]  
\[ t_j \sigma_{ij} + K_3 \partial_z^2 t_i = 0 \quad \text{(stress free ends)} \]  
\[ K_3 \partial_z t = 0 \quad \text{(torque free ends).} \]

Similar to the Föppl-von Kármán number previous attributed to flexible membranes, we can define an equivalent measure of the ratio of the cost of cohesion to bending

\[ \gamma = \frac{YR^2}{K_3}. \]
For $\gamma \gg 1$, interfilament elasticity dominates, leading to easily buckled bundles; alternatively, for $\gamma \ll 1$, the cost of bending filaments dominates, resulting in predominantly straight filaments. A full derivation of the conversion between the parameters in this continuum elasticity model and the 3D discrete model, appears in appendix E.

To analyze the instability behavior in the presence of defects, we consider a reference state of a parallel bundle consisting of straight filaments with $t = 0$. Rather than solve for displacement, we determine only the equilibrium stress generated by a centered disclinations of charge $s$, at $r = 0$, by supplementing force balance with the compatibility condition,

$$Y^{-1}\nabla_\perp^2 \sigma_{ii} = s\delta^2(x_\perp),$$

(4.18)

where $Y = 4\mu(\mu + \lambda)/(2\mu + \lambda)$, is the 2D Young’s modulus. Eqn (4.18), when combined with the condition for vanishing normal stress at the bundle boundary, has the solution

$$\sigma^0_{rr} = \frac{Ys}{4\pi} \ln(r/R); \quad \sigma^0_{\theta\theta} = \frac{Ys}{4\pi} [\ln(r/R) + 1],$$

(4.19)

where $\sigma^0_{ij}$ denotes the stress of the parallel reference state with the centered defect.

We determine the conditions for the solutions to the stability equations outlined in eqns (4.13)-(4.16), by considering solutions that are weakly perturbed from the parallel state, and of the form

$$u(x) = u_0(x) + \epsilon u_1(x) + \epsilon^2 u_2(x) + \epsilon^3 u_3(x) + \ldots,$$

(4.20)

where $\epsilon$ is the amplitude of the deformation, assumed to be arbitrarily small near the point of linear instability (i.e. the supercritical bifurcation point), and $u_n$ represents the $O(\epsilon^n)$ deformation modes. For linear stability, it is sufficient to analyze only the lowest order in $\epsilon$, though if we want to solve for the dependence on the $\epsilon$ distance from the instability, we need to solve to order $\epsilon^3$. 

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Considering only the order $\epsilon^1$ term, and decomposing the displacement into radial and azimuthal components

$$\mathbf{u}_1(\mathbf{x}) = \rho(\mathbf{x})\hat{r} + \tau(\mathbf{x})\hat{\theta}. \quad (4.21)$$

Applying this form to eqn (4.13), and assuming only axisymmetric patterns of deformation, we find the force balance along the $\hat{r}$ direction

$$(\lambda + 2\mu)\partial[r^{-1}\partial_r(r\rho)] - \sigma_{rr}^0\partial_z^2\rho - K_3\partial_z^4\rho = 0, \quad (4.22)$$

and along the $\hat{\theta}$ direction

$$\mu\partial[r^{-1}\partial_r(r\tau)] - \sigma_{r\theta}^0\partial_z^2\tau - K_3\partial_z^4\tau = 0. \quad (4.23)$$

The boundary conditions on the sides of the bundle are simply $\sigma_{rr}^1 = \sigma_{r\theta}^1 = 0$, or specifically

$$\lambda\rho(R)/R = (\lambda + 2\mu)\partial_r\rho(R) = 0 \quad (4.24)$$

$$\partial_r\tau(R) - \tau(R)/R = 0. \quad (4.25)$$

And finally, we have the boundary conditions for the derivatives of the displacements at the ends of the bundles, but we will neglect these by assuming that solutions are periodic, and of the form

$$\rho(\mathbf{x}) = \rho(r)\cos(kz); \quad \tau(\mathbf{x}) = \tau(r)\cos(kz). \quad (4.26)$$

To compare to a finite length bundle, we might consider wavelengths that are commensurate with the bundle length, $k = 2\pi n/L$, though to be clear, these purely
sinusoidal deformations will not allow us to match the free end boundary conditions from eqns (4.15) and (4.16). Presumably, a boundary layer is required to match the purely periodic solutions we consider, to the free-end calculations. Therefore, we work under the assumption that the length scale of this boundary layer will vanish proportional to $\sqrt{K_3/Y}$, and hence can be ignored for large aspect ratio bundles ($L/R \gg 1$), and large bundle Föppl-von Kármán number ($\gamma \gg 1$).

To proceed, we rewrite the equations in dimensionless variables, by measuring all lengths in units of bundle width, $R$, and stresses in units of $Y$. Doing this, and recalling the definition of the 2D Poisson ratio, $\nu = \lambda/(\lambda + 2\mu)$, we rewrite eqns (4.22) and (4.23) as

$$-\frac{1}{2}[r^{-1}\partial_r(r\rho)] + V_\rho(r)\rho(r) = -\alpha_\rho\rho(r)$$

(4.27)

$$-\frac{1}{2}[r^{-1}\partial_r(r\tau)] + V_\tau(r)\tau(r) = -\alpha_\tau\tau(r),$$

(4.28)

using the definitions

$$V_\rho(r) = -\frac{s(1 - \nu^2)}{8\pi} \ln r$$

(4.29)

$$V_\tau(r) = -\frac{s(1 + \nu)}{4\pi}(\ln r + 1)$$

(4.30)

$$\alpha_\rho = \frac{(1 - \nu^2)k^4}{2\gamma}$$

(4.31)

$$\alpha_\tau = \frac{(1 + \nu)k^4}{\gamma}.$$  

(4.32)

In this way, we have recast the linear stability calculation in terms of an eigenvalue problem, with the boundary conditions

$$\nu\rho(1) + \partial_r\rho(1) = 0$$

(4.33)

$$\partial_r\tau(1) - \tau(1) = 0.$$  

(4.34)

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We are interested in the ground state solution, i.e. the smallest values of $\alpha_\rho$ or $\alpha_\tau$ that are consistent with our boundary conditions. This will correspond to the first instability, the lowest value of $\gamma$, at which a given periodic mode, $k$, becomes unstable.

In the next couple sections, we will solve these equations for the most unstable wavenumber, $k$, given centered 5-fold ($s = +2\pi/6$) and 7-fold ($s = -2\pi/6$) disclinations. We will find that the former gives rise to azimuthal patterns of filament tilt that result in a twisted bundle texture, while the latter yields radial patterns of filament splay.

### 4.3 Homogeneously twisted bundles with 5-fold disclinations

In the following sections, we will derive the available modes of deformation for bundles containing various types and locations of defects, by performing energy minimization simulations on bundles represented by our bead-spring model from eqns (4.4) to (4.6). To begin, will first consider bundles containing a single centered 5-fold disclination. The initial state consists of a cylindrical bundles of $N_f = 306$ ($R = 10\Delta_0$), aspect ratios of $L/R = 4$ and 8, and bead spacing along a filament length of $\ell_0 = 0.2\Delta_0$. The spring constant between filaments, $\epsilon$, was fixed, and the bundle-equivalent Föppl-von Kármán number, $\gamma$, was varied between 0.25 and 25,000, by varying the filament rigidity, $B$, in 50 steps. A minimum energy state was found for each value of $\gamma$ using the GSL conjugate gradient package for C [121].

Results are shown in Fig. 4.3, where we plot the mean angle of the outermost filaments with the $z$ axis (henceforth called the *twist angle*, $\theta$), versus $\gamma$. We define $\theta$ as the angle of the outermost filaments with the central $z$ axis. Because $\theta$ may vary between boundary filaments, and even along the length of a single filament, we report the values as $\langle \theta \rangle$, defined as the twist angle averaged over the total length of all outer filaments. These results verify that bundles with 5-fold disclinations are always unstable to twist, even when $\gamma \ll 1$, a results established by Grason [91, 90], using a
Figure 4.3. Energy minimization simulation results for the bead-spring model of a bundle containing a single centered 5-fold disclination. The measured twist angle, $\theta$ is defined in the furthest right example. Blue and purple data points are for $L/R = 4$ and 8, respectively. Example structures are shown for $L/R = 4$ bundles, with select outer filaments colored orange to help highlight the structure. The solid black line is the continuum elasticity result derived by Grason [91].

continuum model that was summarized in section 1.4. The black line in Fig. 4.3, is eqn (1.12), where we have used the fact that $\Omega R = \tan \theta$. This result is in antithesis of defect-induced buckling of 2D membranes, which are only unstable to buckling above a critical $\gamma_m \gtrsim 120$ [78]. A feature that can be attributed to the bending energy of a membrane having a greater contribution to total energy than that of the bending of filaments.

Although the physics of buckling defective membranes and bundles are distinct, their geometry can be made equivalent. Tilting filaments in a bundle effectively alters their spacing by modifying their distance of closest contact. A similar effect is commonly attributed to Gaussian curvature for particles packed on a 2D surface
As originally described in section 2.5, although it is possible to always calculate the equivalent Gaussian curvature for a given pattern of filament tilt, it is not always possible to analytically solve for an exact equivalent surface itself. That is to say, there is not always a unique surface for every pattern of Gaussian curvature (e.g. one can smoothly transition between the two distinct minimal surfaces of a catenoid and helicoid while continuously preserving the Gaussian curvature). However, in the case of a bundle of homogeneous pitch for all filaments, things are simplified by recognizing that every slice of the bundle perpendicular to the centerline z axis is identical up to a rigid rotation. This texture can be assumed because it only generates small tilt-induced strains caused by applying a rigid rotation around the z axis. With the additional assumption of an axisymmetric surface, one is able to solve for the *bundle-equivalent dome* as an exact one-to-one mapping of twist onto curvature, using eqn (4.3) \[98, 97\]. The resulting conversion allows us to transform from curvature to twist, using

\[
K_G(r) = \frac{3\Omega^2}{(1 + \Omega^2 r^2)^2},
\]

where $\Omega = \frac{2\pi}{\text{pitch}}$ is the rate of bundle twist, and $r$ is the radial position of a filament within the bundle.

Up until now, no method has been developed to perform these sorts of mappings for discretely modeled bundles. Here we introduce a formal method for mapping between filament tilt and curvature for our coarse-grained bundles, and compare it to what is already known for the continuum model.

For an arbitrary discrete 2D surface composed of vertices, edges, and triangular faces, the Gaussian curvature can be defined at a single vertex point as

\[
K_G = 3 \left( 2\pi - \sum_{\alpha=1}^{\#f} \psi_\alpha \right) / A,
\]

where $\#f$ is the number of faces at each vertex.
where $\psi_\alpha$ is the internal vertex angle for face $\alpha$, $A$ is the summed area of all the faces attributed to the vertex, the factor of 3 accounts for a single face belonging to three vertices, and the sum is over all faces that contain the vertex point [125, 126]. For a bundle, we can generate a mesh of vertices by performing a Delaunay triangulation over the points of intersection of all filaments with an intersecting plane, here chosen to be the $z$ plane. This mesh is itself flat, with zero Gaussian curvature, but remember that the true spacing between filaments is the distance of closest contact, which may lay out of the plane. Explicitly, if the in-plane spacing between two filaments, $i$ and $j$, is $\vec{\ell} = \vec{x}_j - \vec{x}_i$, then the out of plane distance of closest contact from filament $i$ to filament $j$, is defined as

$$\vec{\ell}_* = \vec{\ell} - t_j(\vec{\ell} \cdot \vec{t}_j),$$

(4.37)

and shown in Fig. 4.4 Therefore, we measure each angle, $\psi_\alpha$, for a given triangle, in a manner that accounts for this modification. This is done by using the standard SSS theorem for a triangle composed of sides $a$, $b$, and $c$, and solving for the angle opposite of side $a$,

$$\psi_c = \arccos \left( \frac{b^2_* + c^2_* - a^2_*}{2b_*c_*} \right),$$

(4.38)

where the subscript, $\ast$, designates that side lengths obey eqn (4.37). In essence, this method works by distorting the dimensions of every triangle based on the relevant dis-
tances of closest contact, then stitches them back together in a manner that preserves the original network topology, but requires out-of-plane orientations.

The application of this mapping, is applied to the previously discussed results of 5-fold disclinations, and shown in Fig. 4.5. The equivalent Gaussian curvature increases

**Figure 4.5.** The top row of images shows the equivalent Gaussian curvature for middle cross-sectional slices of the four example bundles highlighted in Fig. 4.3. The discrete hexagonal regions correspond to Voronoi tessellation of filament intersections with the plane. The color corresponds to the equivalent Gaussian curvature for each filament, calculated using eqns (4.36) to (4.38). The middle row of images shows a reconstruction of the mapped surface. The bottom row of images shows the Gaussian curvature vs a filament’s radial position. Data points are for individual filaments, while the solid curve is derived from eqn (4.35).

with $\gamma$, as the filaments become flexible enough to allow the bundle to develop into a more twisted texture. For low values of $\gamma$, $K_G$ is nearly uniform throughout the entire cross section. This agrees with previous results that show that a lightly twisted bundle with homogeneous pitch, $P$, can be mapped onto an axisymmetric surface that is approximated by a sphere with a radius of $2\pi P/\sqrt{3}$ [98]. For larger values of $\gamma$, $K_G$ continues to increase, and is focused near the centered 5-fold disclination, which is accounted for by the bundle-equivalent dome asymptotically approaching a cylinder.
with zero Gaussian curvature for large $\theta$. This method of calculating $K_G$, is also used in the following sections to understand the buckling behavior of 7-fold disclinations, as well as off-centered 5-fold disclinations.

The bottom row of images in Fig. 4.5, show a full reconstruction of the Gaussian curvature into a complete surface. This structure is generated by first introducing a 2D array of vertices (representing filaments) that share the network topology of the bundle, where the bonds between neighboring vertices are treated as springs, each with a unique preferred spacing that is equal to the measured distance of closest contact from eqn (4.37). The shape is then numerically relaxed in energy using the method of steepest descent, until the stretching energy is sufficiently close to zero. During this process, the dissimilar spring lengths, derived from the dissimilar distances of closest contact, promote a buckled form. The Gaussian curvature of the final structure measured with eqn (4.36), is identical to the Gaussian curvature measured directly from the bundle itself. By comparing Fig. 4.5 to Fig. 4.1(b), it is very clear to see the similarities between the 5-fold disclination-induced buckling of a bundle and a membrane.

### 4.4 Torsional crumpling for 5-fold disclinations

In addition to the homogeneous pitch structures described above for bundles with 5-fold disclinations, we have discovered metastable states that become accessible for large values of $\gamma$. This new class of buckling, we term *torsional crumpling*, is characterized by an alternating direction of twist along the $z$ axis. Example structures for $L/R = 4$, are shown in Fig. 4.6. These energy minimization simulation results consist of a series of steps: 1) begin with an initial crumpled texture with a maximally large value of $\gamma$, and an applied azimuthal displacement pattern corresponding to the desired value of $n$; 2) perform an energy minimization relaxation of the structure; 3) The resulting structure’s twist is plotted in Fig. 4.6, and $\gamma$ is decreased in a stepwise
Figure 4.6. Torsional crumpling instability for bundles containing a centered 5-fold disclination. The various modes are characterized by $n$, which counts the number of times the direction of twist alternates. The $n = 0$ structure is the homogeneous pitch data from the previous section. The lower row of figures correspond to the points of instability, $\bar{\gamma}_*$ (large data points). For $\gamma < \bar{\gamma}_*$, filaments are too stiff to retain the original number of twist alternations (a.k.a. torsional crumples), and instead progress to a lower $n$ configuration, in which case they are no longer plotted for ease of viewing.

fashion; 4) repeat steps 2 and 3, until the minimum desired $\gamma$ is reached. During the course of this routine, eventually $\gamma$ becomes too low for the initial non-zero $n$ to remain stable, and a crumple (e.g. vertical section of a bundle over which the direction of twist alternates) is “ejected” from one of the ends. The Föppl-von Kármán number at which this occurs is designated as $\bar{\gamma}_*$.

Ideally, the ground energy configuration for all values of $\gamma$ is the homogeneous pitch $n = 0$ state, where the azimuthal filament tilt pattern best screens the strain of the defect. Along a crumpled region the twist alternates its direction, during which
the filaments necessarily become locally straight and unable to screen the defect’s strain. Therefore, the strain energy drives the crumple towards an infinitesimally small length in the $z$ direction. Alternatively, the bending energy, though not exactly zero in the $n = 0$ state, is still minimal compared to crumpled states. Therefore, the wavelength of these crumplings, measured to be $\lambda_m$, is set by a competition between these two energies. A natural length scale is already found in the ratio of the cost of bending filaments to the cost of interfilament stretching

$$\lambda = \sqrt{K_3 / Y}.$$  \hfill (4.39)

Therefore, it is easy to surmise that $\lambda_m \sim \lambda$. Fig. 4.7, plots these two length scales together, with a near linear trend strongly suggesting that this is indeed the case. The slope of the best fit (dashed line), yields the remaining numerical constant to be $\lambda_m \approx 2.145\lambda$.

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure4.7.png}
\caption{Plot of $\lambda$, defined in eqn (4.39), versus the measured wavelength of the crumple patterns, $\lambda_m$. Points correspond to the points of instability, $\tilde{\gamma}_s$, from Fig. 4.6. The dashed line is the best linear fit, with a slope of 2.415.}
\end{figure}
At the ends of the bundle, filaments are unable to support the energetically high cost of bending associated with these crumplings. This effectively drives crumplings inward into the bulk. When \( n\lambda_m \gtrsim L \), the crumplings begin to occupy the ends of the bundle, but the free end’s inability to contain them result in their ejection, and therefore a new lower \( n \). To reiterate, this transition occurs at \( \bar{\gamma}_* \).

We can analyze the linear stability of the crumpled states by building upon the continuum elasticity model, laid out in section 4.2. Again, we are considering periodic displacement perturbations away from the reference state of a straight infinite bundle, of the form eqn (4.26), and that obey eqns (4.27) to (4.34). From this foundation, we will determine the relationship between mode \( k \) and the critical \( \gamma_c(k) \), above which, the bundle becomes unstable to torsional crumpling of mode \( k \). From eqn (4.3), we can already see that a twisting is a good guess for the tilt-induced displacement when \( s = +2\pi/6 \), strongly suggesting \( \rho(r) = 0 \), and \( \tau(r) \neq 0 \). Therefore, we begin by making an additional coordinate transformation to convert the second-order linear of eqn (4.28), into a first order, non-linear ODE, which is more practical to solve

\[
\tau(r) = \Omega_0 r \exp \left[ \int_0^r dr' f(r') \right],
\]

(4.40)

where \( \Omega_0 \) is an arbitrary constant. In this parameterization, \( f(r) \) describes the deviation from rigid rotations in the plane, \( r\partial_r(\tau/r) = f(r)\tau \), allowing us to transform eqn (4.28) into

\[
-\frac{1}{2}(f^2 + f' + 3f/r) - \Gamma_\tau (\ln r + 1) = -\alpha_\tau,
\]

(4.41)

where

\[
\Gamma_\tau \equiv \frac{sk^2(1 + \nu)}{4\pi} > 0,
\]

(4.42)

parameterizes the coupling strength to the logarithmic potential. This equation must be solved subject to the boundary condition \( f(1) = 0 \).
Practically speaking, eqn (4.41) can be solved numerically via a simple shooting method [127]: For a given guess of $\alpha_\tau$, shoot from $f(0) = 0$ at $r = 0$ and integrate eqn (4.41) to the boundary; then numerically adjust $\alpha_\tau$ until $f(1) = 0$ is satisfied. The solution for $\alpha_\tau(\Gamma_\tau)$, is plotted in Fig. 4.8(a). We can then compute the critical

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure4.8.png}
\caption{(a) The dependence of the torsion eigenvalue, $\alpha_\tau$, on the effective coupling to the stress-induced "potential", $\Gamma_\tau$, showing a largely linear dependence in both small and large $\Gamma_\tau$ regimes. (b) The stability map of torsional crumples (for $\nu = 1/3$) in terms of the wave vector, $k$, and $\gamma$. Above this line, bundles are unstable to torsional wrinkling. Note that that the stability range of parallel bundles decreases to zero in the long-wavelength limit of $k \to 0$, i.e. homogeneous pitch.}
\end{figure}

Föppl-von Kármán number for a given $k$, above which, the bundle is unstable to torsional crumpling.

$$\gamma_c(k) = \frac{(1 + \nu)k^4}{\alpha_\tau[sk^2(1 + \nu)/4\pi]}.$$ (4.43)

To compare this result to our bead-spring model, we first follow the reasoning of Seung and Nelson [78], to find that the Poisson ratio of a triangular lattice of atoms bonded by Hookean springs is $\nu = 1/3$. Using this value, the final result (and returning $k$ back to its non-unitless form) is shown in Fig. 4.8(b).

We can compare these results to the crumpled bundle structures found in our coarse-grained model, by considering the special values of $\gamma$, at which various transitions occur. While $\gamma_c(k)$ measures the point at which the straight bundle becomes
unstable to a torsional crumpling texture of wave vector \( k \), there also exists a critical \( \gamma \) at which an already crumpled bundle of mode \( k \) becomes unstable to a lower \( k \). In the coarse-grained model, this point is designated as \( \bar{\gamma}_s \), and is a sort of ”upper bound” on \( \gamma_c \). It is difficult to measure \( \gamma_c \) directly in the coarse-grained model because a straight bundle may be unstable to multiple modes of crumpling simultaneously, making it impractical to isolate just one mode. However, it is possible to estimate this value by assuming that the instability occurs at a supercritical bifurcation point, where the twist angle obeys the form

\[
\theta \approx \theta_0 \sqrt{1 + \frac{\zeta}{(\gamma - \gamma_c)}},
\]

(4.44)
in the region of \( \gamma \approx \gamma_c \). In this equation, \( \theta_0 \) is the maximum twist angle far from the transition point (30° for a 5-fold disclination), and \( \zeta \) is a value that regulates the speed of the transition. Even though the structures from the coarse-grained model are far from transition, we can assume they still obey the form of eqn (4.44). Given the simple conversion of \( \pi n/L \approx 2\pi/\lambda_m = k \), a best numerical fit of the data for the \( n = 3 \) solution is shown in Fig. 4.9(a). From this fit, and the subsequent fit for all \( n \), we can estimate a reasonable value for the critical Föppl-von Kármán number, and label it \( \bar{\gamma}_c \). A final comparison of the two models is made in Fig. 4.9(b), where we overlay the linear stability result for \( \gamma_c \), from Fig. 4.8, with the upper and lower bounding estimates of \( \bar{\gamma}_s \) and \( \bar{\gamma}_c \), showing reasonable agreement.

We will now consider the functional advantages of these metastable crumpled states. Returning to our comparison to the model system of a buckled crystalline membrane, there is an intriguing behavior that previously had no known analogy in bundles. This is the ability of the membrane to ”focus” the curvature to the disclination when \( \gamma_m \gg 1 \). At low \( \gamma_m \), the initial instability (for a disclinations of \( s > 0 \)) transforms the flat surface into one with with (positive spherical) Gaussian curvature that is nearly equal everywhere, so as to minimize the cost of bending.
Figure 4.9. (a) Plot of mean twist angle, $\theta$, versus $\gamma$, for $n = 3$ from Fig. 4.6. Two values of $\gamma$ are labeled: $\bar{\gamma}_*$, below which the bundle becomes unstable to a lower value of $n$; and $\bar{\gamma}_c$, a fitted-line estimation of $\gamma_c$. (b) The values of $\bar{\gamma}_*$ and $\bar{\gamma}_c$, for multiple crumpled textures. These provided upper and lower bound estimates of $\gamma_c$ from eqn (4.43).

However, as $\gamma_m$ is increased beyond the critical point, the cost of bending approaches zero, and the membrane adopts a sharper (cone-like) configuration [78]. In the limit of $\gamma_m \to \infty$, all of the Gaussian curvature is focused to a single point. A similar effect is seen in simulations of virus capsids, where a shell continuously transforms from a sphere for small $\gamma_m \ll 1$, to a faceted icosahedron for large $\gamma_m \gg 1$ [87].

Paradoxically, the three dimensional nature of bundles strongly resists any equivalent behavior as a result of the strong drive to retain homogeneous pitch for all filaments.\(^2\) This is even despite the ability to translate twist into Gaussian curvature, as covered in section 4.3. Any deviation from a homogeneous pitch motif systematically destroys the full-length contact between filaments that exist at different radii, $r$. Now consider torsionally crumpled bundles, we find that these unique textures are

\(^2\)In this analogy, membrane Gaussian curvature is equivalent to gradients in the filament tilt pattern.
able to attain a compromise, and allow for twist gradient-focusing as $\gamma \to \infty$. This is a consequence of the ability of a torsional crumple to "reset" the filaments back to the homogeneous pitch pattern, within its longitudinal zone of influence. Because both the wavelength of the crumplings, $\lambda$, and the relative cost of filament bending, both approach zero as $\gamma \to \infty$, this suggests that crumpled bundles may be able to achieve a tilt gradient-focused (i.e. Gaussian curvature-focused) state, similar to that found in crystalline membranes.

To show this, in Fig. 4.10, we have plotted the equivalent Gaussian curvature for individual filaments located at a distance, $r$, away from the center of the bundle, for $n = 0, 2, 4, \text{and } 6$. Note that the total integrated Gaussian curvature is nearly identical for every $n$. However it is clear that the distribution of the curvature depends greatly on $n$. This reveals that the equivalent curvature of the mapped surface (i.e. gradients in the tilt pattern), becomes more focused towards the defect location, as

![Figure 4.10](image-url)

**Figure 4.10.** (a) Equivalent Gaussian curvature for individual filaments located at a distance $r$ away from the center of the bundle, in the central cross section. Results are for bundles of aspect ratio, $L/R = 4$, $\gamma = 25,000$, and $n = 0, 2, 4, \text{and } 6$. (b) Plot of the total energy versus $\gamma$ for various numbers of crumplings, $n = 0$ to 6. (c) Plot of just the cohesive energy in the range of high $\gamma$, showing that $n \neq 0$ bundles are able to overtake their lower $n$ counterparts for larger values of $\gamma$. (d) The reconstructed surfaces for $n = 0$ and $n = 6$, showing the impact of curvature focusing at the top of the dome (i.e. the location of the defect).
both $\gamma$ and $n$ increase. This phenomenon of curvature focusing is exactly what is observed in the analogous 2D crystalline membranes, but is not geometrically attainable for homogeneous pitch bundles of $n = 0$. In Fig. 4.10, we plot the total energy of the filaments (both strain and bending), versus $\gamma$, for the values of $n = 0$ through 6. For all values of $\gamma$, the homogeneous pitch state of $n = 0$ is always lowest in energy, as it has no crumples along which the filaments possess higher bending energy. However, the steeper slopes of larger $n$ modes suggest that they may overtake the $n = 0$ mode as $\gamma \to \infty$. This argument is strengthened by looking at just the cohesive energy alone in Fig. 4.10(c), which shows high $n$ bundles overtaking their lower-$n$ counterparts as $\gamma$ increases. Fig. 4.10(d) shows the reconstructed equivalent surfaces for $n = 0$ and $n = 6$. Notice the (slight) amount of curvature focusing at the top of the dome for the larger $n$, similar to the behavior predicted in [78]. A more advanced model is needed to fully reach the asymptotic limit of $\gamma \to \infty$, as increasing $\gamma$ further in this bead-spring representation leads to difficulties in energy minimization due to the 5+ orders of magnitude difference in strain to bending energy scales.

4.5 Radial crumpling for 7-fold disclinations

In this section, we will perform a similar analysis to the previous, but applied to bundles containing centered 7-fold disclinations. For comparison, a sufficiently flexible crystalline membrane containing a negative disclination ($s = -2\pi/6$), will buckle into a saddle-like shape with negative Gaussian curvature. This configuration increases the azimuthal spacing between atoms, effectively canceling out the azimuthal compressive stresses caused by the defect [78]. For a bundle, the equivalent texture is one of splay, similarly increasing the distance of closest contact between filaments. This feature can be attributed to the negative sign in front of the in the eqn (4.3), for equivalent Gaussian curvature. Importantly, splay does not conserve bundle volume, and therefore can’t continue uninterrupted in long aspect ratio bundles, $L/R \gg 1$. 

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One may surmise that a compromise is reached by sacrificing bending energy for strain energy. This can be accomplished by alternating the direction of splay along the bundles length, similar to the crumpled patterns found for 5-fold bundles, but now directed radially instead of azimuthally. Using both our continuum and bead-spring models, we will explore the nature of this instability, in light of what is known to occur in membranes.

Figure 4.11. Crumpling instability for 7-fold disclinations. Plotted is the mean twist angle $\theta$, versus $\gamma$, for various aspect ratios, $L/R$. The bottom row of figures are example structures for $L/R = 5.4$, showing the developing buckling instability as $\gamma$ increases.

Results using the bead-spring model are found in a manner similar to the steps laid out in the previous section for torsional crumpling, except that $\gamma$ is increased in a stepwise fashion between energy minimizations, rather than decreased. Simulations are run using the same parameters presented in section 4.3, except that for $R/\Delta_0 = 10$, $N_f = 428$, and $L/R = 0.2, 0.3, 0.5, 0.7, 1.1, 1.6, 2.4, 3.6, 5.4$, and 8. Final
results are shown in Fig. 4.11. The most obvious difference between positive (5-fold) and negative (7-fold) disclinations observed here, is that 7-fold disclinations promote filament tilt in the radial direction as opposed to the azimuthal direction. The aspect ratio-dependent sharp jump in $\theta$, as $\gamma$ is increased, is an artifact of the minimization protocol, rather than a representation of the true ground-state structures. Preliminary results show there to exist a continuously smooth-like trend in $\theta$ for the entire range of $\gamma$. Nevertheless, the final buckled states are indeed lower in energy than the straight ones, and we can at least qualitatively review their structures. Following the example structures for $L/R = 5.4$, in Fig. 4.11 (and indeed all of the final structures not shown), a trend is noticed in the evolution of the buckled structure. The initial buckled state (2nd example bundle from left) is very similar to the unbuckled state, except for a slight shearing within the $xy$ plane, which occurs at each end.

This shear pattern is best understood by observing the equivalent Gaussian curvature, in Fig. 4.12. Additionally, the in-plane components of the filament tilt, $t$,
are also overlaid on the curvature mapping, showing that the shearing at the ends is also accompanied by a slight splaying of the filaments, as predicted with eqn (4.3). Meanwhile, at low $\gamma$, the central bulk of the bundle (lower cross-sectional slice in Fig. 4.12), remains straight. This suggests that free ends of the bundle result in a lower $\gamma$ threshold for buckling. Alternatively, for $\gamma = 2,500$, the patterns in filament tilt and mapped curvature appear much more complex, but with a common feature of the largest (most negative) $K_G$ filaments located in the vicinity of the central defect. This characteristic was also present for 5-fold disclinations, where the largest (most positive) $K_G$ filaments were also focused near the center. Note that the integral of $K_G$ over the entire cross-sectional area is always less than zero, for all bundles containing negatively charged, $s < 0$, defects. The full nature of this transition is not yet fully understood, and requires careful simulations that more readily minimize to ground-state structures, in order to fully analyze this behavior. Once this is done, we can examine quantities such as the integrated Gaussian curvature at various locations along the bundle’s height, to determine whether the buckling is identical everywhere, or has an end-specific behavior.

For comparison to bundles in the limit of infinitesimal filament spacing, we perform a linear stability analysis similar to that of the previous section. Our goal is to solve the eigenvalue problem for an undulating radial-splay deformation, determined by the solution to eqn (4.27). Judging from eqn (4.3), we suspect that an equivalent negatively curved mapped surface is achieved with radial patterns of filament tilt. However, strictly speaking, such a pattern cannot continue forever, as this would lead to very large strains as filaments extend out to infinity. Alternatively, the splay cost can be traded for bending, by alternating the direction of splay along the bundle’s length. This validates our choice of a periodic displacement of eqn (4.26), and leads us to an undulatory structure, as shown in Fig. 4.13(a). A 7-fold disclination is defined with a charge of $s = -2\pi/6$, and again we make an additional coordinate
transformation to the radial component of the perturbative displacement to allow us
to solve a first order non-linear ODE

$$\rho(r) = \Lambda_0 r \exp \left[ \int_0^r dr' f(r') \right],$$  \hspace{1cm} (4.45)

where $\Lambda_0$ is an arbitrary constant. Here $f(r)$ describes the deviations from the uniform
splay-rate in the plane, $r \partial_r (\rho/r) = f(r) \rho$, and transforms eqn (4.27)

$$-\frac{1}{2} (f^2 + f' + 3f/r) + \Lambda_\rho \ln r = -\alpha_\rho,$$  \hspace{1cm} (4.46)

where the splay coupling constant

$$\Gamma_{\rho} \equiv \frac{sk^2(1 - \nu^2)}{8\pi} > 0,$$  \hspace{1cm} (4.47)

parameterizes the coupling strength to the logarithmic (splay) potential. Unlike the
case of torsional crumplings, this equation must be solved for a non-zero value of $f(1)$,
to make the vanishing radial stress boundary condition, $f(1) = -(1 + \nu)$. It is
this boundary condition which leads to a minimal value of $\Gamma_\rho$, for which $\alpha_\rho > 0$, a
necessary condition for finding finite values of $\gamma_c$.

In Fig. 4.13(b), we plot the numerical solution of $\alpha_\rho(\Gamma_\rho)$, for $\nu = 1/3$, which shows
that a "bound state" (positive $\alpha_\rho$), does not exist for $\Gamma_\rho \leq 5.5$ (inset). Thus, unlike
the torsional buckling in the presence of a 5-fold defect, which becomes unstable
for arbitrarily weak coupling (or long-wavelengths), here there is a range of long-
wavelengths (small $k$), for which no unstable solution exists at $\gamma$. Using the definition
for the splay eigenvalue in eqn (4.31), we have

$$\gamma_c(k) = \frac{(1 - \nu^2)k^4}{2\alpha_\rho(|s|k^2(1 - \nu^2)/8\pi)},$$  \hspace{1cm} (4.48)
Figure 4.13. (a) An axisymmetric undulatory structure, assumed to be the buckling pattern for a bundle containing a 7-fold disclination. (b) The dependence of the splay eigenvalue $\alpha_\rho$, on the effective coupling to the stress-induced ”potential”, $\Gamma_\rho$, showing negative values for $\Gamma_\rho \leq 5.5$, shown in inset. (c) The stability map of splay undulations in terms of wavevector $k$ and $\gamma$. The minimum $\gamma$ unstable mode occurs for $kR = 21.8$, and $\gamma_c = 13,685$.

which is plotted in Fig. 4.13(b). This figure shows that the minimum unstable value of $\gamma_c$, occurs at a mode $kR = 21.8$, for which $\gamma_c = 13,685$, setting the threshold Föppl-von Kármán number for the splay instability driven by 7-fold disclinations.

This instability analysis doesn’t consider certain features specific to our bead-spring model. Specifically, the bundle ends are ignored entirely in this approximation by assuming $L/R \to \infty$, which is admittedly far from the largest aspect ratio simulated in the discrete model, of $L/R = 8$. The free ends appear to allow for an initial shear-induced buckling behavior, at a $\gamma$ far below the critical $\gamma_c$ predicted for patterns of radial splay. Therefore, we can see the $\gamma_c$ as an upper bound value for what we would expect for finite length bundles. Additionally, our applied perturbation is a simple axisymmetric pattern, which is easy to solve analytically, but not necessarily the most unstable mode of deformation, nor the far-from threshold buckled configuration. Indeed, as seen in Fig. 4.11 under the coarse-grained model, the final states are not simple axisymmetric undulations, but are rather a more complex crumpled texture. To fully understand the two models we would first need adequate simulation
results. Once this is acquired, a comparison could be made by looking at the $L/R$ dependence of the buckling behavior. This would allow us to predict the value of $\gamma_c$ in the asymptotic limit of $L/R \to \infty$, within the bead-spring model, and compare it to the values found using the instability analysis. Additionally, Fig. 4.13(c) predicts a certain behavior of $\gamma_c$ with the wavenumber $k$, which can be analyzed in a manner similar to what is done in Fig. 4.9. And finally, the far from threshold wavelength of the crumpled can be measured and compared to $\lambda = \sqrt{K_3/Y}$, as is done in Fig. 4.7.

4.6 Buckling behavior of off-centered 5-fold disclinations

Thus far we have only considered centrally located defects. These tend to produce axisymmetric patterns of filament tilt (at least for 5-fold disclinations), and lead to a very limited variety of possible structures, where the centerline of the bundle remains relatively featureless and straight. In this section we will overcome this hurdle by allowing for off-centered defect positions. Specifically, we will study the buckling behavior of 5-fold disclinations located at non-central positions in the lattice, which yield helically wound centerlines. This is accomplished using the bead-spring model, with $R = 10\Delta_0$ (about 305 filaments total), $L/R = 10$, and $\ell_0 = 0.5\Delta_0$. The distance of the defect from the center of the bundle, $r_d$, was varied between $0\Delta_0$ and $7\Delta_0$, and $\gamma$ was varied between 0.005 and 1.581. Resulting energy minimized structures for the maximum value of $\gamma$ are shown in Fig. 4.14.

It is clear to see that the buckling behavior is highly dependent on the position of the defect within the bundle’s cross section. For a centered defect of $r_d = 0\Delta_0$, we observe the original homogeneous pitch structure. Alternatively, for $r_d = 7\Delta_0$, the centerline of the bundle becomes helical in nature, while the filaments themselves twist only minimally around the backbone. Varying the position of the defect from centered to off-centered, smoothly transitions between these two states as the helical twist of filaments around the bundle’s centerline is traded for a helical twist of the
Figure 4.14. Low energy 3D structures (left bundles), and equivalent Gaussian curvature maps of the central cross section (right cross sections), for various 5-fold disclinations located at $r_d/\Delta_0 = 1$ through 7. For all examples shown, $\gamma = 1581$, $R = 10\Delta_0$, and $L/R = 10$. A central defect yields a ground state of a homogeneous pitch bundle (top left example). As the defect is moved away from the center, the bundle centerline becomes progressively more helical. Select filaments in the 3D structures are colored orange to help highlight the pattern. Although the largest value of equivalent Gaussian curvature quickly shifts to the outer boundary of the bundle (at around $r_d \approx 3\Delta_0$), the mean position of curvature, $x_K$ (represented with a black dot), defined in eqn (4.49), remains situated close to the defect.

The measured in-plane distance between the helical axis of the bundle centerline, and the bundle’s center of mass, is designated $r_h$. We define the location of this helical axis by tracking the path of the centerline of the bundle, defined as the curve traced out by the center of mass of all the filaments at successive $z$ layers. This centerline closely resembles a helix with a well defined torsion and radius, allowing us to estimate the helical pitch and radius, as reviewed by Kamien [99].

Naively, one may expect that in the limit of $\gamma \to \infty$, the buckled structure will evolve so as to match the helical axis with the defect location, i.e. $r_H \approx r_d$, however, this turns out to be not the case. We can justify the true behavior by employing our equivalent Gaussian curvature mapping from the previous sections. The results of this mapping are also shown in Fig. 4.14, for each value of $r_d$. Due to the gradients in tilt, filaments located closest to the helical axis will possess the greatest Gaussian curvature. However, we notice that the helical axis quickly locates itself outside of
the bundle for \( r_d \gtrsim 2\Delta_0 \). This suggests that the true dependence of the buckling pattern on the location of the defect, is to instead match \( r_d \) with the mean positions of curvature, \( x_G \), defined as

\[
x_G = \frac{\sum_{i=1}^{N_f} x_i K_G(i)}{\sum_{i=1}^{N_f} K_G(i)},
\]

(4.49)

where \( x_i \) is the position of filament \( i \), and \( K_G(i) \) is the equivalent Gaussian curvature for filament \( i \), defined in eqn (4.36). In essence, \( x_G \) calculates the center of mass of all filaments, but weighted by the Gaussian curvature value for each filament. The location of \( x_G \) is represented by the black dots in the cross-sectional images in Fig. 4.14. A plot displaying this near-linear relationship, as well as the behavior of \( r_H \), is shown in Fig. 4.15. The minimal energy states are ones that more closely match the radial distance of \( x_G \) from the center of the bundle, \( r_G \), to the defect position \( r_d \). A similar situation is found in Monte Carlo simulations of crystalline order on weakly curved surfaces [84]. Note that these results are not yet in the limit of zero bending energy, where \( \gamma \to \infty \). As we approach this limit, both \( r_H \) and \( r_G \) will increase, as

![Figure 4.15.](image)

Figure 4.15. Evolution of the buckled bundle structure with radial location of off-centered 5-fold disclinations, and \( \gamma = 1581 \). Blue line corresponds to the distance of the bundle’s centerline to the helical axis, \( r_H/\Delta_0 \). Purple line corresponds to the distance of of the centerline to the mean position of curvature \( r_G/\Delta_0 \). Dashed black line has a slope of one.
the bundle will be less inclined to remain straight. In the end, these findings closely resemble the results of a simplified 2D model by Grason [91], and also help explain why \( r_H > r_d \).

The full \( \gamma \) dependence of the final structures are shown in Fig. 4.16. Values for

![Figure 4.16](image)

**Figure 4.16.** Plotted are the dependence of the twist angle \( \theta \) (a), and centerline helical radius \( r_H \) (b), on the Föppl-von Kármán number, \( \gamma \). Line color and size correspond to a particular value of \( r_d \), with the thick red line representing centered 5-fold disclinations, \( r_d = 0 \).

the twist angle, \( \theta \), and the centerline helical radius, \( r_H \), are plotted against their dependence on \( \gamma \). As a baseline, for a centered 5-fold disclination (red line), we see that the centerline remains straight at all values of \( \gamma \), while only the twist angle changes in accordance with what was established in section 4.3. As \( r_d \) increases, the bundle substitutes twisting filaments around the center of the bundle, with helical writhing of the entire bundle itself. Both transitions are smoothly dependent on \( \gamma \), and there is no critical value required to buckle into these configurations. This is similar to the homogeneous pitch bundles, in that the deformations are largely just rigid rotations and translations of cross-sectional slices of the bundle. Unlike for 7-fold disclinations, this rigid rotations freely allows for these "soft modes" of deformation.
At large values of $\gamma$, $r_H$ first increases with $r_d$, then at $r_d \gtrsim 4$, it begins to decrease again. We expect this phenomena arises from the interaction of defects with the free boundary of the medium. Generally speaking, the strain energy of a centered disclination grows as $E \sim s^2 R^2$. However, as the defect is moved closer to the edge, the condition of a stress free boundary allows the total stress to be relieved, such that in the limit of $r_d \to R$, the energy approaches zero. This can also be translated into equivalent Gaussian curvature, and observed in Fig. 4.14, as a resulting reduction of required curvature as $r_d \to R$. It is clear to see that we have reached the asymptotic limit of highly flexible filaments only for $r_d = 0\Delta_0$, but not for off-centered disclinations. This is especially evident in Fig. 4.16(b), suggesting that a better match of $r_G$ to $r_d$ can be achieved in Fig. 4.15, at higher values of $\gamma$. A full analysis of off-centered defects under the continuum elasticity model is made difficult by being forced to drop the assumption of axisymmetric tilt patterns, and is not attempted here.

On a final note, we recognize that crumpled patterns likely exist for off-centered defect bundles. Although it is difficult to imagine the shape of $\gamma \to \infty$ crumpled structures, we suspect that they will have a similar curvature-focusing effect as they do for the homogeneously twisted bundle. We leave the analysis of these structures for future study.

4.7 Conclusions

In this chapter we explored the buckling behavior of flexible cohesive filament bundle containing various types and locations of defects. Our analysis was performed in the context of the comparable buckling behavior for 2D crystalline membranes. We modeled the bundles using two compatible models. First, a fully 3D bead-spring model of filaments, introduced in section 4.1; and second, a continuum elasticity model, introduced in section 4.2. The bundle-equivalent Föppl-von Kármán number,
γ, was introduced as a means to quantify the energetic cost of interfilament elasticity, versus filament bending.

In section 4.3, we showed that bundles containing centered 5-fold disclinations are unstable to twisting at all values of γ; a behavior that is unlike that of their membrane counterpart, which only buckles above a critical value of γm. Additionally, the patterns of filament tilt within a cross section of the bundles were mapped onto a surface whose Gaussian curvature provided identical distortions to interparticle spacing as tilt does for interfilament spacing.

In section 4.4, we presented an entirely new mode of bundle buckling, termed torsional crumpling, where the handedness of the twist alternates direction along the length. We employed linear stability analysis with the continuum model to derive the critical γc at which a straight bundle becomes unstable to torsional wrinkling. Furthermore, we suggested that these deformations provide a route for bundles to focus gradients in the tilt pattern on to the defect location, much in the same way membranes are able to focus their curvature to the defect in the limit of γm → ∞.

In section 4.5, we performed a similar analysis, but for 7-fold disclinations, and explained how the incompatibility of radial splay with patterns of tilt along the z axis, result in qualitatively different behavior from their 5-fold cousins. Namely, that in the limit of large aspect ratio, L/R → ∞, buckling only occurs above a critical γc. While for finite length bundles, the bead-spring model showed that the free ends allow for a shearing of the filaments along the length, which in turn allow for splay and a slight relaxation of the defect-induced strain energy at values of γ much lower than that predicted by the instability analysis.

Finally, in section 4.6, we used our coarse-grained model to analyze bundles with off-centered defects. We found that these promoted writhing textures, where the centerline of the bundle itself became helical in nature. This behavior was explained using the Gaussian curvature mapping technique, which showed that off-centered de-
fects yield off-centered twist patterns that matched the Gaussian curvature to the defect location. In conclusion, this chapter explained the previously hidden connections that exist between the 2D packing topology of flexible cohesive filaments, and the 3D structures of the bundles they form.
CHAPTER 5
CONCLUSIONS

Throughout this proposal we elucidated the connection between the 2D packing and the 3D configuration of cohesive flexible filament bundles, by developing several models that allowed us to replicate cohesive filament interactions. This allowed us to understand the influence of a bundle’s geometry and structure on its energy.

In chapter 1, we motivated this proposal by first introducing the concepts of chirality and packing. This was followed by a discussion on geometric frustration, specifically related to the twisting of columnar materials. We then closed with continuum elasticity scaling arguments to hypothesize the specific relation between disclination defects and twist.

In chapter 2, we introduced a discrete model of cohesive filament interactions as a means to study the twist-dependent interior packing topology of bundles. We began by defining the concepts of contact and cohesion in our model, and determined the geometry of filaments within a bundle of constant pitch. We then developed a framework for describing the interior packing topology of crystalline bundles as a means to understand the effects of defects on the energy. This exposition was then advanced with the insight gained by mapping the positions of filaments in the cross section of a twisted bundle to the positions of particles on a curved surface, specifically a surface designated as the bundle-equivalent dome.

In chapter 3, we laid aside any specifics of the interior and introduced a continuum model of the surface energy of twisted bundles. We began by defining the twist-dependent bundle geometry for finite-length filaments. Then we combined this with
a notion of cohesive filament interactions to establish the concept of a surface energy. This model was fully examined, with a focus on the ends of the bundles and the effects of slippage from twist. We concluded by adding a filament bending term to our model’s Hamiltonian, and discussed the important parameters necessary to predict whether the ground state of a filament bundle is twisted or untwisted.

Finally, in chapter 4, we constructed a third model of cohesive filament interactions, with a fully 3D discrete bead-spring representation. We discussed the similarities between twisted bundles and curved sheet, and defined an equivalent Föppl-von Kármán number for bundles based on the bending rigidity and cohesive bond strength. Results of this parameter versus twist angle were discussed, along with an exploration into alternating twist structures comparable to the Helfrich-Hurault instability found in tensile-strained bulk columnar materials. We fully analyzed the 3D textures resulting from variations in the type and positions of defects, and explained much of the observed behavior in terms of a discrete version of the mapping of filament tilt onto Gaussian curvature.

There are still many open questions remaining about filament bundles. Specifically, in regards to chapter 4, the buckling behavior for 7-fold disclinations is not yet fully understood, especially the length-dependent behavior. Additionally, other defect types and patterns were not even considered, including: off-centered 7-fold disclinations, dislocations of various positions and orientations, disclinations of higher charge, and multi-defect patterns such as grain boundaries. Also, all of the models presented here assumed fixed filament positions that was identical along the entire length of the bundle. Relaxing this condition could yield interesting 3D patterns, and better predict some biological structures like collagen, where the kinetics of self-assembly permit such loose formations.

We conclude this thesis by stating its value in designing filament bundles. We have discovered the geometrical similarities between curved membranes and filament
bundles, allowing future material scientists to design unique columnar structures using
generic cohesive interactions. Additionally, we have uncovered a library of available
3D structures: twist, undulations, crumples, and writhe; that can now be programmed
into the self-assembly of fibers patterned with the correct packing topology. Perhaps
the best experimental system to pursue as a means to explore our predictions, would
be one similar to the work done by Pokroy and Aizenberg [18]. Highly flexible large
aspect ratio sub-millimeter plastic pillars were adhered to a substrate in a crystalline
lattice and immersed in water. As the water level was decreased, the capillary forces
between neighboring pillars caused them to aggregate into bundles. By selectively
patterning the pillars on the substrate—say with a 5-fold disclination—one could
drive the bundles to twist. A rudimentary model, applicable to both plastic and
hydrogel pillars, is presented in appendix F.
APPENDIX A

DERIVATION OF CONFORMAL MAPPING FOR BUNDLE DELAUNAY TRIANGULATION

True interfilament distance in twisted bundles is not preserved when the cross-sectional packing is projected into the plane (as in a horizontal cross section of the bundle shown in Fig. 1), and hence, determination of the nearest-neighbor bond network requires some care. A very efficient and sufficiently accurate method is to map the filament positions onto the plane via a coordinate transformation that rescales local inter-filament distances in different directions (radially and azimuthally) by nearly equivalent amounts and then perform the standard planar Delaunay triangulation on the transformed array. That is, we use the isothermal coordinate map of filament position \((\rho, \phi)\) in the horizontal section to position \((\hat{\rho}, \phi)\) in the plane. Such transformation is conformal and as a consequence it maps infinitesimal circles on the dome to circles in the plane. For sufficiently small, but finite-size, circles (i.e. \(d/P \lesssim 1\)) the Delaunay triangulation of the mapped positions will give the identical connectivity of nearest neighbors as a triangulation based on true geodesic distances on the bundle equivalent dome.

The coordinate transformation is described by the function \(\hat{\rho}(\rho)\) that transforms twisted bundle metric of eq. (5) into the following form,

\[
ds^2 = \omega^2(\hat{\rho})(d\hat{\rho}^2 + \hat{\rho}^2 d\phi^2), \tag{A.1}
\]

where \(\omega(\hat{\rho})\) describes the conformal scaling of area elements. This transformation satisfies
Figure A.1. On the left is shown the horizontal section of a (5, 5) twisted bundle and on the right the conformal mapping of the that section described by eqn (A.3) with the corresponding Delaunay triangulation connecting the map center line positions.

\[ \frac{\bar{\rho}}{\rho} \frac{\partial \rho}{\partial \bar{\rho}} = \Omega^{-1} \sin \theta(\rho), \]  

(A.2)

from which we find

\[ \bar{\rho} = \frac{\rho}{1 + \sqrt{1 + (\Omega \rho)^2}} e^{\sqrt{1 + (\Omega \rho)^2}}, \]  

(A.3)

and \( \omega(\rho) = [1 + \cos \theta(\rho)] e^{\sec \theta(\rho)} \). In Fig. A.1 we show the planar section of a (5, 5) bundle as well as the conformal transform of the cross section and corresponding triangulation. Note that for this bundle \( d/P = 0.12 \), so that filament cross sections are very nearly circular in the projection.

This triangulation method always produces a bond network with a convex boundary of bonds encompassing the entire bundle. However, in many cases this includes extra long bonds that bridge naturally concave sections of the hull. Because these bonds exist as an artifact of the triangulation and not as a product of the governing interaction energy, bonds along the boundary with \( \Delta^* \geq 1.4d \) are removed from the triangulation. From this final triangulation, simulation results are then classified by their radius \( R/d \), net disclination charge \( Q \), and total disclination number. \( R \) was
calculated to be the mean distance from the center of mass of every filament along the outer hull of the cross section.
Appendix B

Deriving the Equivalent Gaussian Curvature for the Bundle-Equivalent Dome

Starting with eqn 2.17, which gives the azimuthal length, \( \ell(\rho) \), available to pack additional filaments at a radial position of \( \rho \); we will derive the equivalent Gaussian curvature on the mapped bundle-equivalent dome, eqn 2.18. Restating eqn 2.17

\[
\ell(\rho) = P \sin \theta(\rho) = \frac{2\pi \rho}{\sqrt{1 + (\Omega \rho)^2}},
\]

where \( \rho \) travels a radial path along the surface. Similarly, the radius of this loop is

\[
r(\rho) = \ell(\rho)/2\pi = \frac{\rho}{\sqrt{1 + (\Omega \rho)^2}}.
\]

Assuming an axisymmetric shape defined in polar coordinates, we can solve for

\[
\frac{dr}{d\rho} = (1 + (d\rho \Omega^2))^{-3/2},
\]

and given that the surface obeys \( d\rho^2 = dr^2 + dz^2 \),

\[
\frac{dz}{d\rho} = \frac{d\rho \Omega \sqrt{3 + 3d\rho^2 \Omega^2 + d\rho^4 \Omega^4}}{(1 + (d\rho \Omega^2))^{3/2}}.
\]

From here we can solve for the height of the surface as a function of \( \rho \)

\[
z = \int_0^\rho \frac{dz}{d\rho} d\rho,
\]
which is analytically solvable but a bit lengthy to report. Now we have a full parametric equation of the bundle-equivalent dome

\[ \mathbf{R}(\rho) = r(\rho) \cos(\theta) \mathbf{\hat{x}} + r(\rho) \sin(\theta) \mathbf{\hat{y}} + z(\rho) \mathbf{\hat{z}}, \quad (B.6) \]

where \( r \) is defined in eqn B.2, and \( z \) is defined in eqn B.5.

Following a formal definition from ref. [128], a surface’s Gaussian curvature is defined as

\[ K_G = \frac{LN - M^2}{EG - F^2}, \quad (B.7) \]

where \( E, G, \) and \( F \) are the coefficients of the first fundamental form; and \( L, M, \) and \( N \) are the coefficients of the second fundamental form. Following through with the algebra yields our final result

\[ K_G(\rho) = \frac{3\Omega^2}{[1 + (\Omega \rho)^2]^2}. \quad (B.8) \]
APPENDIX C

SUPPRESSING DEFECT GENERATION IN TWISTED BUNDLES

While the preference for twist is driven by effects at the boundary of the bundle, the complex evolution of cross-sectional packing—as evidenced by the universal increase in topological charge of the packing—plays a critical thermodynamic role in stabilizing twisted bundles. Above a critical threshold of twist \( \theta \simeq 22^\circ \), excess 5-fold disclinations are needed in the ground-state packing to screen the elastic effects of the packing frustration generated by twist. Continuum elasticity arguments made in section 1.4 have shown that twist decreases inter-filament spacing between azimuthally-separated neighbors by an amount proportional to \((\Omega \rho)^2\), ultimately leading to an increase in energy density that grows as \((\Omega R)^4\) for defect-free bundles [90]. Hence, in the absence of topological defects which act to “neutralize” the stresses generated by twist, the elastic cost of twisting defect-free bundles would continue to grow unmitigated at large twist angle, likely overwhelming the gains in cohesive energy at the boundary.

We demonstrate the importance of achieving the appropriate defect configuration for stabilizing twist by considering a class of \textit{kinetically-constrained} bundle packings in our numerical simulation model. Unlike our numerical search for ground states described in section 2.3, which explored an ensemble of in-plane packings at each value of twist, in Fig. C.1 we analyze the energetics of simulated packings achieved in the following kinetically-constrained algorithm. Beginning from the energy-minimized packing of an untwisted bundle, we increase \(\Omega\) in small increments of 0.001\(d\). For each \(\Omega\), we perform a steepest-descent minimization of bundle energy (in the \(L \rightarrow \infty\)
Figure C.1. Lower thin line shows the thermodynamically lowest energy ground states vs twist rate $\Omega d$ (same as shown in Fig. 2.9). Upper thick lines represent the constrained ground state energies. $N = 82$ for both.

compares the energy of kinetically-constrained bundles to the simulated ground states presented in section 2.6. For small $\Omega d$, the cohesive energies of both states are identical as expected since no large-scale filament reorganizations are required. This persists until a high enough twist forces the ground state bundle into a new configuration at $\Omega d \approx 0.1$. At this point they cannot reach this new ground state packing as it requires a global rearrangement of the filaments. In the kinetically-constrained packings, such defects only enter at the boundaries of the bundle, migrating slowly towards the center upon further twisting. Fig. C.1 shows that further increase in twist eventually does allow the constrained bundle to overcome the local energy barriers of filament arrangement; however, the total energy of states continues to exceed the ground state packings inhibiting the stability of the twisted bundle relative to the $\Omega = 0$ state.

The discrepancy between ground state packings and this simple model of kinetically-constrained bundles demonstrates two key points about disclinations in twisted fil-
ament bundles. First, changes in surface energy will only stabilize twisted bundles over straight bundles provided that the appropriate, energy-minimizing configuration of disclinations punctuates the cross-sectional packing. And second, the kinetic pathways by which topological defects enter into and migrate throughout the cross section may place strong constraints on whether an externally or intrinsically twisting system of filaments is able to achieve an optimally-twisted state.
APPENDIX D

LATTICE ORIENTATION DEPENDENCE

As described by eqn (3.3), the energy per unit of exposed surface area of a bundle, $\Sigma$, is dependent on the angle, $\Theta$, between the local filament tangent, $T$, and the cutting plane normal vector, $n$. This definition separates the surface corresponding only to the loss in contact lengths between filament pairs, as opposed to surface area associated with filament ends. However, as noted in section 3.2, there is an additional dependence of $\Sigma$ on the orientation of the cutting plane with respect to the lattice directions at the surface, which is defined in terms of the component of $n$ that lies in the horizontal cross section, (shown in Fig. D.1)

$$n_\perp = n - \frac{T(n \cdot T)}{1 - (n \cdot T)^2}.$$  

We consider the loss of contact for a single filament, $i$, for each its six nearest neighbors, $j$,

$$\ell_s(j) = |\tan \Theta|d|\cos \psi_j|,$$

where $\psi_j$ is the angle between $\Delta_{ij}$ and $n_\perp$. Defining the angle of this orientation as $\Psi$, the smallest angle between the bond directions in the hexagonal lattice and $n_\perp$ we have $\psi_j = \Psi + \pi j/3$. Using the fact that area per filament at the cutting surface is $n_0^{-1} \sec \Theta$ and defining the dimensionless parameter $\alpha$ as in eqn (3.3), we have

$$\alpha(\Psi) = \frac{n_0d^2}{2} \sum_{j=0}^{5} |\cos (\Psi + \pi j/3)|,$$
Figure D.1. Top view of bulk filaments in Fig. 3.2 cut by a plane with the normal, \( \mathbf{n} \). Filament \( i \) and its neighbor \( j \) are specified. The interfilament spacing is \( d \), and \( \Psi \) is the angle between the horizontal cross section component of the normal, \( \mathbf{n}_\perp \), and the vector \( \mathbf{X}_j - \mathbf{X}_i \).

where \( n_0 = 2/\sqrt{3}d^2 \). Summing over the filament neighbors we have,

\[
\alpha(\Psi) = \frac{4}{\sqrt{3}} \cos \Psi, \text{ for } -\pi/6 < \Psi < \pi/6.
\]  

Two limiting cases of \( \alpha \): 1) the lattice vector is aligned with \( \mathbf{n}_\perp \) (high surface energy), yielding a maximum \( \alpha(\Psi = 0) = 4/\sqrt{3} \); and 2) the lattice vector is \( \Psi = \pm\pi/6 \) maximally offset from \( \mathbf{n}_\perp \) (low surface energy), yielding a minimum \( \alpha(\Psi = \pi/6) = 2 \).  

For surface elements at the ends of the bundle, the distribution of \( \Psi \) roughly visits all orientation equally, hence, suggesting the appropriate value of \( \alpha \) is average with respect to \( \Psi \): \( \langle \alpha \rangle = 4\sqrt{3}/\pi \approx 2.2 \).
APPENDIX E

COMPARING DISCRETE AND CONTINUUM MODEL PARAMETERS

Here we will compare the parameters for the discrete and continuum models of cohesive filament bundles, so that we are able to fairly compare them. To start, filament curvature is defined from a continuum filament to be

\[
\kappa^2 = \left( \frac{\partial T}{\partial l} \right)^2 \approx \frac{(T_{n+1} - T_n)^2}{\ell_0^2} = 2 \frac{1 - T_n \cdot T_{n+1}}{\ell_0^2}.
\]  
(E.1)

We can convert from the discrete model of bending energy in eqn 4.4, to the continuum model in eqn 4.11

\[
E_b^{(i)} = B \sum_{n=1}^{N_b-1} \frac{1 - T_n \cdot T_{n+1}}{\ell_n}
\]

\[
= B \frac{N_f}{2\ell_0} \sum_{n=1}^{N_f} \sum_{n=1}^{N_b} \kappa^2
\]

\[
= \frac{B}{2} \sum_{n=1}^{N_f} \int_{L} \kappa^2 d\ell
\]

\[
= \frac{1}{2} \frac{2B}{\sqrt{3}\Delta_0^2} \int (\partial z)^2 dV.
\]
(E.2)

where \( N_f = 2\pi R^2 / \sqrt{3}\Delta_0^2 \) is the number of filaments, and \( N_b = L / \ell_0 \) is the number of beads on a single filament. From this we can compare the bending parameters between the two models in the limit of \( N_b \to \infty \) [129].

\[
K_3 = \frac{2B}{\sqrt{3}\Delta_0^2}.
\]  
(E.3)
For the discrete model of the cohesion between filaments, from eqn 4.5 we have

\[
\frac{E_c}{Nb} = \frac{\epsilon}{2} \sum_{Nf} \sum_{\text{neighbors}} (\Delta_{ij} - \Delta_0)^2,
\]

(E.4)

where \(\Delta_0\) is the preferred spacing between filaments. Given the form of the continuum energy per unit bundle length of eqn (1.1), we can follow the steps laid out in reference [78], to arrive at

\[
\frac{E_c}{L} = \frac{\sqrt{3}\epsilon}{4\ell_0} \int 2u_{ij}^2 + u_{kk}^2 dA,
\]

(E.5)

where we have multiplied by an extra factor of 2 to account for the double counting of filament interactions in eqn (E.4). Following through with the conversion gives us a 2D Young’s modulus of

\[
Y = \frac{4\epsilon}{\sqrt{3}\ell_0}.
\]

(E.6)

Some final algebra yields the final version of the bundle-equivalent FvK number, in both the continuum model and discrete model

\[
\gamma = \frac{YR^2}{K_3} = \frac{2\epsilon R^2 \Delta_0^2}{B\ell_0}.
\]

(E.7)
Here we present a rough calculation estimating the filament flexibility and bundle size required to promote twist in a bundle containing a single centered 5-fold disclinations. The cohesive interactions between filaments are derived from the capillary forces between them, trying the reduce surface energy of any water-air or water-oil interfaces.

We begin by estimating the strength of capillary forces of a liquid bridge between two pillars (Fig. F.1) [130]. The general equation for force per unit length with this geometry (ignoring end effects) is

\[
\frac{F_{\text{cap}}}{L} = 2\gamma \left( \sin(\alpha + \theta) + \frac{r}{R} \sin(\alpha) \right),
\] (F.1)
where \( \gamma \) is the surface tension of the liquid, and the other symbols are described in Fig. F.1. The first term of this equation is the force due to surface tension, and the second term arises from capillary pressure. For a rough approximation we will assume that \( \alpha = \pi/2 \) and \( \theta = 0 \), which corresponds to the bridge being flat with no curvature. The surface tension of water at room temperature is \( \gamma = 0.0728 \text{N/m} \), giving us

\[
\frac{F_{\text{cap}}}{L} = 0.146 \text{N/m}.
\]  

We can assume from the notion of the Rayleigh instability, that this bridge will collapse when its length exceeds its width. This allows us to calculate the energy of the bridge to be

\[
\frac{E_{\text{cap}}}{L} = \text{displacement} \times -F_{\text{cap}} = 2r \times -0.146 \text{N/m}.
\]  

To calculate the bending energy required, we can assume the simplest case of five fibers twisting around one central fiber, the centerline of the twisting fibers can be described by the general equation for a helix

\[
\vec{R}(s) = \{ \rho \cos(\Omega s + \theta), \rho \sin(\Omega s + \theta), s \},
\]  

where \( \rho \) is the radius of helix, \( s \) is the arc length position along the curve, and \( \Omega \) is the rate of twist of the bundle [99]. The curvature of the helix based off this formulation is

\[
\kappa = \frac{\rho}{\rho^2 + \Omega^{-2}}.
\]  

Assuming that the fibers with radius \( r \) are in complete contact with each other, the critical value of twist is \( \Omega \rho_{\text{critical}} = \sqrt{2}/9 \). This is the twist needed to achieve a
simple centered five-fold defect [91]. From here we can calculate the curvature of one of these fibers as it twists around the central fiber

$$\kappa = \frac{2r}{4r^2 + \frac{4r^2}{2/9}} = \frac{1}{11r}.$$  \hspace{1cm} (F.6)

The energy per length of bending an elastic rod is

$$\frac{E_{\text{bend}}}{L} = \frac{1}{2}EI\kappa^2,$$  \hspace{1cm} (F.7)

where $E$ is Young’s modulus, and $I$ is the area moment of inertia, in our case of a circular rod, $I = (\pi/4)r^4$ [86]. Inserting in the values for $I$ and $\kappa$ leads to

$$\frac{E_{\text{bend}}}{L} = \frac{\pi}{968}Er^2.$$  \hspace{1cm} (F.8)

Now we have our equation for the energy per unit length of fiber for a capillary bridge (Eq. F.3), as well as the energy per unit length for the required amount of bending needed to wrap five fibers around one central fiber (Eq. F.8). From here we can solve for the critical radius $r_{\text{crit}}$ where these two energies are of equal magnitude. The Young’s modulus for PDMS can range between about $300-3000 kN/m^2$, so we will assume the lower end for flexible fibers to be $E \approx 400 kN/m^2$. Solving for the critical radius gives us $r_{\text{crit}} \approx 220 \mu m$. For fiber radii below this value the twisted state has a lower energy than the untwisted state. Conventional macroscale manufacturing of these pillars (i.e. drilling holes into a sold mold) can only reach a minimal radii of about $600 \mu m$, while microscale techniques can achieve radii down to potentially a few microns [131].

Alternatively, instead of having capillary bridges spanning between each pair of filaments, an easier system to construct may be one where the entire bulk of the
bundle is composed of water, and the bridges between filaments exist only on the outside. A cross section of our system looks like Fig. F.2.

Neglecting friction, gravity and other terms, our energy depends on only the energy of bending filament, $E_b$ and surface tension energy, $E_\gamma$. In the case of a straight bundle, $E_b = 0$; while in the case of a fully twisted bundle we will assume that all the gaps between the outer pillars, $b = 0$, therefore $E_\gamma = 0$. We already established the bending energy in eqn (F.7), and filament curvature in eqn (F.6). Combining this all together, the bending energy per unit length as it depends on the filament diameter, $d$, the number of layers $m$, and rate of twist, $\Omega$, is

$$
\frac{E_b}{L} \approx \frac{\pi}{128} Y d^4 \sum_{l=1}^{m} 5l \frac{\Omega^4 d^4 l^2}{(1 + \Omega^2 l^2 d^2)^2}.
$$

(F.9)

Here we have assumed that all filaments belonging to one layer all lie at a radial distance of $ld$, while in reality this is the maximum radial distance. Anyway, assigning...
the fully twisted value of the rate of twist to be the continuum elasticity predicted value \( \Omega = \sqrt{\frac{1}{3}}/R \), where \( R = md \), gives us

\[
\frac{E_b(m)}{L} \approx \frac{5\pi}{1152} Y d^2 \sum_{l=1}^{m} \frac{l^3}{(m^2 + l^2/3)^2}.
\]  

(F.10)

Now for the surface tension energy, \( E_\gamma \). Ignoring end effects, this is simply

\[
E_\gamma = \gamma A,
\]  

(F.11)

where \( A \) is the total exposed surface area of the liquid between the pillars, which we can break down into \( A = 5mLb(m) \), where \( b(m) \) is the perpendicular spacing between the outer pillars (shown in Fig. F.2), and \( L \) is the length of one pillar. As stated earlier, \( b(m) = 0 \) in the fully twisted state, leading to \( E_\gamma = 0 \). However, for the untwisted state we can use trigonometry to solve for \( b(m) \), giving us \( b(m) \approx (\pi/15)d \). Combining this with eqn (F.11) gives us the final result of

\[
\frac{E_\gamma(m)}{L} \approx \frac{\pi}{3} \gamma dm.
\]  

(F.12)

Now we have the two important energy, eqn (F.10) and eqn (F.12). Within this simple model, the ground state of our bundle will be twisted if \( E_b(m) < E_\gamma(m) \). We can solve this to find the critical filament diameter \( d^* \), below which the ground state is twisted. The surface tension for our oil-water interface is about 1/3 that of air-water surface tension. Combining all the parameters of our hydrogel pillar system:

\begin{align*}
\text{Surface Tension: } & \gamma = 0.0243 \text{ J/m}^2 \\
\text{Young’s Modulus: } & E = 10 \times 10^3 \text{ J/m}^3 \\
\text{Number of Layers: } & m = 2.
\end{align*}

(F.13)
Plugging this in we get a critical diameter of $d^* \approx 1 \text{ mm}$ which is very close to current diameters of $d \sim 0.7 \text{ mm}$. This tells us that our experiments are just barely within range of the upper limit of filament diameter. Again, friction, inhomogeneities, and end effects (both the anchored and free ends) are ignored, which would only lead to a smaller $d^*$ than the one estimated here in this simple model.
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