Orientational ordering in crumpled elastic sheets

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Abstract – We report an experimental study of the development of orientational order in a crumpled sheet, with a particular focus on the role played by the geometry of confinement. Our experiments are performed on elastomeric sheets immersed in a fluid, so that the effects of plasticity and friction are suppressed. When the sheet is crumpled either axially or radially within a cylinder, we find that the sheet aligns with the flat or the curved wall, depending on the aspect ratio of the cylinder. Nematic correlations develop between the normals of the sheet’s facets at relatively low volume fractions and the crumpled object has large density fluctuations corresponding to the stacking of parallel facets. The aligning effect of the wall breaks symmetry and selects the direction of ordering.

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Introduction. – At first sight, a thin sheet crumpled into a ball appears to be a very spatially disordered object. When crushed within a shrinking container a sheet collapses into a complicated 3-dimensional labyrinth of nearly flat facets bounded by ridges or creases. The geometry of a piece of paper balled within your fist has indeed been found to be complex, statistically variable, and not ordered in any obvious fashion [1–5]. However, another limit of this process leads to a different intuition. Imagine crushing a can underfoot, or squashing a crumpled object in a trash compactor: as the thin sheet is confined to volume fractions near unity, intuition suggests that the facets of the crumpled object must lie flat, in parallel stacks. One would not expect this arrangement to be ordered like the folds of an accordion, but nonetheless, the arrangement could have underlying long-range orientational order, with the normals to most facets aligning parallel to the confining direction.

In this article, we present evidence that nematic orientational ordering of surface normals underlies the arrangement of material in a crumpled membrane. The examples above supply the intuition that parallel stacks of material are a likely outcome at high density if you squash the object flat. What if one confines the sheet to high density, but not with flat confining surfaces? One of the principal concerns of this article is whether stacking is merely an outcome of conforming to the crushing boundary with a finite penetration depth into the bulk of the crumpled object or if it is a geometric inevitability in trying to pack a sheet into any volume at very high density. In other words, is nematic ordering a boundary effect, or is it a bulk effect, with the alignment of the nematic selected by the boundary? The experiments we describe here study the development of orientational order in the sheet as a function of both the external boundary’s shape and the degree of confinement, that is, the aspect ratio as well as the volume fraction.

Considerations of ordering in confined flexible objects have been better fleshed out in the situation of a 1-dimensional object confined in a 2-dimensional area. Experiments [6–8] and simulations [9] show that a 1D curve confined within a circle forms a series of loops that conform to the circular boundary. Simulations that use simplified models of bending [10], and mean-field models of elastica [11], have shown the existence of a sharply defined nematic transition where long-range orientational order sets in. For elastic rods of a finite thickness, [12] and [11] predict a jamming transition further within the nematic phase.

Confining a 2D sheet into a 3D volume, however, is qualitatively different. Rather than coiling smoothly, even a purely elastic sheet will localize deformation [13–15] and form sharp ridges and vertices under confinement. Furthermore, the constraints of self-avoidance are more severe in this case. Experimentally, the conformation of a curve in a plane is much easier to visualize than a crumpled membrane in a volume, and, therefore, the 2D analogue has been more studied. However, X-ray tomography has been used to study the internal conformation of aluminum foil crumpled into a ball [4,16]. Lin et al. studied 2D slices
of a crumpled ball and found ordered domains whose size increased with compaction. In our own work [4], we made 3D analyses of the orientation of the sheet within the volume of the ball. The orientation was found to be isotropic at all radial locations within the ball; however, we found evidence of stacks of nematically oriented layers forming preferentially near the boundary and growing with increasing compression. The shape of the boundary was not varied in either of these studies ([4] and [16]), and a spherical boundary with Gaussian curvature is not a favorable geometry for the development of nematic stacks in a thin sheet that does not want to stretch to conform to the boundary.

We have therefore designed an experiment in which we confine a sheet within a cylindrical container and vary the type of confinement by altering either the radius or the height of the cylinder; thus, varying the relative amounts of flat boundary and of boundary with mean curvature. In addition to the geometric constraints of self-avoidance, material properties such as friction and plasticity can hinder relaxation towards a lower-energy state. Simulations [17] and experiments [18] show that in the absence of friction and plasticity, the folds and vertices are mobile, thus allowing the membrane to assume lower-energy configurations. In the current experiments we eliminate plastic effects by using elastomeric sheets so that any deformation is reversible [18]. The sheet is immersed in a fluid, so that lamina of the sheet slide past each other with viscous drag, but without static friction. Finally, we visualize the sheet by optical tomography; unlike the slower X-ray technique, optical scans allow us to observe the time-dependent evolution of the conformation.

**Experimental set-up.** – We make polydimethylsiloxane (PDMS) sheets using a 10 : 1 mixing ratio of Sylgard 184 spread onto a silanized glass substrate and cured in an oven at 70 °C overnight. Squares of area \( A_{\text{sheet}} = 150 \) to 200 cm\(^2\) are cut out of the PDMS and infused with a hydrophilic dye (rhodamine B). The sheet is introduced into the experimental cell where it is submerged in dodecane, which closely matches the index of refraction, and pulling it tighter. Shrinking the volume axially and reducing the diameter of the cylinder. The diameter is defined by \( A = \frac{1}{\phi} \), where \( A \) is the area of a facet whose normal lies in the image plane and \( \phi \) is the volume fraction.

We illuminate a cross-section of the \( XZ \)-plane in the interior of the crumpled sheet (fig. 1) with a 532 nm laser sheet of thickness \( \approx 1 \) mm. The fluorescence of the sheet is imaged with a Nikon D80 camera through a filter that eliminates the illumination line. We work with 2D slices taken at the central cross-section of the cylinder. The thickness of a facet whose normal lies in the image plane is 3 pixels, but facets that are inclined away from this direction present a thicker cross-section. To determine the locations of surface points and find surface normals \( \hat{n} \), a canny edge-detection is applied to the grey-scale image that has been smoothed with a mean filter. The edge-detector divides each image into three regions: surface points, fluid points, and bulk points, where \( m(\vec{x}) = 1 \) and \( \vec{x} \in S \); fluid points, where \( m(\vec{x}) = 0 \); and bulk points, where \( m(\vec{x}) = 1 \) (see fig. 1). In addition, the surface normals, \( \hat{n} \), are evaluated. The edge-detection leads to the loss of some surface points within a stacked group of facets. However, when checked visually, we can see that most of the surfaces are indeed detected. More details of our algorithms and error analysis can be found in [19].

A typical experimental protocol involves crumpling, axially for instance, by reducing the confining height \( h \) from the initial cylinder height in discrete steps towards a maximum volume fraction \( \phi \), and then reversing the motion. Each step is taken at a velocity \( = 0.07 \) cm/s, for a total displacement \( = 0.33 \) cm, followed by waiting a time \( \tau_w \) for the sheet to relax. An image is captured immediately before and after this relaxation period. A movie (crop_small.avi) showing a full cycle is shown in the supplementary material.

In order to quantify the relaxation times of the sheet, we have monitored the motion of the sheet over 10 hours. For the experiments described here, we find it unnecessary to wait the maximum relaxation time since the
quantities we measure are insensitive to a waiting time of $\tau_w > 30$ seconds (this is further discussed later in the text). However, small local motions, presumably triggered by ambient noise, persist for hours.

**Alignment with boundaries.** – To study the effect of the confining boundary on the development of alignment, we determine the orientation of normals, $\hat{n}_i$, to the sheet at all locations and construct the scalar product with the normal $\hat{n} = \hat{x}$ or $\hat{z}$ corresponding to the top or side walls. We note that $\hat{n}$ is the 2D projection in the $XZ$-plane of the true 3D normal to the sheet, so that completely random orientation of the 3D vector leads to an average value of $2/\pi$ for the magnitude of a component of the 2D vector. In addition when the alignment increases (decreases) in the $x$- or $z$-direction, it will decrease (increase) in the other direction.

We first quantify the alignment as a function of the distance from the crushing wall. In order to do this, we divide each cross-section into 10 slices, denoted by $j$. For axial crumpling, each slice is a rectangle of dimension $h/10$ by $d$. For radial crumpling, each vertical slice is $d/10$ by $h$. We then find the average orientation of the points located in each section using the equation,

$$O_j = \frac{\sum_{i=0}^{N_j} |\hat{n}_i \cdot \hat{z}|}{N_j}, \quad j = 1, \ldots, 10, \quad (1)$$

where $N_j$ is the total number of surface points in the $j$-th section.

In fig. 2(a), we show $O_j$ in an axial crumpling experiment, as a function of the distance from the top confining wall, $z_j/h$, where $z_j = (j)h/10$. Even in the initial, least-crumpled, state, there is considerable alignment in the vertical direction near the bottom and top walls, but the alignment decays rapidly in the bulk of the system. As we compress with the flat piston, alignment increases everywhere in the bulk of the cell, until the alignment is extremely high everywhere in the most compressed state achieved in this run. Subsequent release of the compression reverses this trend. A similar progression is shown in fig. 2(b) for radial crumpling, where the alignment in the radial direction increases. In each crumpling geometry, the tendency to align is much stronger than in the case of crumpling within a sphere [4] where all boundaries have Gaussian curvature and defeat the tendency to conform to the boundary. Without the frustration of a spherical wall, the high sheet-to-sheet friction, and the plasticity found in the aluminum sheets used in [4], the PDMS sheets align more easily within the cylindrical confinement.

As discussed above, the orientation of the alignment is governed by the dominant boundary. Next, we explore the evolution of the alignment over the course of many experiments. To this end, we measure the orientation averaged over an entire cross-section. In a cylindrical geometry, the relative surface area of the cylindrical and flat (top and bottom) boundaries is given by the aspect ratio $A = \frac{h}{d(2\pi)}$.

In this case, we evaluate for the axial geometry,

$$O = \frac{\sum_{s=0}^{N_s} |\hat{n}_i \cdot \hat{z}|}{N_s}. \quad (2)$$

Here $N_s$ is the total number of surface points within each image ($N_s = \sum_{j=1}^{10} N_j$).

In fig. 3, we show the evolution of the alignment averaged over the entire cross-section as a function of aspect ratio. We display $O$ vs. $A$ for several axial crumpling and uncrumpling experiments. We point out a few distinct features of the progression: i) The increase in alignment with increase in aspect ratio occurs in all these figures. ii) The initial alignment varies considerably from run to run, and one complete loop of crumpling and uncrumpling does not take the conformation very far from the initial level of alignment. iii) At each aspect ratio we show two data symbols representing images taken immediately after a compression step, and just before the next step is taken. None of the averaged quantities we show are sensitive to the waiting time $\tau_w$ between the steps. Though the deformations are always elastic, and though there is no static friction in these experiments, there is hysteresis between initial and final states over a cycle.

Both small and large values of $A$ might be expected to create alignment by conforming to the developable flat and cylindrical surfaces by disordered accordion- and coil- or scroll-like motifs, respectively. In fig. 4, we confirm this expectation by combining results from both radial and axial crumpling experiments, spanning both sides of $A = 2$, which denotes a square cross-section. At small $A$,
Fig. 3: (Colour on-line) Alignment averaged over cross-section. We show $|\hat{n}_i \cdot \hat{z}|$ averaged over the entire cross-section, denoted by $C$, of an axially crumpled sheet. Data are shown for several instances of crumpling and uncrumpling cycles with the initial and final data points indicated with an open diamond and a cross, respectively. The aspect ratio, $A$, decreases towards the right on the horizontal axis, as shown by the rectangles. Orientation along the $z$-axis grows as $A$ decreases. The value of isotropic orientation in 2D is shown by the dashed line. Even though there is no plastic deformation, the sheet maintains memory of its initial configuration as it returns to a similar average alignment. The waiting time, $\tau_w$, has little effect on the evolution of orientation.

Fig. 4: (Colour on-line) Alignment averaged over the entire cross-section for all geometries. Shown is the alignment, $O$, for varying confinement geometries: radial, axial, axial with two sheets, and hemisphere (axial motion with a spherical cap instead of a flat piston) (red, blue, green, and pink, respectively) as a function of the aspect ratio, $A$. The aspect ratio, $A$, decreases towards the right on the horizontal axis, as shown by the rectangles. The value of isotropic orientation in 2D is shown by the dashed line.

Nematic order and stacking. – Thus far, we have only considered the alignment relative to space-fixed directions set by the normals to the cylinder boundaries. However, one may ask whether orientation is only a boundary effect, with a finite penetration depth, or whether there is a tendency to orient in bulk as the volume fraction is increased. In this latter scenario, the boundary merely breaks symmetry and chooses the direction of orientation.

We assess the development of bulk order within the volume, independently of any space-fixed direction, by computing two distinct two-point correlation functions, one that measures the mass distribution along the normal direction and the other that measures the relative orientation of normals.

The first of these is a straightforward generalization of the radial distribution function, except that the mass density (i.e., pixels with $m(\vec{x}) = 1$) is measured in the direction of the surface normal of every detected surface point. We start from every surface point, $\vec{x}_i$, and measure the mass at a distance $r$, away from the point in the direction of its surface normal, $\hat{n}(\vec{x}_i) = \hat{n}_i$.

$$C_m(r) = \frac{\sum_{i=0}^{N_S} m(\vec{x}_i \pm \hat{n}_i r)}{2 N_S} \quad |\vec{x}_i| \in S, \quad \hat{n}(\vec{x}_i) = \hat{n}_i$$

with $N_S$ equal to the total number of surface points in an entire cross-section.

The density correlation $C_m(r)$ does not account for relative orientation of the two surface points, and includes contributions from unaligned facets.

To account for the relative orientation of sheets in a stack, we compute the normal-normal correlation function as

$$C_n(r) = \frac{\sum_{i=0}^{N_S} |\hat{n}_i \cdot \hat{n}(\vec{x}_i \pm \hat{n}_i r)|}{1 + N_{liq}} \quad |\vec{x}_i| \pm \hat{n}_i d \in S, \quad \hat{n}(\vec{x}_i) = \hat{n}_i$$

where the total number of liquid points between each pair is $N_{liq}$. We use $N_{liq}$ as the denominator to emphasize stacked facets, with no liquid separating them.

In figs. 5(a) and (b) we show for a cycle of axial crumpling and uncrumpling the development of the density correlation, $C_m(r)$, and the normal-normal correlation, $C_n(r)$. There is a robust trend for both correlations to decay slower with increasing $\phi$, even at these rather low volume fractions. Corresponding correlation functions constructed from 2D sections of our earlier experiments [4] with aluminium foil crumpled into spheres are shown in the inset. In those experiments, the overall orientation of surface normals was found to be isotropic, unlike the aligned states achieved in PDMS sheets within a cylinder (cf. fig. 3). However, the evolution of correlations with volume fraction is similar, thus showing that the propensity for bulk orientational ordering persists, even in the absence of a boundary that promotes global alignment.
3.0 demonstrated that the opposite is true. The comparison of (a) with (b) demonstrates that as the aspect ratio, is to serve as the agent of symmetry breaking and choose a preferred orientation for the sheet normals. The orientational order has two related aspects: one of these is that the sheet develops nematic order, and the other is that the sheets stack closely. The normals do not form a dilute, dispersed nematic, but are inhomogeneously clustered in stacks. We are not aware of any other state of matter with closely analogous properties, which presumably arises from the combined nonlocal demands of self-avoidance and of connectivity. In our current experiments, we have not reached a regime of long-range order, but it would clearly be important to study how both these aspects of ordering progress at higher densities, and whether either or both of them lead to true phase transitions, analogous to those predicted [12] for the lower-dimensional system of an elastic rod confined in a 2D space.

Conclusion. – To summarize, we observe that both cylindrical and flat walls promote alignment of a crumpled sheet. These strongly aligned states show accordion-like or scroll-like motifs that may produce the more ordered states identified as folds in [20]. Furthermore, it appears that stacking into nematically ordered layers is not merely a boundary effect. The process of crumpling leads to the development of orientational order at remarkably low volume fractions of between 8% and 20%. The principal role of the walls, as quantified by the aspect ratio, is to serve as the agent of symmetry breaking and choose a preferred orientation for the sheet normals. The orientational order has two related aspects: one of these is that the sheet develops nematic order, and the other is that the sheets stack closely. The normals do not form a dilute, dispersed nematic, but are inhomogeneously clustered in stacks. We are not aware of any other state of matter with closely analogous properties, which presumably arises from the combined nonlocal demands of self-avoidance and of connectivity. In our current experiments, we have not reached a regime of long-range order, but it would clearly be important to study how both these aspects of ordering progress at higher densities, and whether either or both of them lead to true phase transitions, analogous to those predicted [12] for the lower-dimensional system of an elastic rod confined in a 2D space.

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