Ribbon Formation in Twist-Nematic Elastomers

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1. Helicoid to Ribbon

1.1 Formation of twisted ribbons consisting of bilayers of gemini surfactants (two surfactant molecules covalently linked at their charged head groups; here 16-2-16 tartrate at 0.1% in water; horizontal span ~ 10 μm).

1.2 It is observed a smooth transition from **platelet to helix to ribbon** (tubule in the picture)

![Diagram of Tubule, Helix A, Platelet, Helix B](image)

1.3 How does the shape of the twisted ribbons arise from the particular molecular structure of the amphiphiles?

1.4 This is a long lasting story ...

W. Helfrich, J. Prost.  
**Intrinsic bending force in anisotropic membranes made of chiral molecules.**  

**Tuning bilayer twist using chiral counterions.**  

R. Ghafouri, R. Bruinsma.  
**Helicoid to Spiral Ribbon Transition.**  

E. Efrati, E. Sharon, R.Kupferman.  
**Buckling transition and boundary layer in non-Euclidean plates.**  

Y. Sawaa, F. Ye, K. Urayama, T. Takigawa, V. Gimenez-Pinto, R.L.B. Selinger, J.V. Selinger,  
**Shape selection of twist-nematic-elastomer ribbons.**  
1.5 ... and here is what happens to Twist-Nematic Elastomers

Y. Sawaa, F. Ye, K. Urayama, T. Takigawa, V. Gimenez-Pinto, R.L.B. Selinger, J.V. Selinger,
Shape selection of twist-nematic-elastomer ribbons.
2. Nematic Elastomers

2.1 Nematic elastomers exhibit large distortions of a special kind: if the stress-free shape of a mesoscopic chunk of NE is a spherical ball when the appended mesogens are in the disordered, isotropic phase (left), its stress-free shape in the ordered, nematic phase is a spheroid whose polar axis is aligned with the prevailing mesogen direction (right).
3. Isotropic-nematic Phase Transitions

3.1 Nematic direction is represented by

\[ \mathbf{N} = \mathbf{n} \otimes \mathbf{n}, \quad \text{with } \mathbf{n} \text{ a unit vector, called director} \]

3.2 Nematic distortions are then represented by the tensor

\[ \mathbf{U}_o = \lambda_\parallel \mathbf{N} + \lambda_\perp (\mathbf{I} - \mathbf{N}), \quad \lambda_\perp = \sqrt{\frac{J_o}{\lambda_\parallel}}, \quad J_o = \det (\mathbf{U}_o) \]

Phase diagram of a typical NE: strains versus temperature \((J_o = 1)\).
4. Elastic Strain

4.1 Given a volume element $dV$, the elastic deformation $F_e$ measures the difference between its distorted image $dv_o = U_o dV$ and its actual state $dv = F dV$.

$$dv = F dV = F_e dv_o$$

$F_e = F U_o^{-1}$

this is the “further strain”

4.2 The elastic energy $\varphi$ has to be a function of the elastic strain $C_e = F_e^T F_e$:

5. Preparation

5.1 Specimens are prepared in the nematic & wet state, and are initially flat. The nematic configuration is imprinted in the elastomer matrix by the cross-linking reaction in the presence of a nonreactive dopant, and appropriate glass substrates coated with uniaxially rubbed layer.

5.2 The specimen undergoes an anisotropic de-swelling (irreversible) and a temperature-controlled nematic-to-isotropic phase transition (reversible)

\[
\text{de-swelling} \quad \to \quad \text{heating} \quad \to \quad \text{isotropic state}
\]

\[
\begin{align*}
\sim 50\% \, \text{volume reduction} & \quad v = v_\text{d}, \vartheta = \vartheta_p \\
\text{preparation state} & \quad v = 1, \vartheta = \vartheta_p
\end{align*}
\]

5.3 Nematic distortions are then represented by the tensor

\[
U_o = \frac{\lambda_{\parallel}(\vartheta) \alpha_{\parallel}(v)}{\lambda_{\parallel}(\vartheta_p)} N + \frac{\lambda_{\perp}(\vartheta) \alpha_{\perp}(v)}{\lambda_{\perp}(\vartheta_p)} (I - N).
\]
5.4 Nematic-isotropic transition is volume preserving, de-swelling is not:
\[
\lambda_{\parallel}(\vartheta) \lambda_{\perp}^2(\vartheta) = 1, \quad \alpha_{\parallel}(v) \alpha_{\perp}^2(v) = v.
\]

5.5 Let us have a look at the resultant strains:
\[
\lambda_{\parallel}(\vartheta, v) = \frac{\lambda_{\parallel}(\vartheta) \alpha_{\parallel}(v_d)}{\lambda_{\parallel}(\vartheta_p)}, \quad \lambda_{\perp}(\vartheta, v) = \frac{\lambda_{\perp}(\vartheta) \alpha_{\perp}(v_d)}{\lambda_{\perp}(\vartheta_p)}.
\]
6. Chiral

6.1 Chiral geometry: $N$ is on horizontal planes

\[ \theta = \theta(z) \]
\[ n(\theta) = \cos(\theta) e_1 + \sin(\theta) e_2 \]
\[ N(\theta) = n(\theta) \otimes n(\theta) \]
\[ U_o(\theta) = \lambda_\parallel N(\theta) + \lambda_\perp (I - N(\theta)) \]
\[ C_o(\theta) = \lambda_\parallel^2 N(\theta) + \lambda_\perp^2 (I - N(\theta)) \]

6.2 What is the realized configuration?
6.3 The elastic strain must accommodate non-homogeneous and non isotropic distortions; we study two chiral geometries:

L-geom:  
at midplane director $\parallel$ axis

S-geom:  
at midplane director $\perp$ axis

6.4 There are two strategies: twist or bend; the transition from one shape to the other is sharp.
7. L- & S-Geometry

7.1 The handedness is determined by the torsion $b_{o12}$:

$$b_{o12} > 0 \Rightarrow \text{right-handed};$$

$$b_{o12} < 0 \Rightarrow \text{left-handed.}$$

$$b_{o12} = \begin{cases} \frac{1}{2} (\lambda^2_\parallel - \lambda^2_\perp) & \text{L-geometry;} \\ \frac{1}{2} (\lambda^2_\perp - \lambda^2_\parallel) & \text{S-geometry.} \end{cases}$$

![Graph showing the transition between different states and geometries with respect to torsion $b_{o12}$](graph.png)
7.2 Helicoid to Ribbon

\[ \nabla \] smooth transition

\[ \nabla \] buckling

8. Shape Formation

8.1 Shape transition is dependent on the ratio:

\[
\frac{\text{torsional stiffness}}{\text{bending stiffness}} \propto \frac{\text{width}}{\text{height}}
\]

\[\lambda_\parallel\] for helicoid
\[\lambda_{\perp}\] for ribbon

- prep. state
- de-swelling
- heating
- isotropic state

shape transition

Helicoid
Ribbon
Helicoid
Helicoid
Ribbon
9. Shape Transition

narrow
W/H=13

medium
W/H=20

wide
W/H=40
9.1 Narrow VS Wide bars

![Diagram showing the comparison between narrow and wide helicoids and ribbons in different states: dry state, flat state, and isotropic state. The diagrams illustrate the de-swelling and heating processes for both narrow and wide structures, with measurements indicated on the y-axis.](image-url)
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