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 $\sim 10 \, \mu m$).



R. Oda, I. Huc, M. Schmutz, S.J. Candau, F.C. MacKintosh. Tuning bilayer twist using chiral counterions. NATURE, vol. 399, 1999.

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



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

R. Oda, I. Huc, M. Schmutz, S.J. Candau, F.C. MacKintosh. **Tuning bilayer twist using chiral counterions**. NATURE, vol. 399, 1999. 1.4 This is a long lasting story ...

W. Helfrich, J. Prost.

Intrinsic bending force in anisotropic membranes made of chiral molecules.

Physical Review A, vol. 38, n. 6, 1998.

R. Oda, I. Huc, M. Schmutz, S.J. Candau, F.C. MacKintosh. **Tuning bilayer twist using chiral counterions**. NATURE, vol. 399, 1999.

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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**. PNAS, vol. 108, n. 16, 2011.

1.5 \ldots 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.

PNAS, vol. 108, n. 16, 2011.

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).



Isotropic phase

Nematic phase

3. Isotropic-nematic Phase Transitions

3.1 Nematic direction is represented by

 $\mathbf{N} = \mathbf{n} \otimes \mathbf{n}$, with \mathbf{n} a unit vector, called **director**

3.2 Nematic distortions are then represented by the tensor



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 \mathbf{F}_e measures the difference between its distorted image $dv_o = \mathbf{U}_o dV$ and its actual state $dv = \mathbf{F} dV$.



4.2 The elastic energy φ has to be a function of the elastic strain $\mathbf{C}_e = \mathbf{F}_e^{\top} \mathbf{F}_e$:

Y. Sawaa, K. Urayama, T. Takigawa, A. DeSimone, L. Teresi, Thermally Driven Giant Bending of Liquid Crystal Elastomer Films with Hybrid Alignment, Macromolecules, 2010.

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)



5.3 Nematic distortions are then represented by the tensor

$$\mathbf{U}_{o} = \frac{\lambda_{\parallel}(\vartheta) \,\alpha_{\parallel}(v)}{\lambda_{\parallel}(\vartheta_{p})} \,\mathbf{N} + \frac{\lambda_{\perp}(\vartheta) \,\alpha_{\perp}(v)}{\lambda_{\perp}|(\vartheta_{p})} \left(\mathbf{I} - \mathbf{N}\right).$$

5.4 Nematic-isotropic transition is volume preserving, de-swelling is not:

$$\lambda \|(\vartheta) \, \lambda_{\perp}^2(\vartheta) = 1 \,, \quad \alpha \|(v) \, \alpha_{\perp}^2(v) = v \,.$$

5.5 Let us have a look at the resultant strains:



6. Chiral

6.1 Chiral geometry: **N** is on horizontal planes

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:



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} :



7.2 Helicoid to Ribbon



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, PNAS, 2011.

8. Shape Formation

8.1 Shape transition is dependent on the the ratio:

torsional stiffness	\sim	width
bending stiffness	a	height



9. Shape Transition





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Y. Sawaa, K. Urayama, T. Takigawa, A. DeSimone, L. Teresi, **Thermally Driven Giant Bending of Liquid Crystal Elastomer Films with Hybrid Alignment**, Macromolecules, 2010.