

### Cubic Invariant for Artificial Muscles

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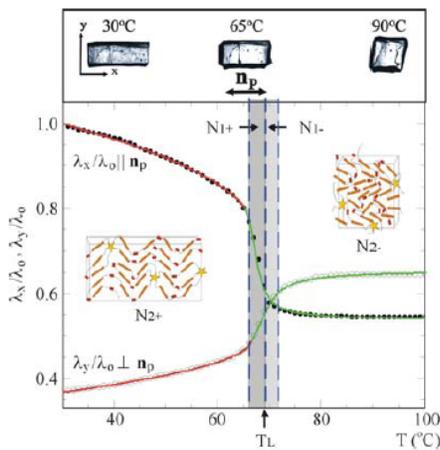


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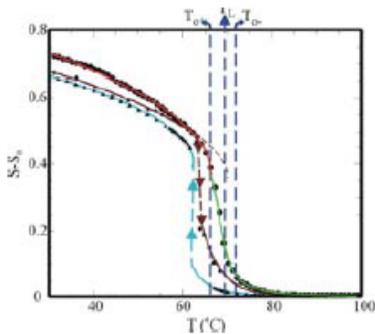


Fig. 2. S-S<sub>0</sub> from [1] and independent birefringence measurements increasing T, t decreasing T [2].

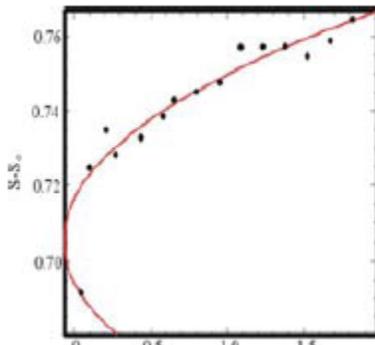


Fig. 3. S-S<sub>0</sub> vs. strain [2]

Liquid crystals are characterized by an axis of orientational order,  $\mathbf{n}_p$ , where  $|\mathbf{n}_p|=1$ . Liquid crystalline elastomers support shear while low molecular weight liquid crystals flow. In monodomain elastomers, supercritical phase transitions with small latent heats result in dramatic muscle action because spontaneous shape changes are in a narrow temperature range. Supercritical first order shape change transitions unfold beautifully in the polarizing microscope (Fig. 1) [1]. The muscle transition for this main chain elastomer [2] occurs in a 5K temperature interval around the temperature,  $T_L \approx 70^\circ\text{C}$  where the cubic invariant,  $I_3$ , changes sign at a continuous uniaxial positive nematic (N1+) to uniaxial negative nematic (N1-) transition. A single Lorentzian connects the constant volume positive biaxial nematic, N2+ to the constant volume negative biaxial nematic, N2-. The two Lorentzians of  $dI_3/dT$ , one for N2+  $\leftrightarrow$  N1+ and the other for N1-  $\leftrightarrow$  N2- merge at  $T_L$ , which is approximately the cross-linking temperature,  $T_{CL}$ . Siloxane cross-linking at  $T_{CL} \approx 60^\circ\text{C}$ , integrates quadratic uniaxial orientational order,  $I_2$ , of the liquid crystalline units into one  $\mathbf{n}_p$  for the elastomer. Uniaxiality for  $\mathbf{n}_p$  is guaranteed in the vicinity of  $T \approx T_{CL}$ . Far from  $T_{CL}$ ,  $\mathbf{n}_p$  is biaxial. Consequently, LCEs require the general description for nematics that includes quadratic,  $I_2$ , and cubic,  $I_3$ , invariants [3]. This introduces a new transition at temperature  $T_L \sim T_{CL} \neq T_{NI}$  where  $I_3$  changes from positive to negative. The “L” in  $T_L$  is for Larkin. The free energy is  $\Phi_{LCE} = -\frac{1}{2}(q_L/q_{NI})(T_L - T_{NI})^2/(T_L - T_{NI})(S - S_0)^2$  where  $q_L/q_{NI}$  is the ratio of the heats of transition at  $T_L \approx 70^\circ\text{C}$  and  $T_{NI} \approx 125^\circ\text{C}$ . So is the Lorentzian base line and  $(S - S_0)^2 = I_3 \cdot 2/3 = 0.55 I_2$ . Fig. 2 shows S-S<sub>0</sub> for the spontaneous shape change in Fig. 1 and for independent birefringence measurement [2]. Fig. 3 shows S-S<sub>0</sub> for relative strain,  $\lambda \parallel \mathbf{n}_p$  at  $T \approx 25^\circ\text{C}$ :  $(S - \sqrt{2}/2)^2 = (\lambda \parallel - 0.957)/\lambda$  [2]. Thus,  $S = S_0 = \sqrt{2}/2$  when  $\lambda \parallel = 0.957$  (not 1) because of spontaneous shape change.

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2. Simon Krause, *Nematische Hauptkettenelastomere Synthese und Untersuchung der mechanischen Eigenschaften und der Ordnungszustandes*, Albert-Ludwigs-Universität Freiburg, Juli (2008).
3. P.B. Vigman, A.I. Larkin and V.M. Filev, *Sov.Phys. JETP* **41**, 944 (1976).