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Sliding friction of nematic elastomers

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Abstract

We discuss steady-state mechanical dissipation at the interface between a nematically ordered elastomer and a substrate of hard asperities. A 'resin-like' approximation is invoked, according to which the nematic director is assumed to be rigidly coupled to the strain tensor in the elastomer. The resulting nematic correction to the friction coefficient scales with the degree of nematic order as Q^2. If the elastomer intercedes at a sheared substrate–fluid interface, there is an associated shortening of the effective hydrodynamic slip length seen by the fluid. This scenario is relevant to the biological functioning of slug pedal mucus.

1. Introduction

At a sheared interface between a viscoelastic medium and a rough substrate there is in general a time-oscillatory strain response in the medium, which contributes to overall sliding friction. Theoretical and experimental studies have shown this contribution to be particularly significant for rubber, due to the combination in this case of low elastic moduli with high viscosity constants [1, 2].

Rubber-like orders of magnitude are broadly reflected in the respective viscous and elastic constants of nematic elastomers [3], so one expects to observe sliding friction of similar origin. On the other hand, nematic ordering considerably enriches the viscoelastic characterization of a medium in respect of the number of constants involved. A generalized account of sliding friction taking all these into account would be a formidable undertaking, and will not be our objective here. We present instead a very simple qualitative perspective, according to which nematic ordering manifests itself via a perturbative modification of one of the rubber viscosity constants.

2. Qualitative viscoelasticity

A defining characteristic of nematic LCs is their ability to transmit a torque. In a bulk nematic fluid, the director field $\boldsymbol{n}(\boldsymbol{r})$ about a concentrated torque $\boldsymbol{\Gamma}$ at the origin is perturbed as [4]

$$\delta\boldsymbol{n}(\boldsymbol{r}) \sim \boldsymbol{\Gamma}/K r, \tag{1}$$

where K denotes the Frank constants governing director elasticity.

In a nematic elastomer, this response is damped by local anchoring of the director to a cross-link matrix [3, 5]. Conversely, *strain* elastic response to an external force $\boldsymbol{F}$ is damped by concomitant director deformation. However, since the Frank elastic terms are second order with respect to strain elasticity, this latter effect is negligible. Thus, for small strains in the classical Hookean regime, we can write as usual for the displacement field in response to a concentrated force at the origin [6]

$$\boldsymbol{u}(\boldsymbol{r}) \sim \boldsymbol{F}/Er, \tag{2}$$

where E is Young's modulus.

We neglect here the general anisotropy in E which characterizes a nematic elastomer. Moreover, we will assume strong anchoring of the director to the cross-link matrix, to the extent that the director is rigidly coupled to the strain tensor u_{ij} in a linear reciprocity relation $\delta\boldsymbol{n} \sim u_{ij}$, again valid only for small strains. In this rigid-coupling sense, we are effectively treating the medium as a nematic 'resin' [3].

These simplifications entail (i) that the director responds to a displacement force $\boldsymbol{F}$ as

$$\delta\boldsymbol{n}(\boldsymbol{r}) \sim \boldsymbol{F}/Er^2, \tag{3}$$

and (ii) that nematic dissipation of the form $\gamma(\partial_t \boldsymbol{n})^2$, where γ has the dimensions of a viscosity, can be expressed equivalently as a translational viscous dissipation $\eta\,\partial_t u_{ij}\,\partial_t u_{kl}$. We neglect further anisotropic characterization of the viscosity.

3. Nematic contribution to sliding friction coefficient

Consider the sub-surface dissipation in an elastic medium as it slides with uniform velocity v_s along, say, the x-axis of a Cartesian coordinate frame. At the origin, a single hard asperity penetrates the plane $z = 0$ of the surface, the tip exerting a concentrated perpendicular force F_z on the medium.

In the linear elastic regime, and in the absence of nematic order, the dissipation density is governed by two viscosity coefficients η and ζ [6]:

$$\dot{\epsilon}_{\rm iso} = 2\eta(\partial_t u_{ik} - \tfrac{1}{3}\delta_{ik}\,\partial_t u_{ll})^2 + \zeta(\partial_t u_{ll})^2 = v_s^2[2\eta(\partial_x u_{ik} - \tfrac{1}{3}\delta_{ik}\,\partial_x u_{ll})^2 + \zeta(\partial_x u_{ll})^2] \tag{4}$$

(with summation over repeated indices)

Substituting equation (2) and neglecting the difference between η and ζ, the volume integration giving the total dissipation around the asperity tip has the form

$$\int_a \dot{\epsilon}_{\rm iso}\,{\rm d}V \sim \frac{\eta v_s^2 F_z^2}{E^2 a^3}, \tag{5}$$

where we identify the length scale a with the diameter of the tip.

Hence, an isotropic elastic medium sliding over a rough surface comprising a density n of identical asperities is resisted by an effective friction force

$$\frac{nA}{v_s}\int_a \dot{\epsilon}_{\rm iso}\,{\rm d}V \sim \frac{\eta v_s L^2}{nE^2 a^3 A}, \tag{6}$$

where A is the area and L the load supported by the asperities. Dividing this force by the load, we define the sliding friction coefficient [2]

$$\mu_{\rm iso} \sim \frac{\eta v_s L}{nE^2 a^3 A}. \tag{7}$$

An analogous nematic contribution follows by expanding in $(\gamma - \eta)$. For example, if the director is oriented along the z-axis ('homeotropically'), then for small strains

$$n_z \simeq 1 \qquad \text{and} \qquad n_i \simeq u_{iz}; \qquad i = x, y \tag{8}$$

such that

$$\gamma(\partial_t \boldsymbol{n})^2 \simeq \gamma v_s^2 \sum_{i=x,y} (\partial_x u_{iz})^2. \tag{9}$$

Putting

$$\dot{\epsilon} = \dot{\epsilon}_{\rm iso} + \delta\dot{\epsilon}_{\rm nem}, \tag{10}$$

with

$$\delta\dot{\epsilon}_{\rm nem} \simeq (\gamma - \eta) v_s^2 \sum_{i=x,y} (\partial_x u_{iz})^2, \tag{11}$$

volume integration yields a nematic component of the friction coefficient:

$$\delta\mu_{\rm nem} \sim (\gamma - \eta) \frac{v_s L}{nE^2 a^3 A}. \tag{12}$$

By analogy with the prediction $\gamma \sim Q^2$ which follows from the Landau–de Gennes theory of thermotropic nematic fluids [4], with Q the nematic order parameter, we postulate

$$\frac{\gamma - \eta}{\eta} \sim Q^2, \tag{13}$$

yielding

$$\delta\mu_{\rm nem} \sim \frac{Q^2 \eta v_s L}{nE^2 a^3 A}. \tag{14}$$

Nematic–isotropic (NI) transitions are ordinarily first order, with typically $\Delta Q \simeq 0.3$. We expect therefore a corresponding discontinuity in the friction coefficient with the onset of nematic order [7].

4. NI-switched hydrodynamic slip length

A related point can be made concerning effective *hydrodynamic slip* if an elastomer film intercedes at a rough substrate shearing a fluid phase. We have in mind the established slippage-modifying effects of grafting a polymer monolayer at a sheared substrate–fluid interface [8].

Consider the laminar flow profile $v(z)$ depicted in figure 1. The thickness of the film is assumed to be large with respect to a, such that the elastomer–fluid interface ('i') can be supposed flat and undeformed by the influence of the asperities at the opposing substrate–elastomer interface. As above, the substrate is stationary in this frame of reference, the film moving tangentially along the x-axis with uniform velocity v_s. The boundary condition on tangential flow is assumed to be 'no slip' [9], i.e., $v(z_i) = v_s$ in our picture. With respect to the substrate, however, this entails a finite effective *fluid–substrate* slip length d defined by

$$d \left.\frac{{\rm d}v}{{\rm d}z}\right|_i = v_s. \tag{15}$$

We obtain d from the steady-state criterion that viscous stress at the z_i-interface, $\sigma_{xz} = \eta_f \, {\rm d}v/{\rm d}z$, where η_f denotes the fluid viscosity, is balanced by the film–substrate friction stress. Using the expressions of section 3,

$$d_{\rm iso} \sim \left(\frac{\eta_f}{\eta}\right) na^3 (EA/L)^2, \qquad \delta d_{\rm nem} \sim -Q^2 \left(\frac{\eta_f}{\eta}\right) na^3 (EA/L)^2. \tag{16}$$

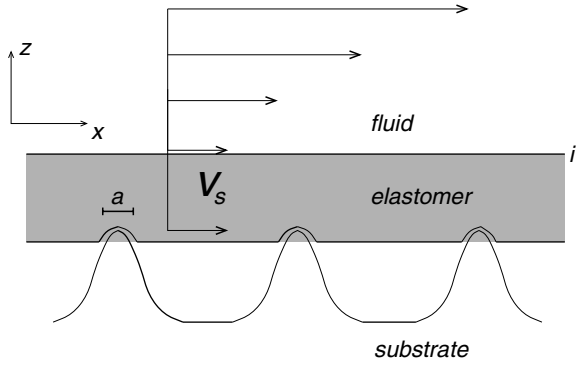

Figure 1. The schematic diagram of the shearing discussed in section 4 of the text. Although the fluid sees no-slip boundary conditions with respect to the elastomer, dissipation at the asperity–elastomer contacts results in an effective hydrodynamic slip length with respect to the substrate.

In the limit of a large load L, hence high friction, d goes to zero and we recover a no-slip effective substrate–fluid boundary condition $v(z_i) = v_s = 0$. Less obviously, slippage increases with the area of real contact $\sim nAa^2$ between the substrate and the film, and with the elastic modulus E.

5. An example from nature: slug locomotion

Mucous secretions are glycoprotein hydrogels, in which the polymer component comprises entangled protein backbones and rod-like polysaccharide side chains. Experimental evidence has suggested that a degree of nematic ordering of the side chains takes place *in vivo*, apparently associated with higher concentration local to a surface [11], such that mucus may sometimes be regarded as a nematic elastomer, at least locally near a surface. The particular type of mucus with which a slug supports itself is known as the 'pedal' mucus. During locomotion, the pedal mucus layer is thought to undergo a 'yield–heal' cycle, alternating between elastic and fluid rheological response [10].

It is interesting in this respect to tentatively interpret mucus–substrate shear mechanics during the 'yield' part of the cycle according to our schematic figure. That is, we assume that a nematically ordered viscoelastic film persists immediately adjacent to the substrate, while the bulk of the pedal layer yields as fluid flow. According to our equation (16), nematic order is associated with enhanced hydrodynamic grip during this part of the yield cycle, a property which might have a functional impact.

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