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# The interplay between boundary conditions and flow geometries in shear banding: hysteresis, band configurations, and surface transitions

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We study shear banding flows in models of wormlike micelles or polymer solutions, and explore the effects of different boundary conditions for the viscoelastic stress. These are needed because the equations of motion are inherently non-local and include “diffusive” or square-gradient terms. Using the diffusive Johnson-Segalman model and a variant of the Rolie-Poly model for entangled micelles or polymer solutions, we study the interplay between different boundary conditions and the intrinsic stress gradient imposed by the flow geometry. We consider prescribed gradient (Neumann) or value (Dirichlet) of the viscoelastic stress tensor at the boundary, as well as mixed boundary conditions in which an anchoring strength competes with the gradient contribution to the stress dynamics. We find that hysteresis during shear rate sweeps is suppressed if the boundary conditions favor the state that is induced by the sweep. For example, if the boundaries favor the high shear rate phase then hysteresis is suppressed at the low shear rate edges of the stress plateau. If the boundaries favor the low shear rate state, then the high shear rate band can lie in the center of the flow cell, leading to a three-band configuration. Sufficiently strong stress gradients due to curved flow geometries, such as that of cylindrical Couette flow, can convert this to a two-band state by forcing the high shear rate phase against the wall of higher stress, and can suppress the hysteresis loop observed during a shear rate sweep.

## I. INTRODUCTION

Microscopic models for certain viscoelastic fluids, such as wormlike micelles [1] and high molecular weight polymeric liquids [2, 3] predict unstable homogeneous stationary states for controlled average shear rates. Specifically, the coupled equations of motion for the fluid flow and microstructural quantities, such as the viscoelastic stress  $\Sigma$  or orientational order, predict a nonmonotonic *constitutive curve*, *i.e.* the steady state total shear stress  $T_{xy}$  (or equivalently the applied torque) as a function of applied shear rate  $\dot{\gamma}$  for homogeneous flows. This constitutive curve typically is multivalued with a region of decreasing shear stress as a function of increasing shear rate, which is a hallmark of hydrodynamic instability [4]. Understanding this nonlinear behavior is important for practical applications such as injection moulding of plastics and drilling muds used in bore holes [5]. The simplest resolution of the instability is an inhomogeneous state with macroscopic regions of high and low shear rates, known as shear bands [6, 7], which has been widely observed in wormlike micelles [8, 9, 10, 11], lamellar surfactant solutions [12], and liquid crystals [13]. The resulting experimental signature of this inhomogeneous state is a stress plateau as a function of controlled average shear rate. We denote this experimentally determined relation between total shear stress and average shear rate as the *flow curve*; in shear banding flow this curve incorporates inhomogeneous flows and is distinct from the constitutive

curve, which cannot be measured because of the constitutive instability. In Fig. 1 the dashed line shows the constitutive curve while the triangles show flow curves that could be measured upon either increasing or decreasing average shear rate ramps.

Experimentally, it is clear that the geometry of the shear cell affects the positioning of the shear bands. The simplest structure is seen in the cylindrical Couette geometry where two bands form, with the higher shear rate band near the inner wall (rotor) [14]. In the cone and plate geometry a different band configuration has been reported: two low shear rate regions next to the cone and plate separated by a high shear rate region [9]. One explanation offered for that result was that secondary flows stabilized the center band [15]. We will show that a boundary condition that prescribes a value for the polymeric stress tensor similar to that of the low shear rate phase can induce this three-band configuration.

The experimental rheological features of shear banding have been studied in detail for wormlike micellar solutions [10, 11, 16]. A constitutive curve such as the dashed line in Fig. 1 can only be inferred because the negative slope region is unstable; however, the experimentally measured flow curve, indicated by triangles, typically shows a stress plateau which spans the range of average applied shear rates at which the system shows shear banding. During shear rate ramps from rest, hysteresis is often observed at the start of this plateau: the stress increases past the plateau stress and follows the

constitutive curve until, at a time that depends on the rate of the ramp, a high shear rate band develops and the total stress decreases onto the steady state plateau [10, 11, 17]. However, if the shear rate is reduced from a point on the plateau, the low shear rate branch of the constitutive curve is intersected directly by the flat plateau. This behavior is reminiscent of nucleation and hysteresis at first order phase transitions, and we will show below that the boundary conditions can influence this hysteresis, in a manner analogous to heterogeneous nucleation.

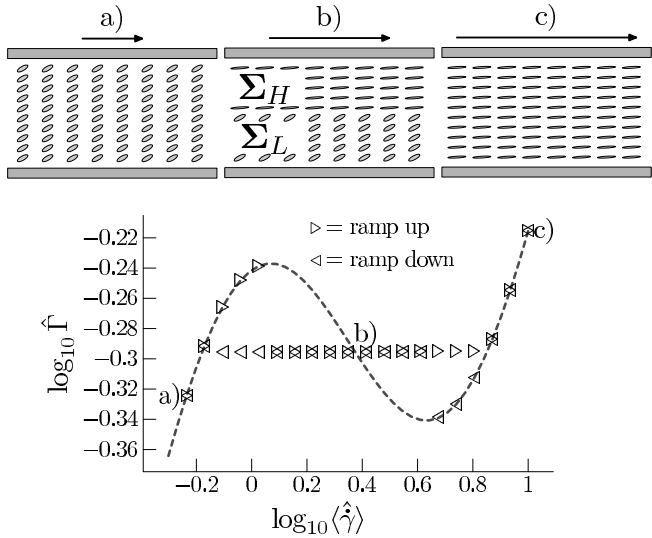


FIG. 1: Dashed line: the constitutive curve (dimensionless specific torque  $\hat{\Gamma}$  as a function of dimensionless shear rate  $\hat{\gamma}$ ) for the diffusive Johnson-Segalman (DJS) model in cylindrical Couette flow. The triangles show simulated steady state flow curves for zero gradient (Neumann) boundary conditions, for increasing ( $\triangleright$ ) and decreasing ( $\triangleleft$ ) the average shear rate ( $\langle \hat{\gamma} \rangle$ ) from small or large average shear rates respectively. The model parameters are  $a = 0.3$ ,  $\epsilon = 0.05$ ,  $\hat{D} = 10^{-5}$ , and the gap size is 0.1% of the radius ( $q = 0.005$ ). The physical difference between the two bands is illustrated by ellipsoids whose principal axes' directions and length ratios coincide with those of the viscoelastic stress tensor  $\Sigma_L$  and  $\Sigma_H$  in, respectively, the low and high shear rate phases.

In modelling shear banding systems such as wormlike micelles the most important degrees of freedom are the fluid velocity  $\mathbf{v}$  and a viscoelastic contribution to the stress,  $\Sigma$ ; here we neglect other potentially important quantities such as concentration and micellar length. Among models with non-monotonic constitutive curves for homogeneous flow, spatially local ones such as the Johnson-Segalman model in its original form [18] show high sensitivity to the flow history and do not give the well-defined and unique stress plateau observed in experiments [19]. In contrast, if a non-local diffusive term (typically proportional to  $\nabla^2 \Sigma$ ) is included in the constitutive equation for the viscoelastic stress (*e.g.* the diffusive Johnson-Segalman (DJS) model), there is only a single value of the total stress for which the interface between

bands is stable and stationary [20]. The diffusive term arises from microscopic physical mechanisms such as the diffusion of molecules that carry stress [21], the persistence length of wormlike micelles [22], or hydrodynamics [23, 24]. The resulting equation governing inhomogeneous steady states is thus a spatial differential equation for the viscoelastic stress, which necessitates a boundary condition that is, at present, unknown. The most frequently used boundary condition on the viscoelastic stress  $\Sigma$  has been that of zero stress gradient parallel to the boundary normal [20]. This boundary condition predicts the flow curve shown in Fig. 1 for the DJS model. As noted above, this flow curve shows hysteretic behavior at the start of the stress plateau similar to that seen in experiment [11], with a stress overshoot during an increasing shear rate ramp.

Other theoretical studies of shear banding have used a fixed value of the polymeric stress [25, 26, 27] at the wall (Dirichlet boundary conditions). Cook and Rossi considered a two-fluid model for wormlike micelles in which the micelles were assumed to align at the wall parallel to the flow direction, in both planar and cylindrical Couette flow. They found that the flow curve deviates significantly from the constitutive curve near the low shear rate branch [25, 26], exhibited hysteresis only at the high shear rate side of the stress plateau, and possessed a three-band state with the boundary condition induced high shear rate bands near the wall. Qualitatively similar results were found by Picard and co-workers in a scalar model for shear banding in a yield stress solid [27].

In this paper we perform a more detailed study of the effects of different boundary conditions for the viscoelastic stress at the wall,  $\Sigma_0$ . We consider strong “anchoring” in which Dirichlet conditions apply, and interpolate between oblate and prolate forms of  $\Sigma_0$ . We also study mixed boundary conditions, in which  $\Sigma_0$  is determined by a balance between spatial gradients and surface anchoring. Moreover, we address the important interplay between the boundary conditions and intrinsic inhomogeneity of the flow determined by the flow geometry (to compare cylindrical and planar Couette flow with cone and plate flow, for example).

The organization of the paper is as follows. In Section II we outline the details for solving the equations of motion within the creeping flow approximation in cylindrical Couette flow, and describe two constitutive equations for the viscoelastic stress: the DJS model [20] and a tube-based model which has been developed for polymer solutions and wormlike micelles [28, 29]. The physical motivation for the boundary conditions is discussed in Section III. After reviewing previous results for Neumann boundary conditions in Section V, the main new results are presented in Section VI. We organize these as follows: (1) the effect of different fixed value (Dirichlet) boundary conditions on the flow curves and hysteresis upon increasing and then decreasing the average shear rate; (2) the interplay between boundary conditions and the stress gradient imposed by the flow geometry (*e.g.*

cylindrical Couette flow) and their effects on the stable position of the shear bands; and (3) the effect of mixed boundary conditions, in which the viscoelastic stress at the surface is influenced by both the bulk constitutive relation and the flow geometry. We demonstrate the possibility of a transition between different effective boundary conditions as a function of the anchoring strength.

## II. EQUATIONS OF MOTION

### A. Creeping Flow Approximation

In a wormlike micellar system we assume that the total stress  $\mathbf{T}$  can be separated into contributions from the Newtonian solvent and a viscoelastic stress  $\mathbf{\Sigma}$  from the micelles:

$$\mathbf{T} = 2\eta\mathbf{D} + \mathbf{\Sigma} - p\mathbf{I}. \quad (\text{II.1})$$

Here  $\mathbf{I}$  denotes the identity matrix,  $\eta$  is the solvent shear viscosity,  $\mathbf{D} = \frac{1}{2} [\nabla\mathbf{v} + (\nabla\mathbf{v})^T]$  is the symmetric velocity gradient tensor, and  $p$  is the isotropic pressure determined by incompressibility ( $\nabla \cdot \mathbf{v} = 0$ ). We are interested here in the creeping flow regime (low Reynolds number), for which the force balance has the form

$$\nabla \cdot \mathbf{T} = 0. \quad (\text{II.2})$$

We perform calculations explicitly for Couette flow between concentric cylinders and assume unidirectional flow  $\mathbf{v} = v(r, t)\hat{\theta}$ , where  $r$  and  $\theta$  are the usual cylindrical coordinates. In this case, Eqs. (II.1-II.2) imply that

$$\eta\dot{\gamma}(r, t) + \Sigma_{r\theta}(r, t) = \frac{\Gamma}{r^2}, \quad (\text{II.3})$$

where

$$\dot{\gamma}(r, t) = r \frac{\partial}{\partial r} \frac{v(r, t)}{r}, \quad (\text{II.4})$$

and  $\Gamma$ , the specific torque per unit cylinder height per radian, is a constant of integration. We assume no-slip boundary conditions on the velocity field. For cylindrical Couette flow with a fixed outer cylinder and a rotating inner cylinder of velocity  $V$ , the no-slip boundary conditions lead to the following global constraint

$$\frac{V}{R_1} = \int_{R_2}^{R_1} \dot{\gamma}(r, t) \frac{dr}{r}, \quad (\text{II.5})$$

which implies that the specific torque  $\Gamma$  is given by

$$\Gamma \frac{R_1^2 - R_2^2}{R_1^2 R_2^2} = 2 \left( \frac{\eta V}{R_1} - \int_{R_1}^{R_2} \Sigma_{r\theta} \frac{dr}{r} \right). \quad (\text{II.6})$$

To simplify the equations we define the spatial variable

$$x = \frac{1}{q} \ln \left( \frac{r}{R_1} \right), \quad (\text{II.7})$$

where  $q = \ln \frac{R_2}{R_1}$ . Making use of this variable change, the creeping flow equation becomes

$$\eta\dot{\gamma} = \Gamma e^{-2qx} - \Sigma_{r\theta}, \quad (\text{II.8})$$

with the specific torque is given by

$$\Gamma = \frac{2q}{1 - e^{-2q}} (\langle \Sigma_{r\theta} \rangle - \eta \langle \dot{\gamma} \rangle), \quad (\text{II.9})$$

where the spatial averages are simplified to  $\langle (\cdot) \rangle = \int_0^1 (\cdot) dx$ . This expression can be used to calculate  $\Gamma$  for an imposed average shear rate, and hence render Eq. (II.8) an integral equation determining the relation between the local and average shear rates and viscoelastic shear stress. A complete description of the dynamics requires an equation of motion for the viscoelastic stress  $\mathbf{\Sigma}$ , examples of which are described below.

### B. Constitutive Equations

#### 1. Diffusive Johnson-Segalman (DJS) Model

One of the simplest tensorial models that produces a non-monotonic constitutive curve is the DJS model, which has been studied in detail [18, 19, 20, 30]. Physically it is motivated by neglecting all but the lowest modes of vibration of polymer chains, thereby representing them as elastic dumbbells with span  $\mathbf{R}$  [31]. The viscoelastic stress tensor  $\mathbf{\Sigma}$  is related to the extension of the dumbbells by  $\mathbf{\Sigma} = G(-\mathbf{I} + k\langle \mathbf{R}\mathbf{R} \rangle)$ , where expressions for the normalization  $k$  and the modulus  $G$  can be derived in terms of the parameters in the underlying microscopic polymeric models. Here  $\langle (\cdot) \rangle$  denotes a thermal average. The DJS model [20, 32] provides the constitutive equation for the evolution of the viscoelastic stress:

$$\overset{\diamond}{\Sigma} + \frac{1}{\tau} \mathbf{\Sigma} = 2 \frac{\mu}{\tau} \mathbf{D} + \mathcal{D} \nabla^2 \mathbf{\Sigma}, \quad (\text{II.10})$$

where

$$\overset{\diamond}{\Sigma} = (\partial_t + \mathbf{v} \cdot \nabla) \mathbf{\Sigma} + (\mathbf{\Omega} \cdot \mathbf{\Sigma} - \mathbf{\Sigma} \cdot \mathbf{\Omega}) - a(\mathbf{D} \cdot \mathbf{\Sigma} + \mathbf{\Sigma} \cdot \mathbf{D}), \quad (\text{II.11})$$

$\tau$  is a relaxation time, the ‘‘polymer’’ viscosity  $\mu$  is related to the modulus by  $G = \mu/\tau$ , and  $\mathbf{\Omega} = \frac{1}{2} [\nabla\mathbf{v} - (\nabla\mathbf{v})^T]$ . The total stress comprises the viscoelastic stress of the DJS model and a Newtonian contribution, according to Eq. (II.1), and the viscosity ratio  $\epsilon \equiv \eta/\mu$  controls the balance between the two stresses. The ‘slip parameter’  $a$ , which describes non-affine stretch of the dumbell with respect to the extension of the flow, allows for a non-monotonic constitutive curve for  $0 < |a| < 1$  and  $\epsilon < 1/8$ .

The ‘‘diffusion’’ term  $\mathcal{D} \nabla^2 \mathbf{\Sigma}$  describes non-local relaxation of the viscoelastic stress and is necessary to describe strongly inhomogeneous flow profiles [20]. Because of this term the steady shear banding state obeys a spatial differential equation, which must solved subject to boundary conditions specified at the walls of the flow cell. The

solvability condition for a stationary interface leads to a unique total shear stress plateau for imposed average shear rates in the non-monotonic portion of the constitutive curve [24]. The characteristic width  $\ell$  of the interface between shear bands is given by  $\ell = \sqrt{\mathcal{D}\tau}$ .

For convenience we define the modulus  $G = \mu/\tau$ . In cylindrical Couette flow the constitutive equation has the following components [20]:

$$\mathcal{L}\Sigma_{rr} = -(1-a)\dot{\gamma}\Sigma_{r\theta} + \frac{2\mathcal{D}e^{-2qx}}{q^2R_1^2}(\Sigma_{\theta\theta} - \Sigma_{rr}) \quad (\text{II.12a})$$

$$\mathcal{L}\Sigma_{\theta\theta} = (1+a)\dot{\gamma}\Sigma_{r\theta} - \frac{2\mathcal{D}e^{-2qx}}{q^2R_1^2}(\Sigma_{\theta\theta} - \Sigma_{rr}) \quad (\text{II.12b})$$

$$\mathcal{L}\Sigma_{r\theta} = \dot{\gamma} \left[ G - \frac{1-a}{2}\Sigma_{\theta\theta} + \frac{1+a}{2}\Sigma_{rr} \right] - \frac{4\mathcal{D}e^{-2qx}}{q^2R_1^2}\Sigma_{r\theta}, \quad (\text{II.12c})$$

where

$$\mathcal{L} \equiv \frac{\partial}{\partial t} + \frac{1}{\tau} - \frac{\mathcal{D}e^{-2qx}}{q^2R_1^2} \frac{\partial^2}{\partial x^2}. \quad (\text{II.13})$$

## 2. Reptation-Reaction and Rolie-Poly Models

In Cates' model [33] of wormlike micellar solutions the micelles are assumed to relax in a tube, and the deviatoric viscoelastic stress is defined by  $\Sigma = G(-\mathbf{I} + 3\langle \mathbf{u}\mathbf{u} \rangle)$ , where the unit vector  $\mathbf{u}$  describes the local orientation of tube segments. The viscoelastic stress is given by a history integral over the second moment of the correlation function  $\langle \mathbf{u}\mathbf{u} \rangle$ , weighted by the distribution of tube segments as they are created and subsequently vacated by the wormlike micelles. The resulting integral equation can be re-written as the following differential equation by using a decoupling approximation to remove fourth order moments [29]:

$$\overset{\nabla}{\Sigma} + \frac{1}{\tau}\Sigma = 2G\mathbf{D} - \frac{2}{3}\Sigma : \nabla\mathbf{v}(\mathbf{I} + (1+\beta)[\Sigma/G]) + \mathcal{D}\nabla^2\Sigma, \quad (\text{II.14})$$

where

$$\overset{\nabla}{\Sigma} = (\partial_t + \mathbf{v} \cdot \nabla)\Sigma + (\boldsymbol{\Omega} \cdot \Sigma - \Sigma \cdot \boldsymbol{\Omega}) - (\mathbf{D} \cdot \Sigma + \Sigma \cdot \mathbf{D}). \quad (\text{II.15})$$

and  $\beta = 0$ . A stress diffusion term, which did not appear in the original formulation, has been included here. The non-linear term in this equation preserves the traceless property of the deviatoric stress.

Interestingly, for  $\beta \neq 0$  this model is identical to an extension of the original Doi-Edwards (DE) theory of entangled polymers [2], which has a constitutive instability and therefore predicts shear banding. This extension to

the DE theory incorporates the enhanced release of polymer entanglements due to convection[34], which increases the polymer stress and can “cure” the DE instability for sufficiently strong convected constraint release (CCR). CCR and tube stretching were incorporated in [3] to describe polymer melts and wormlike micelles, and later simplified to a differential version called the Rolie-Poly model [28]. In the version of the Rolie-Poly model used in Eq. (II.14) the tube length is assumed to be relaxed (“non-stretching”). The CCR parameter  $\beta$  ( $0 \leq \beta \leq 1$ ) corresponds to the frequency of the release of polymer entanglement constraints due to convection by the flow, and Ref. [28] used  $\beta = 1$  to model a well-entangled polymer melt without a constitutive instability. For small enough  $\beta$  a constitutive instability results.

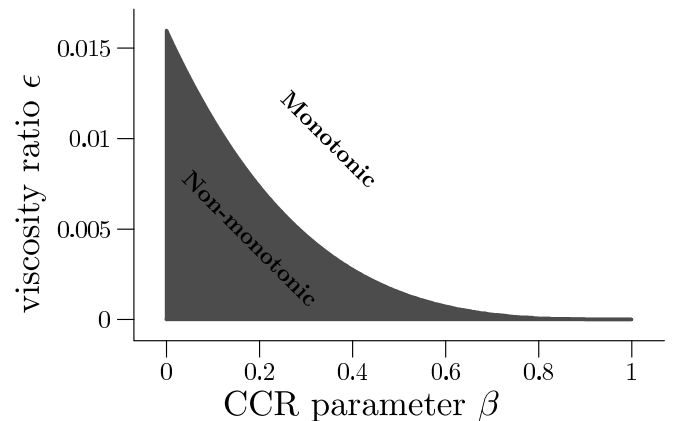


FIG. 2: Regions of parameter space  $\beta$  and  $\epsilon$  for which the DRP model has either a monotonic (unshaded) or non-monotonic (shaded) constitutive curve.

The DE constitutive instability is thought to be an unphysical prediction of polymer melts [3], but a series of recent experiments have reignited the interest in this instability by showing data, such as velocity profiles, consistent with shear banding in polymer *solutions* [35, 36, 37]. Hence, the Rolie-Poly model is an alternative simple constitutive model to the DJS model for wormlike micelles, and by tuning the parameter  $\beta$  it can be used to address polymer solutions and melts.

As with the DJS model, we define the modulus by  $G = \mu/\tau$  and the ratio of Newtonian (or “solvent”) to viscoelastic stress by  $\epsilon = \eta/\mu$ . For a given  $\epsilon$  a non-monotonic constitutive curve is found if the degree of convected constraint release  $\beta$  is small enough (Fig. 2). We will refer to the model described above as the diffusive Rolie-Poly (DRP) model. In cylindrical Couette flow the components of the DRP model evolve according to



$$\mathcal{L} \Sigma_{rr} = -\frac{2}{3} \dot{\gamma} \Sigma_{r\theta} [1 + (1 + \beta)(\Sigma_{rr}/G)] + \frac{2\mathcal{D}e^{-2qx}}{q^2 R_1^2} (\Sigma_{\theta\theta} - \Sigma_{rr}) \quad (\text{II.16a})$$

$$\mathcal{L} \Sigma_{\theta\theta} = -\frac{2}{3} \dot{\gamma} \Sigma_{r\theta} [1 + (1 + \beta)(\Sigma_{\theta\theta}/G)] + 2\dot{\gamma} \Sigma_{r\theta} - \frac{2\mathcal{D}e^{-2qx}}{q^2 R_1^2} (\Sigma_{\theta\theta} - \Sigma_{rr}) \quad (\text{II.16b})$$

$$\mathcal{L} \Sigma_{r\theta} = G\dot{\gamma} + \dot{\gamma} \Sigma_{rr} - \frac{2}{3} \dot{\gamma} \Sigma_{r\theta} (1 + \beta) [\Sigma_{r\theta}/G] + \frac{4\mathcal{D}e^{-2qx}}{q^2 R_1^2} \Sigma_{r\theta}. \quad (\text{II.16c})$$

### III. BOUNDARY CONDITIONS

#### A. Liquid crystal analogy

To motivate the possible boundary conditions in micellar systems, we first recall the situation of nematic liquid crystals confined between walls, in which the boundary conditions are a balance between surface and bulk interactions [38]. The free energy depends on the nematic order parameter  $\mathbf{Q} = \langle \mathbf{u}\mathbf{u} \rangle - \frac{1}{3}\mathbf{I}$ , where the unit vector  $\mathbf{u}$  is parallel to the liquid crystalline mesogen [38]. A simple free energy functional for a nematic liquid crystal is [38]

$$F_{\text{LC}} = \int_V [f_h^b(\mathbf{Q}) + \frac{1}{2}\mathcal{K}(\nabla\mathbf{Q})^2] dV + \frac{1}{2}W_Q \int_{\text{walls}} \text{Tr}(\mathbf{Q} - \mathbf{Q}_0)^2 dS, \quad (\text{III.1})$$

where  $f_h^b(\mathbf{Q})$  is the homogeneous part of the bulk free energy, typically expanded in Landau form as  $f_h^b(\mathbf{Q}) = \frac{a}{2}\text{Tr}\mathbf{Q}^2 + \dots$ ; and  $\mathcal{K}$  is a Frank elastic constant. In the surface free energy,  $W_Q$  specifies the strength of the wall potential which penalizes deviations away from a specific order parameter tensor  $\mathbf{Q}_0$ . More complex surface free energies are possible [39], and one may alternatively consider boundary conditions that influence the orientation, but not magnitude, of  $\mathbf{Q}$  at the wall.

The spatial dependence of the order parameter is found by demanding that the free energy functional be stationary with respect to varying  $\mathbf{Q}$ . Stationarity in the bulk leads to the following differential equation,

$$\frac{\delta F_{\text{LC}}}{\delta \mathbf{Q}} = [a\mathbf{Q} + \dots] - \mathcal{K}\nabla^2 \mathbf{Q} = 0, \quad (\text{III.2})$$

whose boundary condition arises from requiring zero variation at the wall:

$$\mathcal{K} \hat{\mathbf{n}} \cdot \nabla \mathbf{Q} + W_Q (\mathbf{Q} - \mathbf{Q}_0) = 0. \quad (\text{III.3})$$

Here  $\hat{\mathbf{n}}$  is the outward unit normal from the fluid at the wall. Physically the boundary condition is a balance of surface and bulk mesogen torques at the wall.

The bulk equation defines a correlation length  $\ell = \sqrt{\mathcal{K}/a}$ , which controls the decay of order parameter fluctuations away from the surface, while the boundary defines an *extrapolation* length

$$\xi = \frac{\mathcal{K}}{W_Q}. \quad (\text{III.4})$$

which controls wall anchoring [38]. The extrapolation length  $\xi$  roughly sets the scale for the gradient  $\nabla \simeq 1/\xi$

induced at the wall by the boundary conditions, in the absence of bulk Frank stresses. For small  $\xi$  the effective gradient at the wall is very large, so the wall potential has a strong effect. Conversely, for large  $\xi$  the characteristic gradient is small, and the anchoring potential has a weak effect.

The extrapolation length should thus be compared with the bulk healing length or correlation length  $\ell$  to assess the relevant regime. For strong anchoring ( $\xi \ll \ell$ ) the boundary conditions effectively impose  $\mathbf{Q}_0$  at the walls, while for weak anchoring ( $\xi \gg \ell$ ) the boundary conditions are dictated by the Frank elastic term, resulting in Neumann boundary conditions, or  $\nabla \mathbf{Q} = 0$ . Mixed boundary conditions apply between these two extremes [38].

#### B. Viscoelastic stress

Based on the liquid crystalline example, we apply similar boundary conditions to the viscoelastic stress; the details are given in Appendix A. The viscoelastic stress is analogous to the liquid crystalline order parameter, and the stress diffusion constant is analogous to the Frank elastic constant (according to  $\mathcal{D}\tau \rightarrow \mathcal{K}$ ), and there is a corresponding anchoring potential, leading to

$$\mathcal{D}\tau \hat{\mathbf{n}} \cdot \nabla \Sigma + W(\Sigma - \Sigma_0) = 0. \quad (\text{III.5})$$

In this formulation the anchoring potential has the dimensions of length.

As shown in Appendix A, the anchoring potential  $W$  can be expressed as  $W = W_0/G$ , where  $G$  is the modulus and  $W_0$ , with dimensions of energy per area, penalizes

deviations of the polymer deformation  $\mathbf{\Lambda}$  from a reference strain  $\mathbf{\Lambda}_0$ . A simple, albeit weak, contribution to the anchoring potential comes from the effect of the wall on nearby micellar or polymer conformations. In Appendix A we estimate  $W \simeq 2R_g$ , where  $R_g$  is the radius of gyration.

By analogy with the liquid crystal example above, the extrapolation length is given by

$$\xi = \frac{\mathcal{D}\tau}{W} = \frac{\sqrt{\mathcal{D}\tau}}{W} \ell, \quad (\text{III.6})$$

where the interfacial width  $\ell$  is set by the diffusion constant according to  $\ell = \sqrt{\mathcal{D}\tau}$ . The two characteristic lengths are comparable when  $\xi_c/\ell \simeq 1$ , or as noted above, we expect weak anchoring (large  $\xi$ ) to yield effectively Neumann boundary conditions (zero gradient) [19, 20, 26]. Alternatively, very strong anchoring might be encouraged by specific wall treatments such as rubbing or grooving the walls along a particular direction  $\hat{\mathbf{m}}$ , *i.e.* specifying  $\Sigma_0 \sim \hat{\mathbf{m}}\hat{\mathbf{m}}$ . This would give very small  $\xi$  and effectively Dirichlet boundary conditions, as used by Cook and co-workers [25, 40].

To determine the extrapolation length an estimate for the diffusion constant  $\mathcal{D}$  is necessary. Two physical effects that can lead to non-local dynamics and hence stress diffusion are (1) the semi-flexibility of wormlike micelles [22] and (2) diffusion of micelles that carry stress [40]. In the former case the characteristic length scale is set by the persistence length  $\ell_p$ . In Appendix B we estimate these two contributions to be

$$\mathcal{D}\tau = \begin{cases} \frac{\ell_p^2}{126} & (\text{semi-flexibility}) \\ D_{\text{tr}}\tau & (\text{translational diffusion}), \end{cases} \quad (\text{III.7})$$

where  $D_{\text{tr}}$  is the translational diffusion coefficient.

If we consider effects due to semiflexibility, combined with the estimate for  $W$  due steric wall interactions, we estimate anchoring length to be

$$\xi \simeq \frac{\ell_p^2}{126R_g}. \quad (\text{III.8})$$

For typical giant micelles for which  $L \simeq 200 \mu\text{m}$ ,  $\ell_p \simeq 20 \text{ nm}$ , we find  $R_g \simeq 18\ell_p \simeq 670 \text{ nm}$ , or  $\xi \simeq 0.05 \text{ \AA}$ . The interfacial width in this case is  $\ell \simeq 0.09\ell_p \simeq 180 \text{ \AA}$ , which would imply strong anchoring.

Alternatively, if we naively apply the estimate for the contribution of translational diffusion to semidilute micellar solutions with  $D_{\text{tr}} \simeq 10^{-8} \text{ cm}^2/\text{s}$ ,  $\tau \simeq 1 \text{ s}$ , one finds  $\mathcal{D}\tau \simeq 1 \mu\text{m}^2$ , leading to  $\xi/\ell \simeq 0.8$ . Hence in this case one might expect the effective boundary conditions to be somewhere between Neumann and Dirichlet.

We stress that these estimates are necessarily very crude, and should ultimately be replaced by more precise calculations.

## IV. METHOD OF CALCULATION

### A. Non-dimensional parameterization

For both models we express all stresses in units of the modulus  $G$ , and express time in units of the micellar (polymer) stress relaxation time  $\tau$ . In cylindrical Couette flow the natural scale for length is the quantity  $R_1q$ , which for small  $q$  is identical to the gap size  $R_2 - R_1 = R_1(1 - e^{-q})$ . Hence the dimensionless quantities are:

$$\hat{t} = t/\tau \quad \hat{\gamma} = \dot{\gamma}\tau \quad (\text{IV.1a})$$

$$\hat{\Sigma} = \frac{\Sigma}{G} \quad \hat{D} = \frac{\mathcal{D}\tau}{R_1^2q^2} \quad (\text{IV.1b})$$

$$\hat{W} = \frac{W}{R_1q}. \quad (\text{IV.1c})$$

In cylindrical Couette flow the boundary conditions are

$$-\hat{D} \frac{\partial}{\partial x} \Sigma + \hat{W}(\Sigma - \Sigma_0) = 0 \text{ at } x = 0 \quad (\text{IV.2})$$

$$\hat{D}e^{-q} \frac{\partial}{\partial x} \Sigma + \hat{W}(\Sigma - \Sigma_0) = 0 \text{ at } x = 1, \quad (\text{IV.3})$$

where the change in sign arises from the orientation of the surface normal  $\hat{\mathbf{n}}$ .

We use  $\epsilon = 0.05$ ,  $a = 0.3$  for the DJS model and  $\epsilon = 0.01$ ,  $\beta = 0$  for the DRP model, which places the latter in a non-monotonic regime of parameter space (Fig. 2). For the shear rate startup calculations shear rates of  $\langle \hat{\gamma} \rangle = 3.8$  (DJS model) and  $\langle \hat{\gamma} \rangle = 9$  (DRP model) were used, which are approximately in the middle of the respective stress plateaux.

### B. Numerical Methods

We solve the creeping flow equation (Eq. II.8) for the steady state banding profiles of the two models (Eqs. II.12 and II.16) for different imposed boundary conditions and parameter values. In all cases an average shear rate  $\langle \dot{\gamma} \rangle$  is imposed, so the local shear rate is eliminated using Eq. (II.8) to obtain a set of coupled second order partial differential equations for  $\Sigma_{rr}$ ,  $\Sigma_{\theta\theta}$  and  $\Sigma_{r\theta}$ . This set of equations is then solved subject to the chosen boundary conditions by evolving them using the Crank-Nicolson algorithm [41].

Two protocols were followed; the first for shear rate sweeps to calculate flow curves, and the second to calculate shear rate startups from rest. In the first protocol, shear rate sweeps, an initial viscoelastic stress profile was generated by a set of the 20 longest wavelength Fourier modes that fit the desired boundary conditions. Randomly assigned amplitudes for modes of the stress components  $\Sigma_{\alpha\beta}$  were taken from a uniform distribution such that the maximum total value for any component 1. The equations of motion were then evolved to find the



steady state for the first average imposed shear rate in the sweep, which was on either the low or high shear rate flow branch of the constitutive curve. This state was then used as the initial condition for the next average shear rate in the sweep (either incremented or decremented). This process was continued until the desired region of the flow curve had been calculated. At each shear rate the system was evolved typically 500 relaxation times  $\tau$ , using a time step of 0.05; it was checked that the steady state results did not change for smaller time steps.

In the second protocol a range of initial viscoelastic stress configurations were subjected to a given step in the shear rate from zero, and evolved to ensure that the same steady state was reached. Using random initial conditions often produced multiple bands in this protocol; these multiple bands often did not anneal into two bands, which is probably a consequence of both the slow motion of interfaces in one dimensional systems and the fact that multiple interface solutions are known to be locally stable when the interface width is much smaller than the system size [42]. Because we are not interested, here, in the details of this degeneracy, we used a spatially smooth initial condition for the viscoelastic stress tensor which would encourage the simplest band configuration commensurate with the given boundary conditions. A variety of different smooth initial conditions were used to ensure that the results obtained were robust.

For smaller  $\hat{D}$  and  $q$  steady state could take as long as  $10000\tau$  to attain. In the shear banding regime the positions of the interfaces between bands were monitored to ensure that they had stopped moving. In nearly flat geometries with weak curvatures and/or sharp interfaces ( $q < 10^{-4.5}$ ,  $\hat{D} < 10^{-3}$ ), it was difficult to reliably determine the steady state interfacial position; this slow dynamics of fronts in one dimension is well known [43]. Hence we did not study this range of parameter space for this protocol.

### C. Notation for (Dirichlet) Boundary Conditions

A convenient parameterization of the Dirichlet boundary condition  $\Sigma_0$  arises from the physical interpretation of viscoelastic stress tensor in terms of the second moment of unit vector orientations,  $\Sigma = G(3\langle \mathbf{u}\mathbf{u} \rangle - \mathbf{I})$ . In its principal frame  $\Sigma_0$  can be written (in dimensionless form) as

$$\frac{1}{3}\hat{\Sigma}_0 + \frac{1}{3}\mathbf{I} = \begin{pmatrix} \frac{2S+1}{3} & 0 & 0 \\ 0 & \frac{1-S}{3} - b & 0 \\ 0 & 0 & \frac{1-S}{3} + b \end{pmatrix}, \quad (\text{IV.4})$$

where  $S = \frac{3}{2}\langle \cos^2 \theta \rangle - \frac{1}{2}$  and  $b = \langle \sin^2 \theta \cos 2\phi \rangle$ . The conventional spherical polar coordinates  $\theta$  and  $\phi$  describe the orientation of unit vectors with respect to the principal axes. The parameter  $S$  specifies the degree of order along the principal direction, and  $b$  specifies the degree of biaxiality;  $S$  and  $b$  obey  $-\frac{1}{2} < S < 1$ , so  $-\frac{1-S}{3} < b < \frac{1-S}{3}$ .

The order parameter may be illustrated by plotting an ellipsoid with principal axes parallel to and proportional to those of the tensor (Fig. 3).

By symmetry, we expect the principal axes of  $\Sigma_0$  to coincide with the axes chosen, although in principle one could include non-trivial tilt angles with respect to the surface, as is found in the behavior of liquid crystals at some interfaces. One could also consider, as was done in Refs. [25, 26], a preferred direction of alignment along the wall. Below we will consider the natural principal axes set by the wall, the principal axes determined by the steady state bulk solutions, and a restricted set of angles rotated with respect to the natural axes.

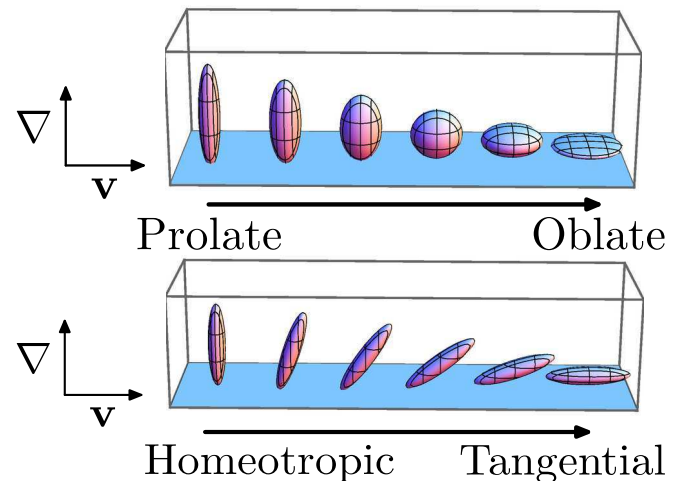


FIG. 3: Tuning between (top) prolate and oblate boundary alignment, and (bottom) homeotropic and tangential alignment. In (top) the principal axes remain unchanged and the parameters  $S$  and  $b$  are varied, while in (bottom)  $S$  and  $b$  are fixed to a prolate distribution, and the angle of orientation with respect to the surface is changed.

## V. REVIEW: NEUMANN BOUNDARY CONDITIONS

We first review the results for Neumann boundary conditions, calculated previously for the DJS model [20]. In this case the diffusion term selects the value  $T_{xy}^*$  of the total shear stress plateau [24]. At other values for the total stress the interface moves at a speed  $c \sim T_{xy} - T_{xy}^*$  [44]. In a flow geometry such as cylindrical Couette flow, in which the total stress is inhomogeneous in steady state, the interface lies at the position of the flow cell at which the total stress is equal to  $T_{xy}^*$  (as long as the stress gradient is negligible over the scale of the interfacial width  $\ell = \sqrt{\hat{D}}$ ) [44, 45]. Hence the stress gradient drives the interface to the correct position. In a flat geometry with no stress gradient the speed of approach to the steady state position commensurate with the imposed average shear rate is determined by interaction with the distant wall,

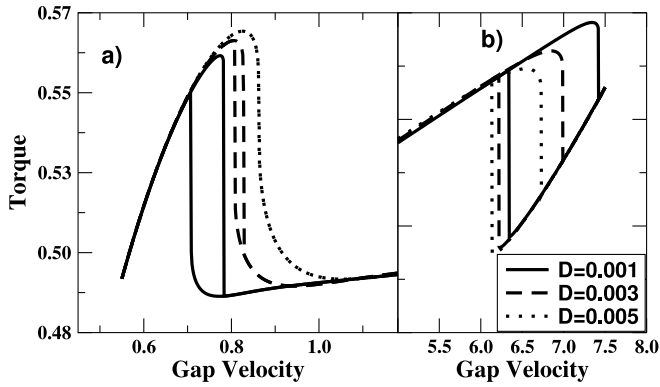


FIG. 4: Hysteresis observed in the DJS model in Couette flow for  $q = 0.00995$  upon increasing and decreasing the average shear rate (or the gap velocity) along the either edge of the stress plateau (From [20]).

and vanishes in the limit of a small diffusion coefficient [46].

The flow curve in cylindrical Couette flow has a stress plateau with a slope  $\partial T^*/\partial \langle \dot{\gamma} \rangle \sim e^q - 1$ , which vanishes in the planar limit  $q = 0$ . In either case the flow curves display hysteresis at either edge of the stress plateau (Fig. 4) [20]. During a shear rate sweep from rest the fluid follows the low shear rate branch to a total wall stress  $T_{r\theta} > T_{xy}^*$ , until a high shear rate band forms and the stress decreases onto the stress plateau. For a downward shear rate sweep from on the stress plateau the high shear rate band shrinks until the interface “touches” the wall; at this point a banding solution is unstable with respect to a homogeneous phase with a higher stress on the low shear rate branch. The width of the hysteresis loop decreases upon decreasing the diffusion constant  $\hat{D}$ . Hysteresis of this sort is frequently seen in wormlike micellar solutions at the low shear rate edge of the stress plateau [10, 11, 17]. The nature of nucleation of a shear banded state from a homogeneous phase is unknown and worthy of study in its own right. Similar behavior is predicted near the high shear rate edge of the stress plateau; this has rarely been studied wormlike micelles, because in many cases the high shear rate band is unstable or unattainable.

Fig. 5 shows the position of the interface between bands as a function of curvature  $q$ , defined as the position of midpoint between the high and low values of  $\Sigma_{r\theta}$ . The high shear rate band appears to shrink at high curvatures (large  $q$ ) because of the total stress gradient; this larger gradient effectively concentrates the shear rate near the inner wall, so that the shear band occupies a slightly narrower region. The viscoelastic stress tensor of the high shear rate band of the DRP model, while strongly ordered, is only slightly more aligned with the velocity direction than in the low shear rate state. The high shear rate state has an alignment angle of  $26^\circ$  which is comparable to that reported in experiments on worm-

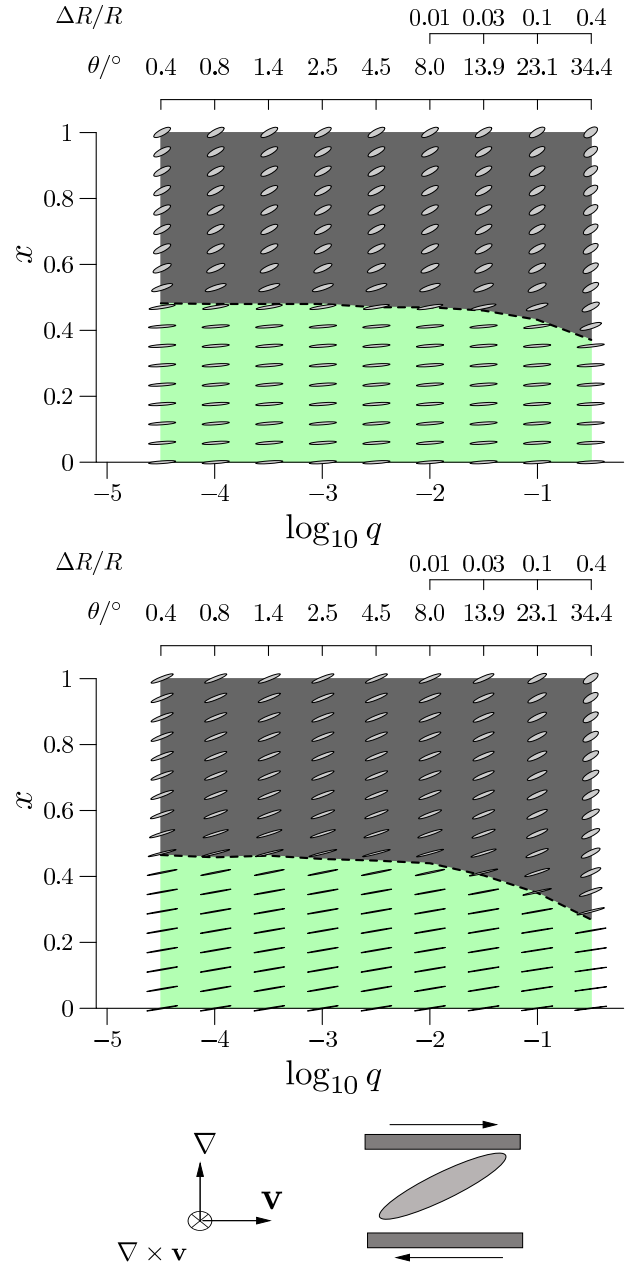


FIG. 5: Position  $x$  of the interface between the shear bands for Neumann boundary conditions for the DJS (top) and DRP (bottom) models, for  $\hat{D} = 1.6 \times 10^{-3}$ . The ellipses show the eigenvalues and principal axes of the viscoelastic stress tensor, which in all cases is nearly prolate in the velocity gradient plane. The coordinate  $x$  is parallel to the velocity gradient,  $\nabla$ .

like micelles [16]. This contrasts with the DJS model, in which the viscoelastic stress tensor is strongly aligned with the flow in the high shear rate band. Hence the DRP model may be a more realistic model for wormlike micelles than the DJS model.

The upper abscissas of Fig. 5 shows the stress differences in terms of geometric parameters for cylindri-

cal Couette flow,  $\Delta R/R$  and an equivalent cone angle  $\theta$  for cone and plate flow. In Couette flow,  $\Delta R/R \equiv (R_2 - R_1)/R_1 = 1 - e^{-q}$ , and the relative stress difference between the two cylinders is  $\Delta T_{r\theta}/T_{r\theta} = 1 - e^{-2q}$ . In cone and plate geometry the relative stress difference is a weak function of the cone angle,  $\Delta T_{\theta\phi}/T_{\theta\phi} = \tan^2 \theta$  [47]. Hence, the cone angle  $\theta$  that is roughly equivalent to a given Couette curvature satisfies  $\tan^2 \theta = 1 - e^{-2q}$ .

For reference, the selected stresses for the two models in a flat geometry, in the limit of an infinite system and Neumann boundary conditions, are:

$$\frac{\hat{T}_{r\theta}^*}{1 + \beta} = 0.62431 \quad \text{DRP model} \quad (\text{V.1})$$

$$\hat{T}_{r\theta}^* \sqrt{1 - a^2} = 0.4829 \quad \text{DJS model.} \quad (\text{V.2})$$

## VI. RESULTS

We first consider weakly curved Couette flow, and study the effect of different Dirichlet boundary conditions and the magnitude of the diffusion coefficient, or equivalently the widths of interfaces, on the flow curves and attendant hysteresis. Then we study how the stress profiles and configuration of bands depends on the competition between the stress gradient due to the curvature of Couette flow and the boundary conditions. Finally, we consider mixed boundary conditions, in which the value at the wall can adjust depending on the competition between boundary parameters and the stress gradient.

### A. Effect of different Dirichlet boundary conditions for fixed geometry

#### 1. Prolate and flow-aligning anchoring

Here we choose the degree of curvature to be  $q = 0.005$ , which corresponds to radii  $R_2 \simeq 1.005R_1$ , and enforce  $\Sigma_0$ , the value of the viscoelastic stress at the boundaries (Dirichlet boundary conditions). The principal axes of  $\Sigma_0$  are chosen to be parallel to the flow, gradient and vorticity directions. The parametrisation of Eq. (IV.4) was then applied so that  $\Sigma_0$  is completely aligned in the flow direction, tangential to the walls, for  $S \rightarrow 1$ .

Figure 6 shows the flow curves and shear rate profiles for upward ( $\triangleright$ ) and downward ( $\triangleleft$ ) sweeps of the DJS model for flow-aligning prolate boundary conditions,  $S = 1.0, b = 0$ , similar to the value  $\Sigma_H$  of the high shear rate branch. For low shear rates the stress is lower than that of the low shear rate constitutive curve; profiles 1 and 4. This can be traced to a lubricating layer (evident in the inset of profile 1) induced by the wall, which has a higher shear rate and hence reduces the stress for a given imposed shear rate. Hysteresis is not seen at the low shear rate edge of the plateau, presumably because

of this lubricating layer. Hence the effect of the boundary condition is similar to heterogeneous nucleation or a wetting: it provides a site upon which the shear band can easily grow, and can eliminate the hysteresis so that the shear band grows smoothly from the wall without requiring “nucleation”. Hysteresis is only seen at the high shear rate side of the stress plateau, as shown in profiles 3 and 6, which are at the same imposed shear rate but after different histories. This behavior should be contrasted with Neumann boundary conditions, in which hysteresis was predicted at both edges of the stress plateau.

In the stress plateau region the high shear rate band lies near the inner cylinder (near  $x = 0$ ), for both increasing and decreasing shear rate sweeps (profiles 2 and 5). Hence the higher total stress near the wall induces preferential growth of the inner cylinder’s lubricating layer upon increasing the average shear rate, while the low shear rate band preferentially forms at the outer wall upon decreasing the average shear rate.

#### 2. Homeotropic anchoring

The converse behavior is observed when the boundary conditions are less favorable to the high shear rate phase. Fig. 7 shows the flow profiles and flow curves for  $S = -0.5, b = -0.5$ , which corresponds to homeotropic orientation with principal axis in the gradient direction. In this case a hysteresis loop is found at the low shear rate end of the plateau (profiles 1 and 4), and not at the high shear rate end (profiles 3 and 6). This boundary condition induces a more viscous layer of low shear rate material near the wall, even well into the stress plateau (profiles 2 and 5). Profiles 2 and 5 are not quite superposable, indicating that steady state was probably not exactly attained for these profiles; indeed, this value  $\hat{D} = 4.4 \times 10^{-4}$  is at the lower limit of our computational capabilities. The more viscous layer near the boundary increases the total stress on the high shear rate branch above than that of the constitutive curve (profiles 3 and 6), as opposed to the lubricating layer found for the flow-aligning boundary condition.

The shear rate adjacent to the wall is extremely high (inset to profile 1), even higher than that of the high shear rate phase. This is because the principal axes of the boundary condition  $\Sigma_0$  are aligned with the velocity, gradient, and vorticity, such that the shear component  $\Sigma_{0r\theta}$  vanishes. To compensate for this vanishing contribution to the shear stress, a very high shear rate is required to obtain the necessary value for the total shear stress. For boundary conditions conducive to the low shear rate phase, the viscoelastic stress tensor  $\Sigma$  adjacent to the wall nonetheless “heals” to the value characteristic of the low shear rate branch, whereas in the flow-aligning case it heals to the value characteristic of the high shear rate branch. Although our numerics can adequately resolve the sharp gradient near the wall, it is likely that higher order gradients are necessary for a

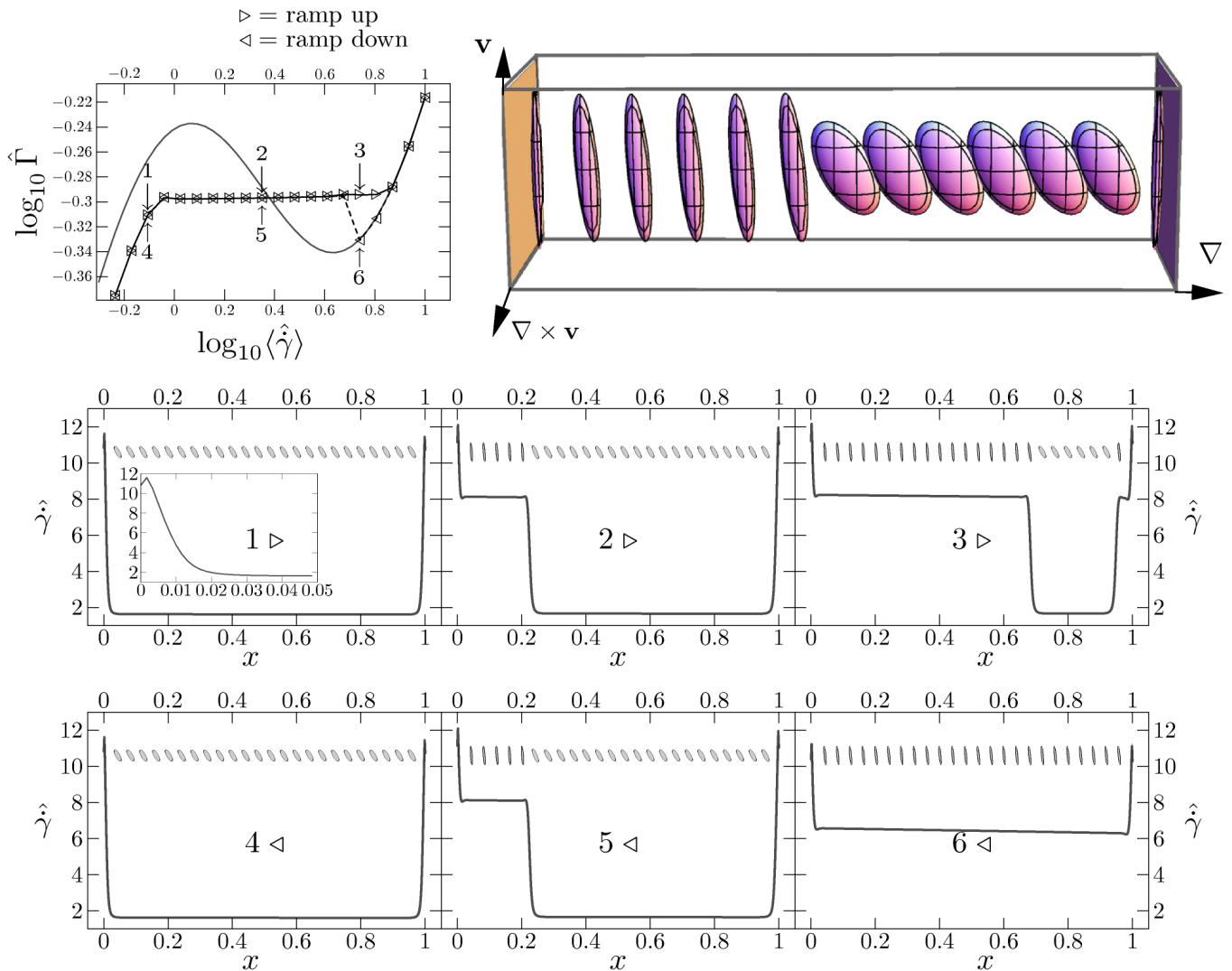


FIG. 6: Top left: constitutive curve (solid line) and flow curves (triangles) for upward and downward sweeps of the DJS model with  $\hat{D} = 4.4 \times 10^{-5}$ , in cylindrical Couette flow with  $q = 0.005$ . The boundary condition is  $\Sigma_0 = \text{diag}(2, -1, -1)$ , as illustrated by the ellipsoids (top right). For the noted average imposed shear rates, figures (1)-(6) show the steady state shear rate profiles  $\hat{\gamma}(x)$ , and cross-sections of the ellipsoids corresponding to  $\Sigma(x) + \mathbf{I}$  in the velocity-flow gradient plane. In all cases the ellipsoids are nearly prolate. The inset in (1) shows the detail of the shear rate profile near the inner wall  $x = 0$ .

constitutive model to give physically accurate results.

### 3. Variation with anchoring angle and from prolate to oblate anchoring

A summary of the behavior of the DJS and DRP models for a range of boundary conditions is shown in Fig. 8. There are essentially three types of behaviors: hysteresis loops can be observed on (1) the high shear rate end of the stress plateau, (2) the low shear rate end of the stress plateau, or (3) both ends of the stress plateau. Hysteresis vanishes on those flow branches whose viscoelastic stress, loosely, is similar to the imposed boundary value

$\Sigma_0$ . The two models have broadly similar qualitative behavior. [The apparently greater slope across the plateau for the DRP model (Fig. 8.2) is due to the curvature of Couette flow and the smaller vertical scale.]

The behavior was similar when the boundary conditions were altered by choosing a prolate  $\Sigma_0$  and rotating its principal axis from the flow gradient (homeotropic) to the velocity (tangential) directions. For tangential boundary conditions (resembling the high shear rate state) there was no overshoot at the start of the plateau either on ramping up the shear rate or down. However on ramping down the shear rate the flow curve undershoots the stress plateau at the high shear rate end of the plateau. As the boundary condition is rotated the

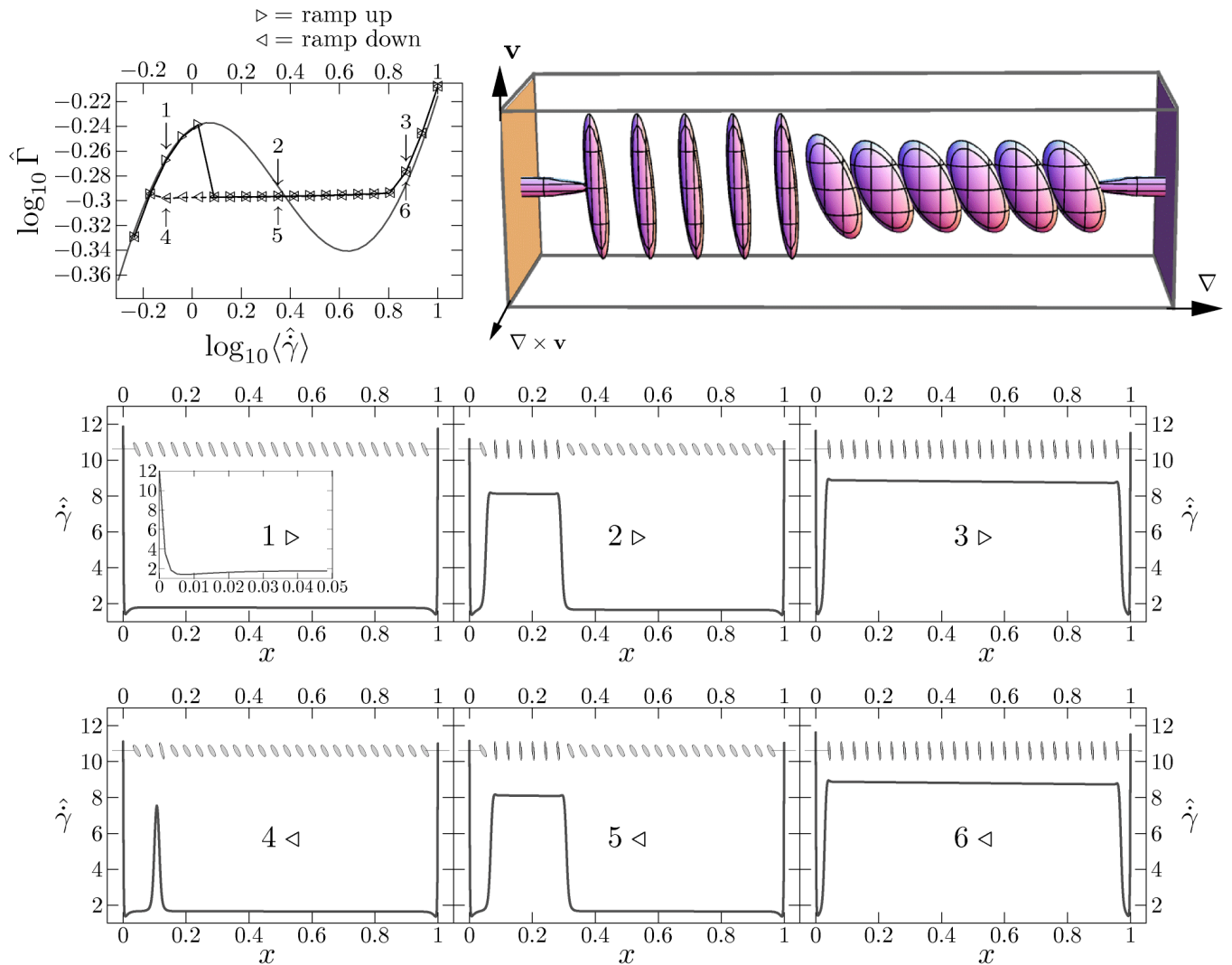


FIG. 7: Top left: constitutive curve (solid line) and flow curves (triangles) for upward and downward sweeps of the DJS model with  $\hat{D} = 4.4 \times 10^{-5}$ , in cylindrical Couette flow with  $q = 0.005$ . The boundary condition is  $\Sigma_0 = \text{diag}(-1, 2, -1)$ , as illustrated by the ellipsoid (top right). (1)-(6) show the shear rate profiles  $\hat{\gamma}$ , and cross-sections of the ellipsoids corresponding to  $\Sigma(x) + \mathbf{I}$  in the velocity-flow gradient plane. Except for very near the boundaries, the ellipsoids are nearly prolate. The inset in (1) shows the detail of the shear rate profile near the inner wall  $x = 0$ .

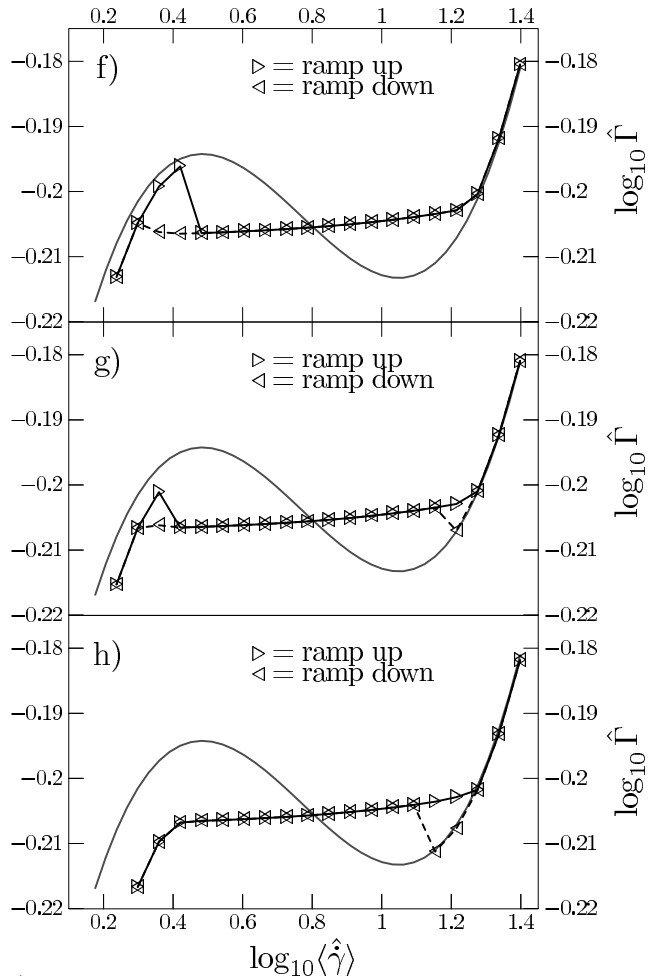
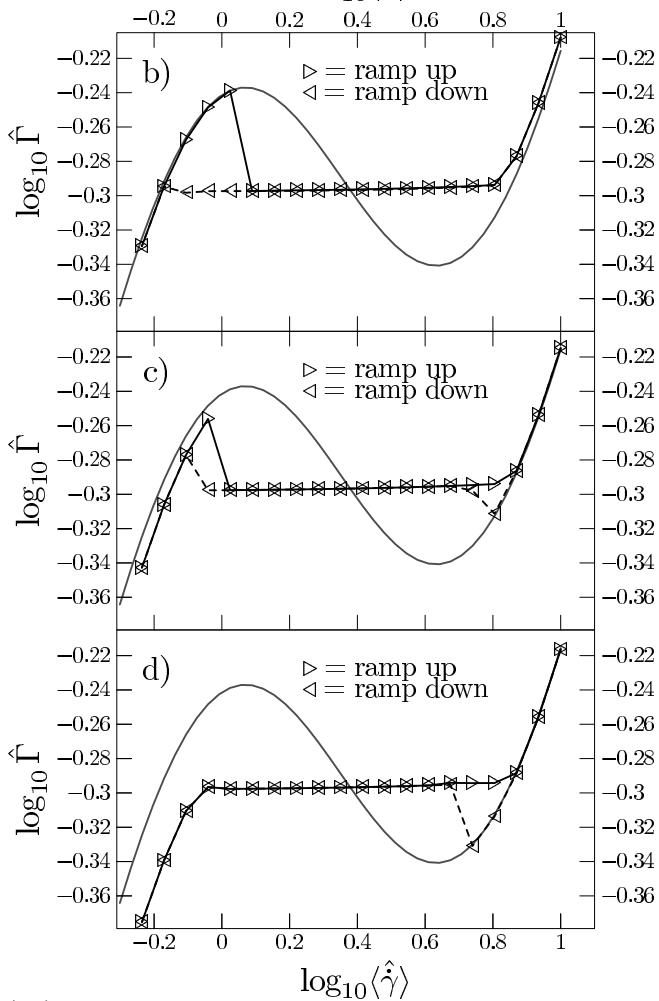
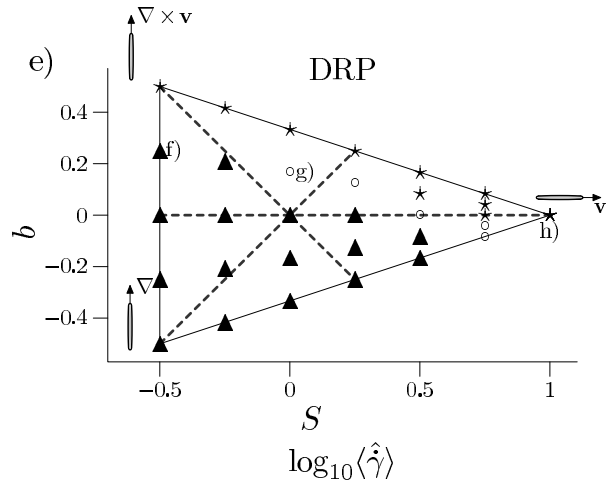
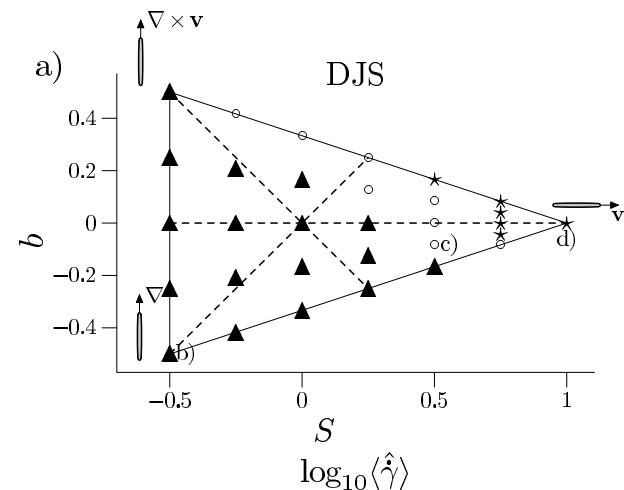
hysteresis at the high shear rate end vanishes, and starts to develop at the low shear rate end. For homeotropic alignment (resembling the low shear rate state) the hysteresis completely vanishes at the high shear rate end of the plateau, and hysteresis develops at the low shear rate end.

#### 4. Effect of the diffusion constant

Having determined that similarity of the imposed viscoelastic stress  $\Sigma_0$  to that of either the high or low shear

rate phases is crucial, we focus for the remainder of this paper on boundary conditions for which  $\Sigma_0$  is one of these values. For the DJS model with  $\epsilon = 0.05$  and  $a = 0.3$  these values are





(8.1) Parameter space and characteristic flow curves: DJS Model.

(8.2) Parameter space and characteristic flow curves: DRP Model.

FIG. 8: (a,e) Regions of  $S - b$  parameter space for  $\Sigma_0$  that give rise to different signatures of hysteresis during shear rate ramps, for  $q = 0.005$ , and  $\hat{D} = 4.4 \times 10^{-5}$ . The dashed lines on (a,e) correspond to uniaxial order interpolating between an oblate “pancake” and a prolate “needle” (at  $S = -0.5, b = \pm 0.5$ , and  $S = 1$ ), aligned along the vorticity, flow gradient, and flow directions. Hysteresis occurs at the low shear rate end of the stress plateau ( $\blacktriangle$ , b,f); at the high shear rate end of the stress plateau ( $\star$ , d,h); or at both ends shear rate end of the stress plateau ( $\circ$ , c,g).



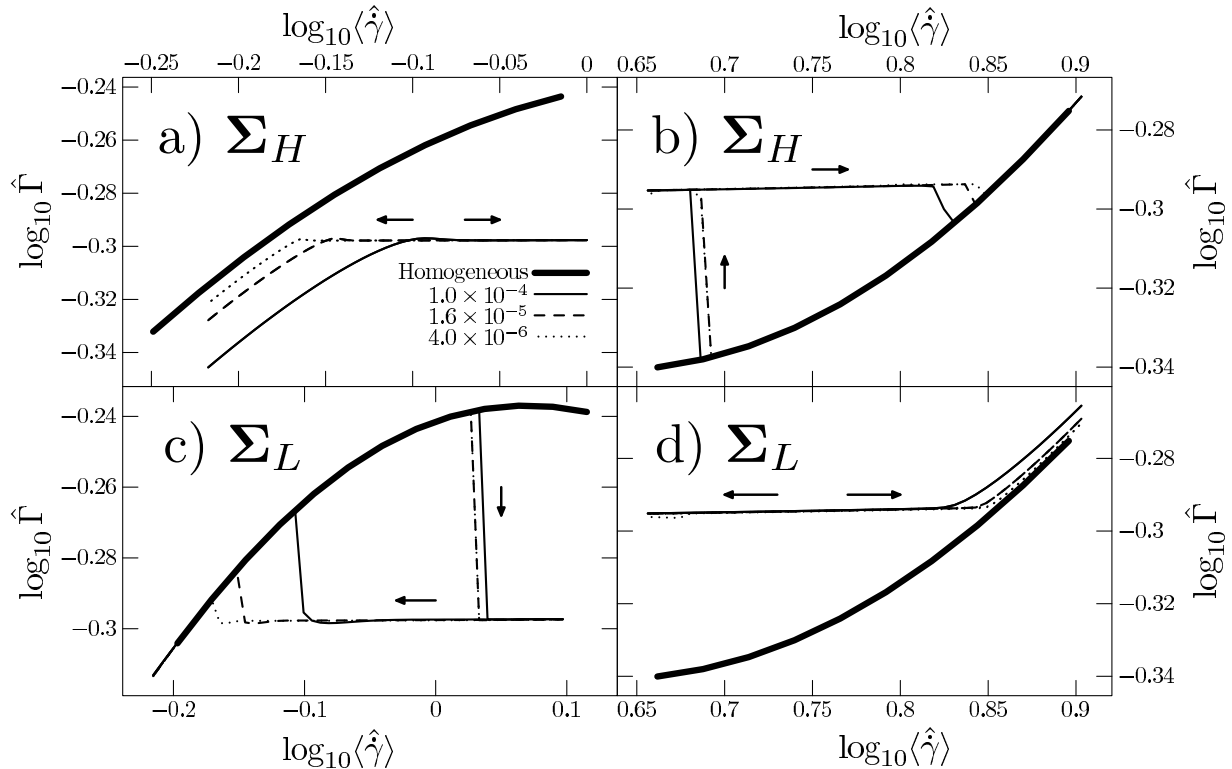


FIG. 9: Constitutive curve and flow curves for the DJS model, for  $q = 0.005$ , and diffusion constants  $\hat{\mathcal{D}} = 1.0 \times 10^{-4}$  (solid),  $1.6 \times 10^{-5}$  (dashed) and  $4.0 \times 10^{-6}$  (dotted). (a,c) show the start of the plateau, while (b,d) show the end; and the boundary condition  $\Sigma_0$  equal to the viscoelastic stress  $\Sigma_H$  or  $\Sigma_L$  of, respectively, the high (a,b) or low (c,d) shear rate branch. The arrows indicate the direction in which the hysteresis loops are encircled (b,c) or the shear rate swept (a,d).

$$\Sigma_L : \quad (\Sigma_{L\theta\theta}, \Sigma_{Lrr}, \Sigma_{Lr\theta}) = (0.40, -0.21, 0.47) \quad \text{low shear rate branch} \quad (\text{VI.1a})$$

$$\Sigma_H : \quad (\Sigma_{H\theta\theta}, \Sigma_{Hrr}, \Sigma_{Hr\theta}) = (1.4, -0.75, 0.12) \quad \text{high shear rate branch,} \quad (\text{VI.1b})$$

and for the DRP model with  $\beta = 0$  and  $\epsilon = 0.01$  they are

$$\Sigma_L : \quad (\Sigma_{L\theta\theta}, \Sigma_{Lrr}, \Sigma_{Lr\theta}) = (0.81, -0.40, 0.60) \quad \text{low shear rate branch} \quad (\text{VI.2a})$$

$$\Sigma_H : \quad (\Sigma_{H\theta\theta}, \Sigma_{Hrr}, \Sigma_{Hr\theta}) = (1.7, -0.85, 0.44) \quad \text{high shear rate branch.} \quad (\text{VI.2b})$$

The extent of the hysteresis depends on the magnitude of the diffusion constant. Fig. 9 shows how  $\mathcal{D}$ , or equivalently the width  $\ell \sim \sqrt{\mathcal{D}}$  of the interface, influences the flow curves, for the two different boundary conditions of the DJS model. As the diffusion constant is reduced the heterogeneous flow curve approaches the homogeneous constitutive curve, and the degree of hysteresis decreases. This occurs for both the DRP (not shown) and DJS models. As noted above, this is because the interfacial layer occupies a progressively smaller fraction of the sample: the separation between the heterogeneous flow curve and homogeneous constitutive curve is proportional to  $\sqrt{\mathcal{D}} \sim \ell$ . This behavior should be contrasted with the Neumann case (Fig. 4), in which the flow curve

matches the constitutive curve for all  $\mathcal{D}$ .

## B. The effects of flow geometry and total stress gradient

### 1. Dirichlet boundary conditions

For a flow cell with a stress gradient, such as a cylindrical Couette cell, the interface in simple banding flow with Neumann boundary conditions lies at the position in the flow cell where the stress is equal to the selected total shear stress  $T_{r\theta}^*$ . With Dirichlet boundary conditions there are typically three bands and two interfaces,

because of the boundary layer imposed by the wall. We now study how the position of these bands varies as a function of the flow geometry and diffusion coefficient. Calculations were performed using the second protocol outlined in Sec. IV: starting up from rest with a suitable initial condition and evolving at fixed average strain rate to steady state. The boundary condition  $\Sigma_0$  was chosen to be either  $\Sigma_L$  or  $\Sigma_H$ .

In a flat geometry the phase near the wall is that most similar to the imposed boundary conditions, ideally resulting in a symmetric three band configuration stabilized by the very weak interactions of the interfaces with the walls. For strong enough curvature we expect the high shear rate phase to preferentially occupy the high stress regions. This is indeed what we find. For increasing curvature the three band configuration becomes increasingly asymmetric, as seen in both the DJS and DRP models (Figs. 10 and 11). A crossover from three banded to nearly two-banded behavior occurs at smaller  $q$  values (weaker curvature) for smaller  $\mathcal{D}$ . The three band state is most pronounced when the boundaries induce the low shear rate phase  $\Sigma_0$ .

The behavior described above can be rationalized as follows. For a symmetric flat system ( $q = 0$ ) both interfaces lie at the selected stress, since the total stress is uniform. The very weak effect a slightly curved system  $q \gtrsim 0$  breaks the symmetry of the shear banded structure, and only one (or neither) interface can lie at the selected stress; at the same time, the stress gradient would favor locating the high shear band near the inner cylinder. These two tendencies adjust the band configuration such that high (low) shear rate bands grow (shrink) near the inner cylinder and shrink (grow) near the outer cylinder. For example, for boundary conditions that favor the low shear rate band the band near the inner cylinder should become narrower with increasing curvature (as seen in Fig. 10a,c). However, a larger diffusion coefficient broadens the interfaces between bands, which means that the walls can strongly influence the bulk behavior. Correspondingly, for large  $\mathcal{D}$  the position of the center shear band moves more smoothly as a function of the geometric curvature  $q$ . For example, compare Fig. 10c ( $\mathcal{D} = 10^{-2}$ ) with Fig. 10a ( $\hat{\mathcal{D}} = 10^{-3}$ ).

Just as for Fig. 5, the effective cone angles are again shown in the upper abscissas in Figures 10 and 11. For small diffusion constant, Fig. 10a shows that in cone and plate flow with a typical angle of  $\theta = 4^\circ$ , a three banded structure could be observed, while the third band would be much smaller in Couette flow with  $\Delta R/R = 0.05$ , and could easily be interpreted as a two band structure. Thus, a three band structure, as observed by Britton and Callaghan [9], could be consistent with boundary conditions that favor the low shear rate form of the viscoelastic stress.

Finally, we note that these results are consistent with those of Cook and co-workers [25, 26], who studied a two-fluid model of wormlike micelles. In cylindrical Couette flow with  $q = 0.064$ ,  $\hat{\mathcal{D}} = 10^{-3}$  they also found a three-

band state, but did not explore the effects of geometry or diffusion coefficient.

## 2. Competing boundary conditions

Finally we allow surface anchoring to compete with the bulk spatial gradients, according to Equation (III.5). By varying  $W$ , or equivalently the extrapolation length  $\xi = D\tau/W$ , the boundary condition at the wall can be tuned smoothly from a fixed value  $\Sigma_0$  as  $W \rightarrow \infty$  ( $\xi \rightarrow 0$ ) to zero gradient as  $W \rightarrow 0$  ( $\xi \rightarrow \infty$ ). As above, we consider boundary conditions in which  $\Sigma_0$  is the viscoelastic stress of either the low or high shear rate branch,  $\Sigma_L$  or  $\Sigma_H$ . We vary both the extrapolation length  $\xi$  and the geometric curvature  $q$ , and monitor the behavior of a well-formed shear band, with an imposed average shear rate such that the high and low shear rate bands are roughly the same size. We find qualitatively similar results for both the DJS and DRP models.

Figure 12 shows the results for the DRP model for boundary conditions  $\Sigma_0 = \Sigma_L$ . Consider changing the anchoring strength at fixed  $q$  (profiles {i,ii,iii,iv} in Fig. 12). For weak anchoring strength (large  $\xi$ , profile a), the effective boundary condition at the inner wall is nearly zero gradient (Neumann), and the high shear rate band lies near the inner wall. Upon increasing the anchoring strength (reducing  $\xi$ ) the boundary conditions deviates slightly from Neumann conditions before reverting to Dirichlet conditions for strong enough anchoring (small  $\xi$ ), thus imposing  $\Sigma_L$  at the wall and inducing three bands. The crossover occurs when the extrapolation length is of order the interfacial thickness,  $\xi \simeq \ell = \sqrt{D\tau}$ .

In all cases the interface closest to the outer cylinder (at larger  $x$ ) remains at the selected stress  $T_{r\theta}^*$  while the inner interface is at a higher stress, as can be seen by the intersections of the shear rate profiles as a function of total stress in Fig. 12(iii,iv). Upon approaching the flat limit both interfaces approach the selected stress. The degree of anchoring strength above which the boundary conditions become Dirichlet-like depends weakly on the geometric curvature. For a more highly curved geometry a larger anchoring potential (smaller  $\xi$ ) is required to enforce the boundary condition  $\Sigma_0 = \Sigma_L$ . This is because the higher stress at the inner wall competes with the tendency of the wall to induce the low shear rate band.

We have examined whether or not this ‘‘surface transition’’ between different anchoring states has an appreciable mechanical signal. The torque at the inner cylinder, as would be measured in an experiment, changes by of order 1% upon tuning between Neumann-like and Dirichlet-like boundary conditions; hence this has only a small effect on the macroscopic response of the system. The degree of anchoring is only weakly affected by the overall average shear rate; only when one interface approaches the wall is there an effect, and again this is reflected in only a small change in the measured total

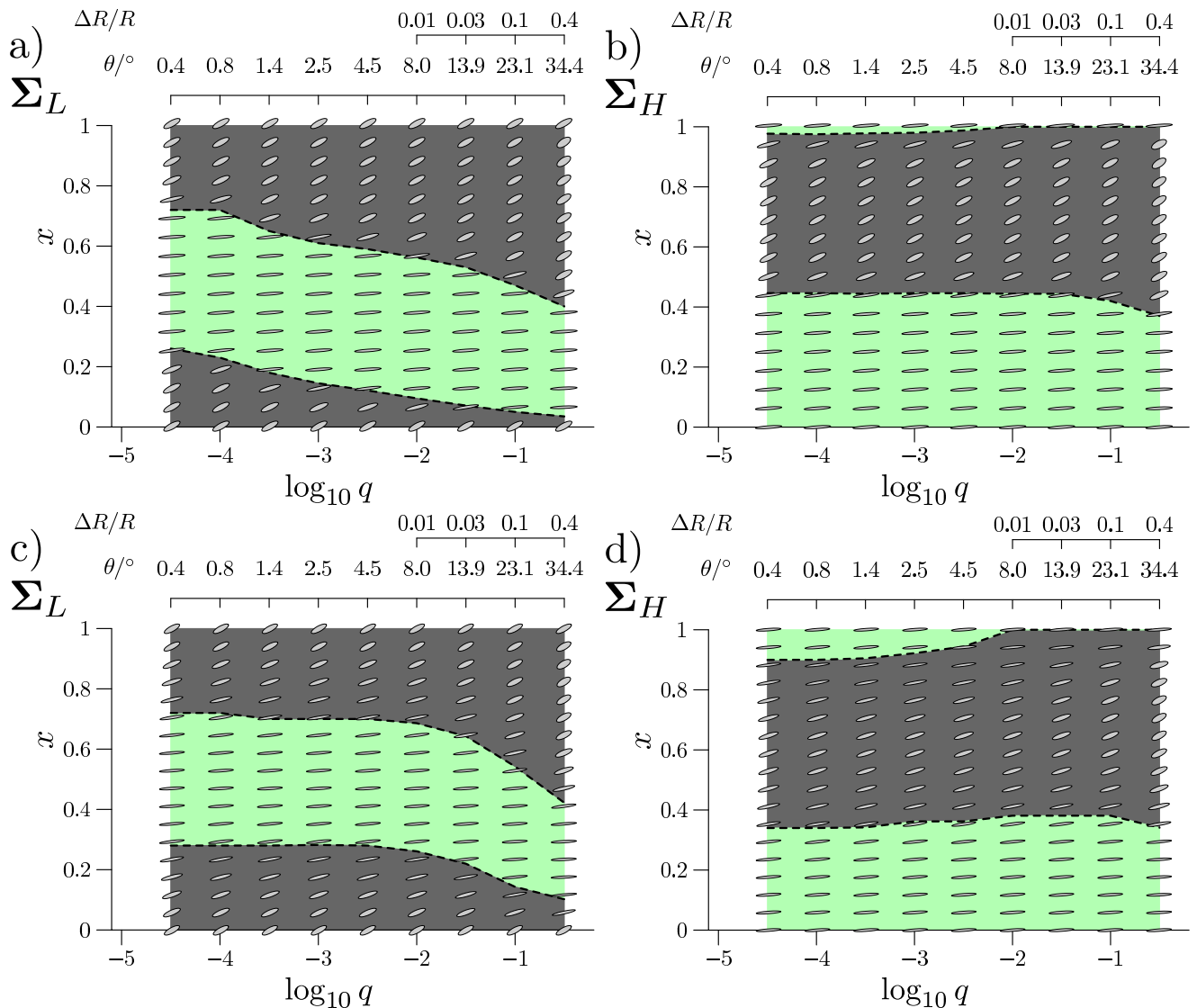


FIG. 10: Viscoelastic stress ellipses as a function of position  $x$  and curvature  $q$ , for the DJS model with  $\langle \hat{\gamma} \rangle = 3.8 (\log_{10} \langle \hat{\gamma} \rangle = 0.58)$ . The diffusion constant is  $\hat{D} = 10^{-3}$  (a,b) and  $\hat{D} = 10^{-2}$  (c,d). The high shear rate phase is lightly shaded (green), and the low shear rate phase is dark grey. The boundary conditions are  $\Sigma_0 = \Sigma_H$  or  $\Sigma_L$ . The top axes show the equivalent Couette cylinder gap  $\Delta R$  relative to the inner cylinder radius  $R$ , and equivalent cone angles  $\theta$  if we assume that the stress gradient mimics that found in cone and plate flow.

stress.

Similar behavior is found for  $\Sigma_0 = \Sigma_H$ , with a few differences. In this case the interface closest to the inner wall lies at the selected stress, as can be seen in the profiles for  $\dot{\gamma}(T_{r\theta})$  in Fig. 13; this implies that for both  $\Sigma_0 = \Sigma_L$  and  $\Sigma_0 = \Sigma_H$  the interface that separates an inner band at high shear rate from an outer band at low shear rate lies at the selected stress. Another difference is that the geometric curvature  $q$  has a very weak effect on the effective boundary condition, because the higher stress at the inner wall doesn't compete with the boundary condition.

## VII. CONCLUSION

### A. Summary of results

We have performed a numerical study of the effect of the boundary conditions on the viscoelastic (polymer) stress for the diffusive Johnson-Segalman (DJS) and diffusive non-stretching Rolie-Poly (DRP) models, in cylindrical Couette flow. The main results are the following:

- a. Dirichlet boundary conditions (fixed  $\Sigma_0$ ) influence the possible hysteretic behavior for the flow curve: for  $\Sigma_0$  resembling the high shear rate phase a hysteresis loop occurs at the high shear rate end of

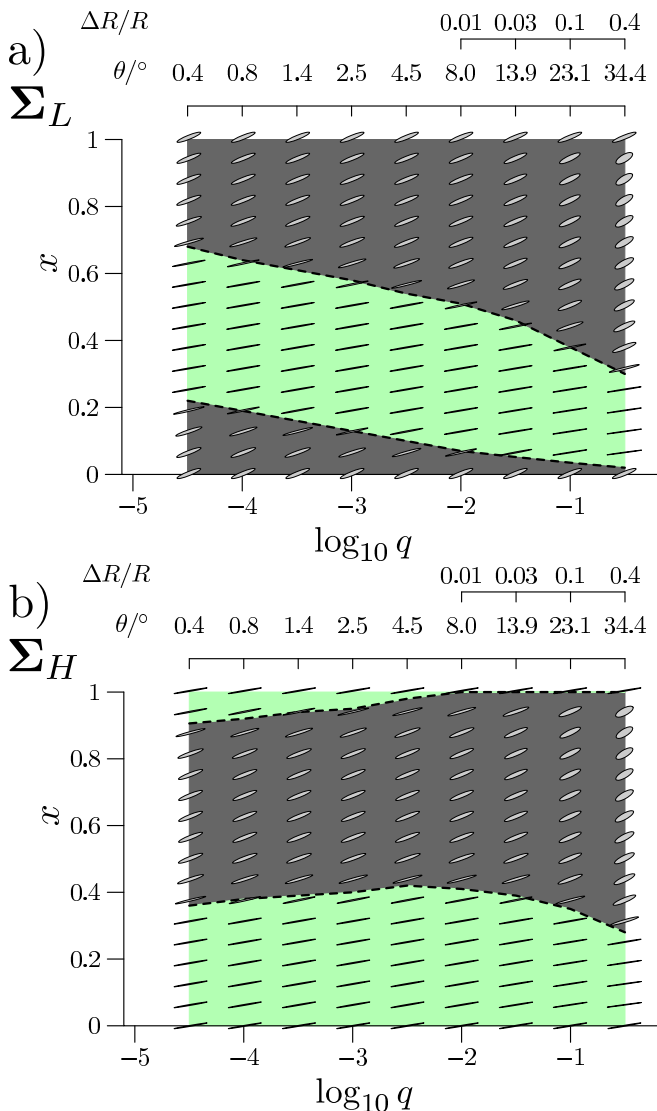


FIG. 11: Viscoelastic stress profiles, shown as ellipses as a function of position  $x$ , for the DRP model in different degrees of curvature  $q$ , for  $\hat{D} = 10^{-3}$  and  $\langle \hat{\gamma} \rangle = 9$  ( $\log_{10} \langle \hat{\gamma} \rangle = 0.95$ ). The high shear rate phase is lightly shaded (green), and the low shear rate phase is dark grey. The boundary conditions are  $\Sigma_0 = \Sigma_H$  (right) or  $\Sigma_0 = \Sigma_L$  (left). The upper abscissa shows the equivalent geometrical parameters for cylindrical Couette ( $\Delta R/R$ ) and cone and plate ( $\theta$ ) geometries.

the stress plateau, while for  $\Sigma_0$  resembling the low shear rate phase a hysteresis loop occurs at the low shear rate end of the stress plateau.

- b. The walls can induce a lubrication (or thickened) layer that suppresses the hysteresis that would occur upon sweeping the shear rate into a banded state from a flow branch with characteristic dissimilar to the boundary layer.
- c. As with Neumann boundary conditions, the hysteresis loop shrinks with decreasing diffusion con-

stant.

- d. For Dirichlet boundary conditions the stress distribution in the gap depends on both the boundary condition and the stress gradient. A three-band state is stable for weak curvature such as cone and plate flow, which becomes more asymmetric or even two-banded as the stress gradient increases to that of typical cylindrical Couette flow.
- e. For mixed boundary conditions the strength of wall anchoring determines whether the effective boundary conditions are Neumann or Dirichlet. The crossover occurs when the extrapolation  $\xi$  length is of order the interfacial width  $\ell$ , or  $W \simeq \sqrt{D\tau}$ .

This is the first use of the DRP model to study shear banding. The model is microscopically motivated by either polymer solutions or wormlike micelles, and its non-monotonic behavior arises from better-understood physics (convected constraint release competing with tube alignment) than that in the DJS model (where the “slip parameter”  $a$  is necessary). The physics of convected constraint release yields a less well aligned state in the high shear rate phase, whose alignment angle, compared to that from the DJS model, is closer to that seen experimentally [16]. Numerically, interfaces typically travel faster in the DRP model than in the DJS model, and hence can be more quickly and reliably calculated, and the DRP model is more robust to large time steps and inhomogeneous initial conditions.

## B. Discussion and outlook

An important open problem is the relation of wall slip to shear banding flows. A boundary condition such as  $\Sigma_0 \sim \hat{\mathbf{v}}\hat{\mathbf{v}}$ , which imposes alignment parallel to the flow direction  $\mathbf{v}$ , yields velocity profiles that could be interpreted in terms of wall slip, because of the lack of a shear component of the viscoelastic stress at the wall. The banding profiles in Fig. 6, for example, have small regions of very high shear rate near either wall. Becu *et al.* measured a correlation between band motion and wall slip in a solution of CTAB wormlike micelles [48, 49]. They used smooth and sand-blasted Couette cells and found different velocity profiles and interface dynamics in the banding regime. It is possible that these differences are a combination of both different intrinsic boundary conditions and different degrees of wall slip.

Another signature of this effect would lie in the measurement of the low shear rate branch before shear banding occurs: the flow cell (for example, the smooth one) that induced tangential ordering would have a smaller stress at a given shear rate, due to the associated lubrication layer, than would the flow cell (*e.g.* the sand-blasted one) with less preference for the high shear rate branch.

In related work, Manneville and co-workers [50] measured the rheology and velocity profiles in a triblock

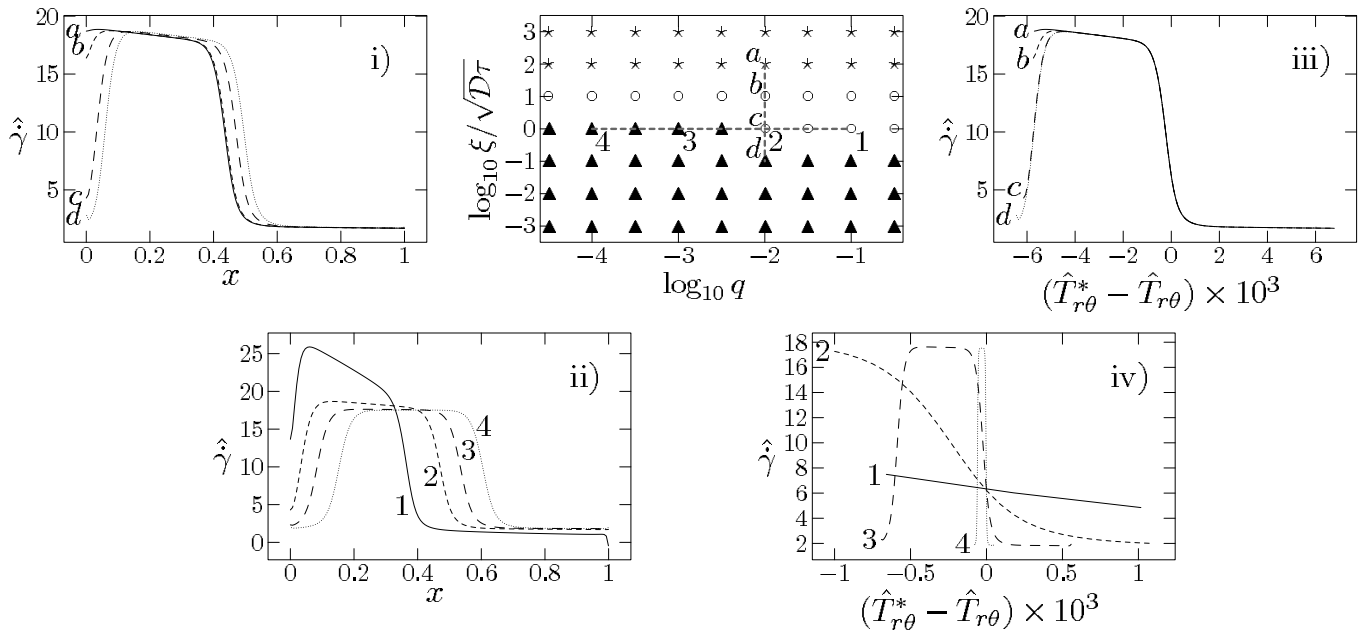


FIG. 12: Flow profiles as a function of extrapolation length  $\xi = \hat{D}/\hat{W}$  and geometrical curvature  $q$  for the DRP model with mixed boundary conditions, for  $\langle \hat{\gamma} \rangle = 3.8$ ,  $\hat{D} = 7 \times 10^{-4}$  and  $\Sigma_0$  equal to the viscoelastic stress in the low shear rate branch,  $\Sigma_L$ . (i,iii) Shear rate  $\hat{\gamma}$  as a function of position  $x$ ; the labels  $a, \dots, d$  and  $1, \dots, 4$  refer to the points in the  $(\xi, q)$  parameter map in the center. (ii, iv): Shear rate as a function of the deviation of the total stress  $T_{r\theta} \sim e^{-2qx}$  from the selected stress  $T^*$ . (Center) Map of different effective boundary conditions as a function of extrapolation length  $\xi$  and curvature  $q$ :  $\star \simeq$  Neumann boundary conditions;  $\blacktriangle \simeq$  Dirichlet conditions;  $\circ \simeq$  mixed boundary conditions.

copolymer solution that forms wormlike micelles, using heterodyne light scattering. At  $T = 37^\circ$  the solutions did not shear band but did exhibit wall slip, within the  $\simeq 40 \mu\text{m}$  resolution of the technique. At a higher temperature  $T = 39.4^\circ$ , believed to correspond to longer micelles, the solution exhibited a three-band shear banding scenario with the high shear rate bands near the inner wall. These data are consistent with the walls inducing preferential ordering parallel to the walls with increasing temperature and longer micelles, so that that wall slip could be either “true” slip or an apparent slip due to the specific nature of the boundary conditions. Other mechanisms for slip, such as micelles detaching from the wall and disentangling near the surface, may also be relevant in this case [51, 52, 53].

In principle, mixed boundary conditions allows for multiple stable inhomogeneous solutions, which could be susceptible to non-linear perturbations such as local flow inhomogeneities, thermal fluctuations, asperities, motor noise, *etc.* The solutions found here switched between effective Neumann and Dirichlet boundary conditions as a function of anchoring strength, but easily externally controllable quantities such as flow geometry and imposed average shear rate had very weak effects, and the total stress differences between the two kinds of anchoring was small (typically less than a percent).

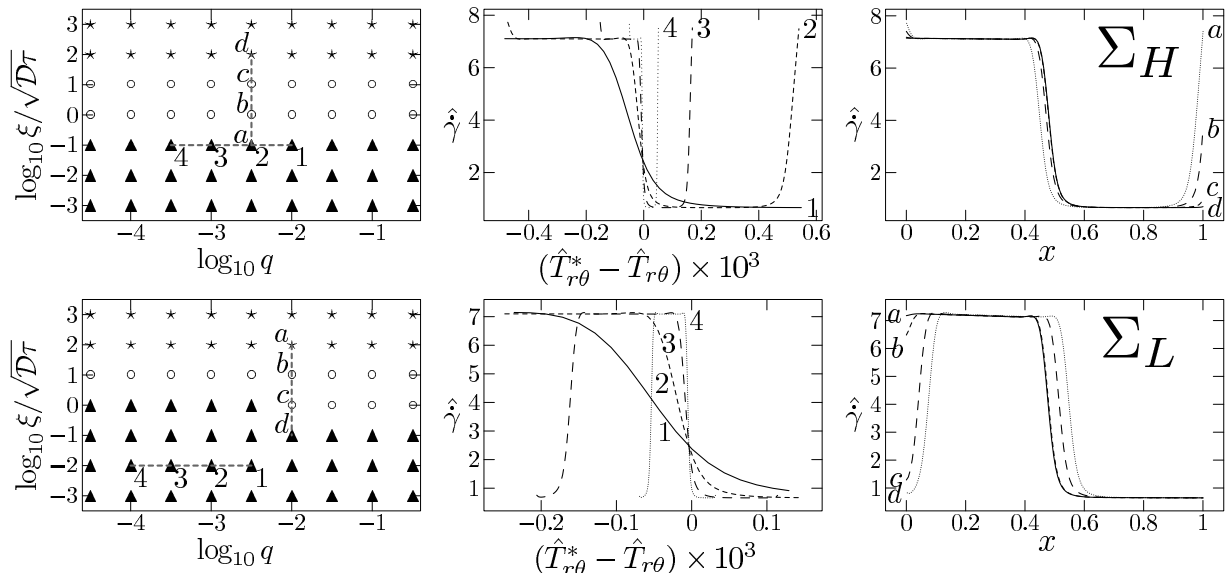
One could, in principle, envision more complex surface anchoring behavior, such as a multi-welled wall po-

tential that would govern a surface phase transition between two different wall orientations  $\Sigma_{0a}$  and  $\Sigma_{0b}$ . The order parameter in the bulk could then, in principle, drive the boundary condition between the two potential wells, which would then have more dramatic consequences. Such a bulk-driven wall transition could also be triggered by fluctuations, would then lead to erratic band motion coupled to an apparent wall slip, depending on the details of the specific measurements.

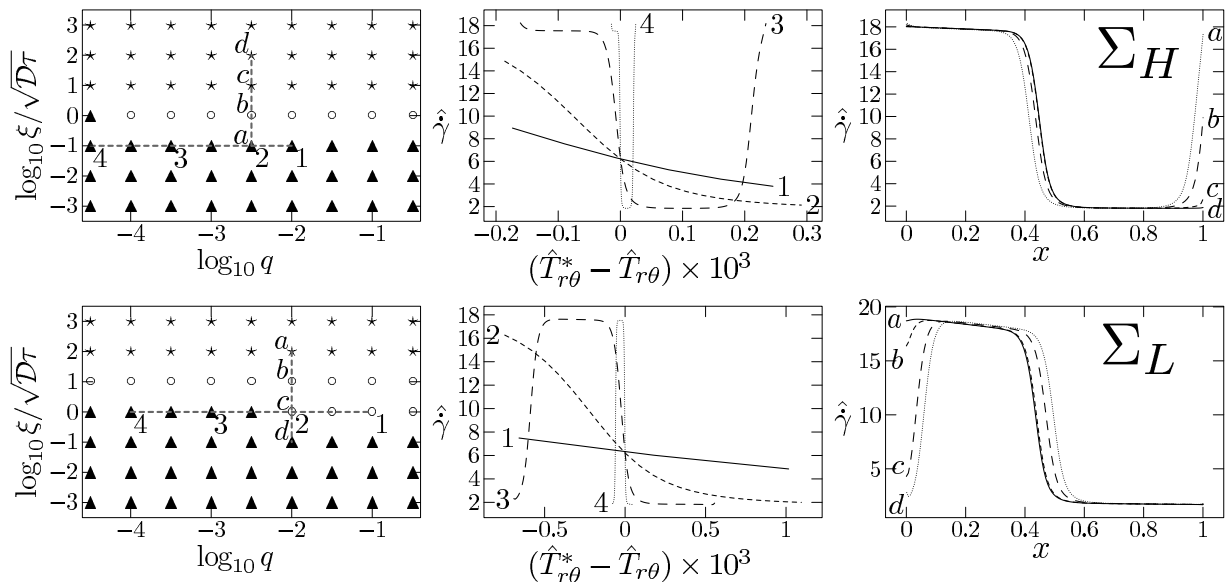
Another physical effect that has not been considered here is the concentration degree of freedom, which in some cases probably leads to a depletion layer (or in rarer cases perhaps coagulation) layer near the surface. Ref. [26] included concentration in their treatment, but without any specific wall-concentration coupling aside from a zero flux condition. This is a promising direction for future study.

## VIII. ACKNOWLEDGEMENTS

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(13.1) DJS model with mixed boundary conditions.



(13.2) DRP model with mixed boundary conditions.

FIG. 13: Left: Map of effective boundary conditions as a function of curvature  $q$  and extrapolation length  $\xi$  and selected shear rate profiles as a function of position and total stress, for  $\mathcal{D} = 7 \times 10^{-4}$ :  $\star \simeq$  Neumann boundary conditions;  $\blacktriangle \simeq$  Dirichlet conditions;  $\circ \simeq$  intermediate boundary conditions. Shear rate as a function of local total stress (middle) and position (right) for the DJS and DRP models with boundary conditions  $\Sigma_0 = \Sigma_H$  and  $\Sigma_0 = \Sigma_L$ .

## APPENDIX A: VISCOELASTIC STRESS BOUNDARY CONDITIONS

The dynamics of the DJS or DRP model are formulated in terms of the viscoelastic (or polymeric) contribution to the total stress. The free energy of a polymer solution is more commonly expressed in terms of the nonlinear deformation tensor  $\mathbf{\Lambda}$ . A simple free energy functional that includes surface, bulk, and distortion elastic

terms is

$$F = \frac{1}{2} \int d^3r \left[ G \mathbf{\Lambda}^2 + K (\nabla \mathbf{\Lambda})^2 \right] + \frac{1}{2} \int_S d^2r W_0 (\mathbf{\Lambda} - \mathbf{\Lambda}_0)^2, \quad (\text{A.1})$$

where  $K$  is analogous to the Frank elastic terms that penalize distortions of order parameter in nematic liquid crystals, and  $G$  is the modulus. For ordinary polymer (rubber) elasticity the deformation tensor is  $\mathbf{\Lambda} \simeq \langle \mathbf{R}\mathbf{R} \rangle / (Nb^2) - \frac{1}{3} \mathbf{I}$ , where  $N$  is the number of Kuhn steps



per strand,  $b$  is the Kuhn length, and  $\mathbf{R}$  is the end-to-end distance of the strand, and the modulus is thus given by  $G \simeq 3k_B T c_s$ , where  $c_s$  is the number of strands per unit volume. A simple dynamics and boundary conditions for such a free energy (based on the analogy with liquid crystals) is

$$\partial_t \mathbf{\Lambda} = -\frac{1}{\zeta} [G\mathbf{\Lambda} - K\nabla^2 \mathbf{\Lambda}] \quad (\text{A.2})$$

$$0 = W_0 (\mathbf{\Lambda} - \mathbf{\Lambda}_0) + K\nabla^2 \mathbf{\Lambda}. \quad (\text{A.3})$$

$$F = \frac{1}{2} \int d^3r \left[ \frac{1}{G} \mathbf{\Sigma}^2 + \frac{K}{G^2} (\nabla \mathbf{\Sigma})^2 \right] + \frac{1}{2} \int_S d^2r \frac{W_0}{G^2} (\mathbf{\Sigma} - \mathbf{\Sigma}_0)^2 \quad (\text{A.4})$$

$$\partial_t \mathbf{\Sigma} = -\frac{G}{\zeta} \left[ \mathbf{\Sigma} - \frac{K}{G} \nabla^2 \mathbf{\Sigma} \right] + \dots \quad (\text{A.5})$$

$$0 = W_0 (\mathbf{\Sigma} - \mathbf{\Sigma}_0) + K \hat{\mathbf{n}} \cdot \nabla \mathbf{\Sigma}, \quad (\text{A.6})$$

where  $\hat{\mathbf{n}}$  is the outward unit vector normal to the surface. On the other hand, the dynamics of the DJS model is

$$\partial_t \mathbf{\Sigma} = -\frac{1}{\tau} [\mathbf{\Sigma} + \mathcal{D}\tau \nabla^2 \mathbf{\Sigma}] + \dots, \quad (\text{A.7})$$

from which we identify  $\tau = \zeta/G$  and  $\mathcal{D}\tau = K/G$ . Thus, the boundary condition for the DJS model, which follows from this correspondence and from Eq. (A.6), is

$$0 = \frac{W_0}{G} (\mathbf{\Sigma} - \mathbf{\Sigma}_0) + \mathcal{D}\tau \nabla \mathbf{\Sigma}. \quad (\text{A.8})$$

Here  $W_0$  is the surface anchoring term that penalizes the strain variable at the surface according to Eq. (A.4). In the main text we have used  $W \equiv W_0/G$ .

## APPENDIX B: ESTIMATES OF $W$ AND $\mathcal{D}$

One mechanism for such a potential is the steric exclusion of micelles at the wall, which would favor an oblate deformation  $\mathbf{\Lambda}$  and hence an oblate viscoelastic stress. Such an effect was found in Monte Carlo simulations of polymer melts, which showed a decrease in the radius of gyration  $R_g^\perp$  normal to the wall of 20% and a corresponding increase parallel to the wall, independent of the attraction of chain segments to the wall [54]. Assuming weak perturbations due to the wall potential, the free energy governing subsequent perturbations is governed by the entropic elasticity of polymer chains:  $F/\text{chain} \simeq \frac{3}{2} k_B T \text{Tr} (\mathbf{\Lambda} - \mathbf{\Lambda}_0)^2$ . Converting to a wall potential according to

$$\frac{F_{\text{wall}}}{\text{Area}} \simeq \frac{F}{\text{chain}} c_s 2R_g \quad (\text{B.1})$$

$$= \frac{3k_B T c_s b}{\sqrt{6N}} \text{Tr} (\mathbf{\Lambda} - \mathbf{\Lambda}_0)^2, \quad (\text{B.2})$$

Since the viscoelastic stress  $\mathbf{\Sigma}$  is related to the strain by a modulus,  $\mathbf{\Sigma} = G\mathbf{\Lambda}$ , we can rewrite the equations above as

we estimate  $W_0 \simeq k_B T c_s b \sqrt{6/N}$  (according to Eq. A.4), and  $W \simeq b \sqrt{2N/3} = 2R_g$ . Here,  $c_s$  is the number of strands per unit volume.

The stress in a solution of semiflexible polymers, such as wormlike micelles, has a contribution  $\mathbf{\Sigma}$  from the tube orientation  $\langle \mathbf{u}\mathbf{u} \rangle$ , as well as a liquid crystalline contribution  $\mathbf{\Sigma}_{LC}$ . Although liquid crystalline effects are only appreciable near an isotropic-to-nematic transition, they are nonetheless present. Hence, the total stress takes the form [55, 56]

$$\mathbf{T} = \mathbf{\Sigma} + 2\eta \mathbf{D} + \frac{\delta F_{LC}}{\delta \mathbf{Q}} + \dots, \quad (\text{B.3})$$

where  $F_{LC}$  is the liquid crystalline contribution to the free energy and the neglected terms are non-linear in both  $\mathbf{Q}$  and  $\mathbf{D}$ .

For a semiflexible polymer solution  $F_{LC}$  has been calculated by Liu and Fredrickson, and depends on the persistence length  $\ell_p$ . To lowest order in the nematic order parameter  $\mathbf{Q}$  it is

$$F_{LC} = \frac{1}{2} \int_V [a \text{Tr} (\mathbf{Q}^2) + \mathcal{K} \partial_\mu \mathbf{Q} : \partial_\mu \mathbf{Q}], \quad (\text{B.4})$$

where only a single Frank constant has been included here. This leads to a liquid crystalline contribution to the stress of the form

$$\mathbf{\Sigma}_{LC} = a \mathbf{Q} - \mathcal{K} \nabla^2 \mathbf{Q}. \quad (\text{B.5})$$

In this case the parameters are [22]

$$\mathcal{K} = \frac{45}{126\pi} \frac{k_B T \ell_p \phi}{D^2}, \quad a \simeq \frac{45\phi k_B T}{\pi D^2 \ell_p}, \quad (\text{B.6})$$

where  $\phi$  is the volume fraction  $\phi$  [22] and  $D$  is the micellar diameter. For wormlike micelles  $\ell_p \simeq 20$  nm,  $D \simeq 2$  nm

[33], leading to  $\mathcal{K} \simeq 0.37 \phi k_B T / \text{nm}$ . Upon converting the entire description to one in terms of the total micellar stress  $\Sigma_{\text{tot}} = \Sigma + \Sigma_{LC}$  and performing a gradient expansion, one finds the following contribution to the stress diffusion coefficient due to Frank-like elasticity:

$$\mathcal{D}\tau \simeq \frac{\mathcal{K}}{a} \simeq \frac{\ell_p^2}{126}. \quad (\text{B.7})$$

There may of course, be other non-local contributions, which could depend on micellar size, concentration, hydrodynamics, or other physics.

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