A rheological theory for liquid crystal thin films

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Abstract A macroscopic rheological theory for compressible isothermal nematic liquid crystal films is developed and used to characterize the interfacial elastic, viscous, and viscoelastic material properties. The derived expression for the film stress tensor includes elastic and viscous components. The asymmetric film viscous stress tensor takes into account the nematic ordering and is given in terms of the film rate of deformation and the surface Jau mann derivative. The material function that describes the anisotropic viscoelasticity is the dynamic film tension, which includes the film tension and dilational viscosities. Viscous dissipation due to film compressibility is described by the anisotropic dilational viscosity. Three characteristic film shear viscosities are defined according to whether the nematic orientation is along the velocity direction, the velocity gradient, or the unit normal. In addition the dependence of the rheological functions on curvature and film thickness has been identified. The rheological theory provides a theoretical framework to future studies of thin liquid crystal film stability and hydrodynamics, and liquid crystal foam rheology.

Key words Nematic liquid crystals · Thin films · Film viscoelasticity · Anisotropic film dilational viscosities · Anisotropic film shear viscosities

Introduction

Interfacial viscoelasticity is one of the foundational elements of interfacial dynamics and interfacial rheology (Slattery 1990; Edwards et al. 1991). For interfaces between viscous Newtonian fluids it has been found that the main interfacial material properties are the interfacial tension, the interfacial dilational viscosity, and the interfacial shear viscosity. These material properties enter into the description of a wide range of interfacial phenomena such as interfacial stability, thin liquid film hydrodynamics, thin liquid film stability, and emulsion and foam rheology (Slattery 1990; Edwards et al. 1991). This paper is concerned with the rheology of thin nematic liquid crystalline films.

The physics of nematic liquid crystalline surfaces is currently an active area of research (Jerome 1998; Sonin 1995) since many applications of liquid crystalline materials involve multiphase systems, where interfaces play significant roles. To model dynamical interfacial phenomena involving nematic liquid crystal phases requires incorporation of viscoelastic modes that describe material properties such as the dynamic interfacial tension and interfacial viscosities. At the present time interfacial orientation phenomena and orientational transitions are well characterized experimentally and theoretically (Jerome 1998; Sonin 1995; Faetti 1991; Barbero and Durand 1996). On the other hand a fundamental understanding of interfacial dissipative phenomena is less developed (see, for example, Rey 1999a).
A hallmark of the bulk mechanical behavior of nematic liquid crystalline materials is the anisotropic character of the viscoelastic modes (Larson 1999). The anisotropic Frank elasticity is now well established and responsible for many pattern selection phenomena in the presence of external fields (Larson 1999). A well-known example is the splay-avoidance mechanism active in deformations of nematic polymers. Viscous anisotropies are also well-characterized experimentally (de Gennes and Prost 1993; Lee and Meyer 1991; Srinivasarao 1995; Larson 1999) and theoretically (Ehrentraut and Hess 1995; Lee and Meyer 1991). One prominent example here is the ordering in magnitude of the three Miesowicz shearc viscosities, where the largest flow resistance is obtained when the average molecular orientation is fixed along the velocity gradient direction and the smallest when the average molecular orientation is fixed along the vorticity. In addition the shear flow-alignment properties that dictate whether the average orientation is close to the flow direction depends in part on the difference between two Miesowicz shear viscosities. These few examples show the prominence of anisotropic viscoelasticity in the bulk behavior of nematic liquid crystals.

Recently it has been shown that anisotropic viscoelasticity plays a similarly prominent role in the interfacial behavior of interfaces involving nematic liquid crystals. The anisotropic elastic nature of nematic interfaces is well understood (Jerome 1998; Sonin 1995). Elastic models for deforming nematic-viscous fluid interfaces have already been proposed (Jenkins and Barrat 1974; Ericksen 1979; Virga 1994; Rey 2000) and used to describe nemato-capillarity phenomena such as bending stresses and Marangoni flows (Rey 1999b, 2000). Models for liquid crystals in 2D have also been given (Papenfuss and Muschik 1992). The presence of interfacial viscous modes and compressibility has been recently characterized (Rey 2000). Given advances on studies of liquid crystalline surfaces and interfaces, the framework already developed can naturally be used to describe the behavior of thin nematic liquid crystalline films.

Thin films are formed during spreading of liquid over solids, between approaching liquid droplets, and in the formation of foams (Edwards et al. 1991). Thin liquid films arise in a number of practical applications ranging from foams, emulsions, filtration of solids from a suspension, among others and understanding them is vital to the food, pharmaceutical, cosmetics, and mining industries. Regarding liquid crystal thin films they are currently used as foam stabilizers (Myers 1999) but many other applications are likely to materialize in the near future. Thus a better fundamental understanding of liquid crystal thin film rheology is highly desirable.

The objectives of this paper are: (1) to derive a rheological model for isothermal nematic liquid crystal- line thin films, and (2) to define and characterize the main elastic, viscous, and viscoelastic material properties of these films. The approach is based on the direct method to model thin films as single membranes and the derived balance and constitutive equations are analogous to those of interfaces. The reader is referred to Rey (2000) for details on modeling the viscoelasticity of nematic interfaces, and to Coutris (1993) and Dikmen (1982) for technical details on direct models for thin films.

The organization of this paper is as follows. The next main section presents the balance equations and constitutive equations for interfaces between nematic liquid crystals and isotropic viscous fluids. Subsections define the geometric properties of the nematic-isotropic viscous fluid interface, present the objective kinematic measures that include the surface rate of deformation tensor and the surface Jaumann derivative of the nematic orientation field, discuss the interfacial mass, angular momentum, and linear momentum balance equations, discuss the components of the surface elastic stress tensor, and derive the constitutive equations for the viscous interfacial extra stress tensor and the viscous interfacial molecular field. The next main section presents and discusses the balance equations and constitutive equations for nematic thin films. This section describes the single nematic membrane model. Subsections discuss the film mass, angular momentum, and linear momentum balance equations, also discuss in detail the elastic stress tensor for nematic films, and use the film dissipation function to derive constitutive equations for the film extra stress tensor and for the film viscous molecular field. The penultimate main section presents and discusses the characteristic rheological functions of nematic thin films, including the dynamic film tension, the film dilational viscosity, and the film shear viscosity. The final section presents the conclusions.

**Balance equations and constitutive equations for nematic interfaces**

In this section we consider the interfacial torque balance equation and interfacial linear momentum balance equations that govern the dynamics of the interfacial director orientation, and the shape of the deforming interface between a nematic liquid crystal (N) and an isotropic viscous fluid (I), under isothermal conditions.

**Nematic-isotropic fluid interface**

The nematic-isotropic viscous fluid interface, denoted by N/I, is characterized by a unit normal κ, directed from the nematic (N) phase into the isotropic (I) phase, whose mean curvature is given by Edwards et al. (1991):