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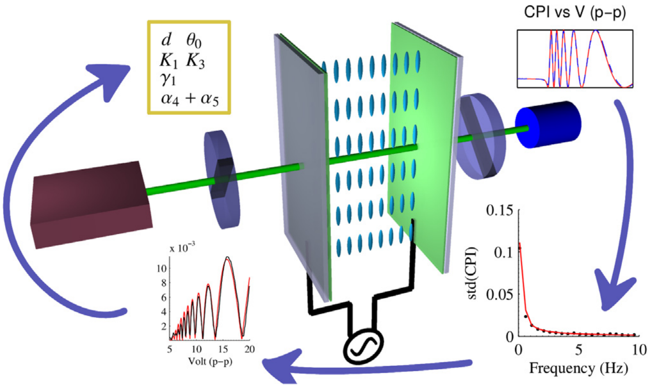
Lifting degeneracy in nematic liquid crystal viscosities with a single optical measurement

T.P. Bennett [a], M.B. Proctor [b,1], M. Kaczmarek [b], G. D'Alessandro [a,*]

[a] Mathematical Sciences, University of Southampton, Southampton, UK
[b] Physics and Astronomy, University of Southampton, Southampton, UK

GRAPHICAL ABSTRACT



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ABSTRACT

The viscosity of complex, anisotropic fluids, such as liquid crystals or their colloidal suspensions, is characterized by a number of coefficients. Methods to measure them are, typically, sensitive only to their particular combinations, hence unable to determine them individually. Using an Ericksen-Leslie model and propagation of light through aligned layers of such materials, we show theoretically and verify experimentally how this degeneracy can be lifted by exploiting both the amplitude and frequency of the voltage applied to the cell as control parameters.

1. Introduction

Viscosity and visco–elastic coefficients play an important role in the dynamics of soft matter either in the form of pure complex fluids or of suspensions of micron or nanometer size objects. For example, when nanoscale objects, such as carbon nanotubes [1] or inorganic nanoparticles [2] were dispersed in liquid crystals, the host viscosity as well as elastic constants were modified. In another study, Faucheux et al. [3] used an asymmetric potential in optical tweezers to realize a thermal ratchet whose time scales depend on the particle diffusion coefficient. Hough and Ou-Yang [4] also experimented with optical tweezers to trap two particles

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* Corresponding author.
E-mail address: dales@soton.ac.uk (G. D'Alessandro).

[1] Current address: NKT Photonics, Unit 20 Compass Point, Ensign Way, Southampton SO31 4RA, UK.

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in a viscous fluid and study the fluid–induced correlations between their periodic motion. Viscosity is also very important in biological suspensions: for example, Chengala and co–workers explored the role of viscous shear in the flow of alga in microfluidic channels [5].

In many of these examples light was used to analyze the system response to induced motion. Here we adopt a similar principle to study the viscosity of an optically active system. We set the system in motion using a periodic force and measure its response by detecting the effect on the intensity of a light beam propagating through it. The system we focus on as a demonstration of principle is a nematic liquid crystal (LC) cell forced by an oscillating AC electric field. However, this technique could, in principle, be applied to other systems. For example, it could be particularly useful for characterizing colloidal suspensions of inorganic, plasmonic, ferroelectric or ferromagnetic nanoparticles.

The interest in LCs stems from their ubiquity in optoelectronic applications, where their birefringence and reorientation properties make them materials of choice for electrically tunable optical devices. Moreover, the speed at which LC molecules respond to a field is a critical property when considering device designs or new LC materials and it is, therefore, important to develop simple and quick methods to measure their viscosity and, hence, their response time.

In fact, the time response of an isotropic liquid to an external stimulus is inversely proportional to its viscosity. LCs are anisotropic liquids and their flow behavior is described by more than one viscosity coefficient, as is the case for the Erickson–Leslie–Parodi [6–8] theory for the director flow and alignment, which uses the five Leslie viscosity coefficients, α_{1-5}, though not all of them are equally critical in determining the switching dynamics of an LC device. For example, to first approximation the switch-off time of a planar LC cell of thickness d is given by [7]

$$\tau_{\text{off}} = \frac{d^2\gamma_1}{K_1\pi^2},$$

where K_1 is the splay elastic constant of the LC and γ_1 is its rotational viscosity, which can be written in terms of the Leslie viscosities as $\gamma_1 = \alpha_3 - \alpha_2$. Hence, determining the response time relies on accurately measuring several Leslie coefficients and, in particular, γ_1.

As the complexity of new LCs, as well as different electrode geometries, increases to achieve faster and stronger reorientation, the contribution and the values of different Leslie viscosities can play an important role. A typical example is the ‘‘kick-back effect” in a splay cell [6]: when a large field is applied, the nematic in the center of the cell aligns. However, at the edges of the cell, a thin layer exists in which the director is rotated through $\pi/2$ radians with respect to the alignment at the center in order to obey the boundary conditions. When the external field is switched off, the director begins to relax at the sides of the cell, where the distortion is large. This reorientation induces shear fluid flow, transferring angular momentum towards the center of the cell. This results in the counter-intuitive situation where the distortion at the center of the cell initially increases before relaxing. This kick-back effect, and the resulting reorientation angle, can be particularly pronounced if a large electric or magnetic field is applied and then turned off. This angle can be analytically related to the Leslie viscosity coefficients $\alpha_{2,4,5}$ [6,9].

There are various approaches to determine some combination of the Leslie viscosity coefficients, including mechanical methods [10,11] such as measuring the damping of a submerged oscillating plate under different director fields. Optical methods, used either in splay geometry [12] or in twisted cells [13], rely on measuring the transient optical response of a nematic under the application of a step-wise voltage. There are a number of advantages and drawbacks for each technique: direct mechanical methods tend to give the most accurate measurements of viscosity coefficients, but rely on bespoke equipment; measuring the transient optical response requires a smaller volume of LC [11] with respect to mechanical methods, but places undue weight on the initial director configuration. Scattering experiments [11,14], provide accurate measurements of γ_1, but requires an uncommon optical set-up. In another approach, the oscillatory response of a LC subjected to a static magnetic field and a sinusoidal electric field has been studied using NMR measurements [15]. Interestingly, in this work the oscillations of the director could be modeled neglecting fluid motion.

In this paper we describe a new, efficient and accurate optical method to determine the LC dynamical properties using light as a measuring tool and the frequency and amplitude of an applied AC field as the control parameters. It is known in the literature that optical measurements cannot easily separate the effects of different viscosity parameters and, hence, measure them. Kelly et al. [13] use cross-polarized intensity (CPI) measurements of switch-on and switch-off dynamics in a twisted cell to measure γ_1 and $\alpha_4 + \alpha_5$. They use low and large amplitude switching to measure γ_1 and $\alpha_4 + \alpha_5$, respectively. This paper builds on earlier work by Cossalter et al. [16,17] which suggested that all viscosities could be measured using switch-on and switch-off dynamics. Kelly et al. doubt this statement, but do not labor this point further. Grinfeld et al. [9] show very elegantly that a two-mode linearised solution of the Ericksen-Leslie equations in a planar cell is able to describe quite accurately the kick-back dynamics. The authors indicate that it could be possible to use this solution to obtain physical parameters of the LC, but do not dwell further on this point except to note that the quantity that is normally measured is the CPI rather than the LC deflection and this may affect the fitting procedure. This is indeed a key point, as we illustrate in this paper.

In the next section we introduce an Ericksen-Leslie model of the director dynamics in a planar LC cell and its effect on the CPI of a light beam propagating through the cell. In Section 3 we explain how CPI measurements suffer from degeneracy in the viscosity coefficients and show how this can be lifted using the frequency and amplitude of the voltage applied to the cell as control parameters. This is, in a narrow sense, the frequency domain equivalent of the time-domain technique of Kelly et al. [13], but the detailed analysis in this section allows us to put it on firmer grounds, while the ease of control of the frequency of the driving voltage gives us great flexibility in choosing optimal measurement regimes. We use this new procedure to measure the viscosities of some standard LCs: we describe the experimental set up in Section 4 and the fitting procedure in Section 5. The paper is closed by a discussion of the advantages and disadvantages of the method and its possible extensions.

2. Flow in an AC-driven planar cell

Optical methods to determine elastic constants, pretilt, birefringence and cell thickness are based on CPI measurements (see Fig. 1 for a typical experimental setup). Normally, in these experiments the frequency of the applied voltage has a high value (1–10 kHz) so that the LC responds only to the rms voltage amplitude and the CPI is constant in time. We extend this method by allowing the frequency to have sufficiently low values so that the LC is able to follow the driving voltage: the director oscillates about an average deflection, determined by the amplitude of the applied field, and modulates the phase lag of the light and, hence, the CPI. Its standard deviation is a good measure of the modulation amplitude. The director oscillations are coupled to the fluid flow within the cell and, as a result, optical measurements are sensitive to multiple viscosity coefficients.