Measurement of viscosity of lyotropic liquid crystals by means of rotating laser-trapped microparticles

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It is impossible to use the conventional methods based on the bulk LCs with uniform orientation to perform the rheological study on micro- and nano-scales and for small amounts of the material.

Laser tweezers and optically trapped rotating microspheres have been previously used to probe complex fluids (including thermotropic LCs) at mesoscales [9–13]. For example, the rotating-particle microrheological method based on laser tweezers has been used for measuring the viscosity of fluids such as water and cellular sap [9]. In this work, we extend this rotating-particle method of Friese et al. [14] and Bishop et al. [9] to the study of LC fluids and measure effective viscosity coefficients of these complex soft matter systems. Instead of applying the laminar flow of LCs, we rotate an optically anisotropic microsphere by means of a trapping laser beam to exert shear stress on the surroundings. Circularly polarized light carrying the spin angular momentum (SAM) allows us to apply a torque and rotate a microparticle by means of an exchange of optical angular momentum (OAM) between the beam and the object [14,15]. We utilize a rotating laser-trapped optically anisotropic microsphere to locally measure the effective viscosity of lyotropic LC fluids in cholesteric and lamellar phases. This method does not require perfectly aligned samples (which are difficult to achieve for lyotropic LCs), avoids artifacts su-3(r)-5a(2(e)4)-7(r)-5f7 0.325 12(u)(m)2ting f7(a)-3in2

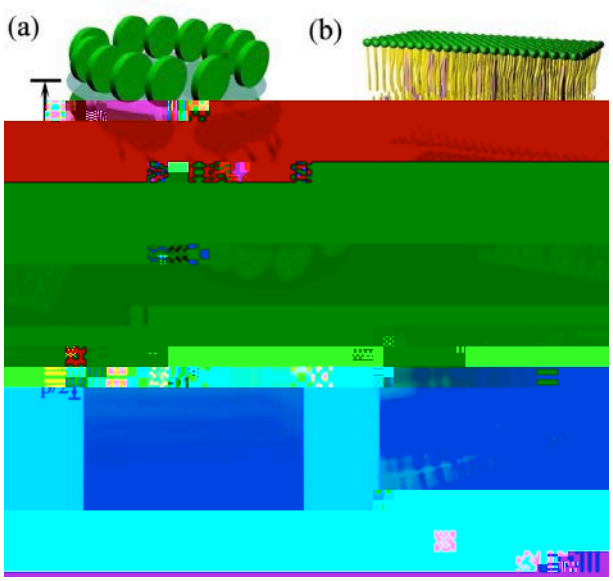

Fig. 1. Schemes of (a) cholesteric LC phase formed by self---

determined by the balance of viscous and optical torques and thus allows one to measure the viscous drag force and an effective viscosity coefficients for purely viscous fluid. The effective birefringence of the vaterite particles Δn_v is about 0.06 [15], which is about an order of magnitude larger than that of used lyotropic LCs.

3. Optical tweezers and imaging techniques

Optical trapping is performed using an inverted microscope with a 100× oil immersion objective lens with high numerical aperture (NA = 1.3) integrated with a charge coupled device (CCD) camera, Fig. 3(a). The power of laser beam at 1070nm from an Ytterbium fiber laser (IPG Photonics) is controlled by a half-wave plate (λ/2) and a polarizer and then the light is focused to a diffraction-limited spot in the sample by the objective. A quarter-wave plate (λ/4) located immediately before the objective converts the linearly polarized light into a circularly polarized beam that is then used to rotate birefringent objects, Fig. 3(b). After passing through the particle, the laser light is collected by a condenser and then split into two orthogonal linearly polarized beams by another quarter-wave plate with the fast axis orientated at 45° to the axes of a polarizing beam-splitter (PBS) cube [9,24]. The two linearly polariz10<005tT 0[mponents tT 0[rrespond to the left- and right-handed circularly polarized

lateral plane of the microscope. The three-dimensional (3D) **n**(**r**) around colloids is also determined with submicron lateral and vertical resolution by use of an anisotropic fluorescent dye and detecting the polarized fluorescence signal using FCPM. We have also utilized two-photon excitation fluorescence polarizing microscopy (2PEF-PM) for imaging of **n**(**r**) patterns and spatial location of the microparticles used as microrheology probes. This technique constitutes of the multimodal nonlinear polarizing microscopy described in details elsewhere [25,26]. The 2PEF-PM imaging is performed by use of excitation with a 960nm femtosecond pulse from a tunable (680-1080 nm) Ti:sapphire oscillator (140 fs, 80 MHz, Chameleon Ultra-II, Coherent). We use epi-detection mode and interference filters to separate the fluorescent signal from the excitation light [19]. This nonlinear optical process of two-photon absorption of the dye molecules attains the high intrinsic submicron resolution in both axial and radial direction as well as a stronger sensitivity to molecular orientations as compared to FCPM [25,26]. FCPM and 2PEF-PM imaging was performed by using a 100 × objective with NA = 1.4 or a 60× objective with NA = 1.42 and yield consisteosiz23(t)-9(hn)8(n)-12inuiphire oscic-9(h)tasi Fscii

desired location within the 3D volume of the sample and in this way assure that the measurement is done in a region with a desired uniform **n**(**r**) and far away from defects, Fig. 4. On the other hand, the influence of defects on mechanical response of the LC can be probed by deliberately doing the microparticle rotation experiment in the proximity of defects which are rather abundant in lyotropic LCs (Fig. 4). The possible artifacts due to the optically anisotropic nature of LCs can be mitigated by aligning them to have a specific well-defined orientation of **n**(**r**) with respect to the optical axis of the microscope, like in the case of homeotropic alignment of the studied LC in the lamellar phase that has **n**(**r**) parallel to the axis of trapping laser beam [Fig. 1(b) and Fig. 2(b)] or planar alignment of cholesteric lamellae with the helical axis along the cell normal [Figs. 1(a), (c), Fig. 2(a), and Fig. 4(c), (f)].

sample in the absence of the birefringent bead, its polarization state remains unperturbed, indicating that the possible optical artifacts due to the LC medium can be neglected. When this light propagates through the trapped bead, the optical axis of the bead aligns parallel to the beam's lateral plane due to the a

and the bead is rotating around an axis normal to the lamellae, the geometry of our experiment assures that the velocity and velocity gradient directions are parallel to the layers (since the shear flow created by the rotating vaterite is parallel to the layers). We have also assured that the lamellar LC with homeotropic **n**(**r**) along the trapping beam's axis has no measurable effect on circular polarization of the used trapping light. The vaterite particle causes weaker

impinge on the rheological studies of LCs made of nanoparticles and newly-synthesized organic molecules, which are often available only in tiny εl-level amounts. Our method also allows one to avoid the effects of presence or flow-induced generation of defects on viscosity measurements. At the same time, it can be also used to probe how defects modify mechanical response of various LC fluids. From the standpoint of view of the rheological study of lyotropic LCs, the main advantage of our method as compared to the passive particle microrheology techniques is that the spatial position of particles within the sample as well as velocity and velocity gradient directions with respect to the LC director can be controlled, so that the influence of low-quality alignment (inherent to lyotropic LCs) or effects of particle localization into defects can be mitigated.

Acknowledgments

This work was supported by the International Institute for Complex Adaptive Matter (ICAM-I2CAM), Renewable and Sustainable Energy Initiative and Innovation Initiative Seed Grant Programs of University of Colorado, the NSF grants DMR 0645461, DMR-0820579 and DMR-0847782, the National Basic Research program of China (2004CB719800) and the Australian Research Council. We thank Paul Ackerman, Noel Clark, Angel Martinez, Julian Evans, and Rahul Trivedi for discussions. We also thank Bohdan Senyuk for his help with the 2PEF-PM imaging experiments.

#155473 - $15.00 USD Received 27 Sep 2011; accepted 11 Nov 2011; published 23 Nov 2011