Direct observation of columnar liquid crystal droplets

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Supplementary Information

1 Experimental Methods

1.1 Particle synthesis and characterization

Gibbsite platelets were synthesized through hydrothermal treatment of aluminum alkoxides in acidic environment, following the procedure developed in the Van 't Hoff laboratory^{1,2}. An acidic aqueous solution of hydrochloric acid (HCl 0.09 M, 37%, Merck), aluminum sec-butoxide (0.08 M, 95%, Fluka Chemika) and aluminum iso-propoxide (0.08 M, 98+%, Acros Organics) was mechanically stirred for 10 days. Subsequently, the mixture was heated in a water bath at 85 °C for 72 h. Then, the colloidal dispersion was centrifuged at 1000g (overnight, 15-20 h) in order to remove the smallest particles and decrease the polydispersity. Finally, the dispersion was dialyzed against demineralized water (Visking regenerated cellulose tubes, MWCO 12 000-14 000).

The resulting platelets were analyzed with transmission electron microscopy (TEM, Tecnai 10, FEI Company) and atomic force microscopy (AFM, Digital Instruments, used in tapping mode with standard TESP silicon tip). 1(a) shows an AFM image of the hexagonal gibbsite platelets. This analysis showed that the platelets have an average corner-to-corner distance of 207 nm ($\pm 35\%$) and thickness of 8.2 nm ($\pm 46\%$), both with a unimodal distribution. However, the platelet aspect ratio (diameter/thickness) has a bimodal distribution, with a narrow peak of small aspect ratio (thick) platelets and a broader peak of large aspect ratio (thin) platelets (see 1(b)).

1.2 Sample preparation

The colloidal stability of the gibbsite platelets was enhanced by surface treatment with aluminum chloro hydrate (ACH). At a pH around 4, ACH forms aluminum polycations that adsorb on the particle surface, giving rise to a steep repulsive particle interaction³. Subsequently, the gibbsite dispersions were adjusted to an ionic strength of 10^{-2} M NaCl and concentrated via centrifugation to reach the desired particle volume fraction, which was 30 and 36 v/v%. Samples were prepared in 0.2 mm and 0.05 mm × 4 mm × 40 mm VitroCom flat glass capillaries, that were filled by capillary action. The highly viscous suspensions at 36 v/v% were centrifuged for a few minutes after filling of the capillary in order to remove air bubbles and drive the suspension towards the capillary bottom.

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1.3 Naked-eye observation and polarized light microscopy

The capillaries were photographed with a Nikon Coolpix 995 digital camera. The samples were studied in more detail with a Nikon LV100 Pol polarized light microscope equipped with $10 \times$ and $20 \times$ Nikon CFI Plan Fluor objectives and a 5 megapixel CCD camera (MP5, QImaging). Bragg reflections were observed with the polarized light microscope while the sample was illuminated with a white light source (model 190, Dolan Jenner).



Fig. 1 Colloidal gibbsite platelets. (a) Atomic force micrograph of the gibbsite platelets. (b) Histograms of the platelet diameter (corner-to-distance), thickness and aspect ratio (diameter / thickness), measured with AFM.

2 Columnar droplets in the nematic phase

Figure 2 shows columnar droplets in the columnar phase that follow exactly the director field of the nematic phase.



Fig. 2 Micrographs of columnar droplets forming in the nematic phase, with the droplets exactly following the nematic director field, including a defect with topological charge +1/2. (a) Observed between crossed polarizers; (b) with retardation plate added; (c) with only the polarizer. Polarizer and analyzer are indicated with arrows and the slow axis of the retardation plate with a dashed arrow.

3 Caption movie Verhoeff_LCdroplets.mov

Polarization microscopy of liquid crystal droplets moving through the isotropic middle phase of a gibbsite suspension that exhibits a three phase equilibrium. Nematic droplets (tactoids) originate in the columnar bottom phase (not visible here) and rise through the isotropic phase towards the nematic top phase, while columnar droplets originate in the nematic phase and sediment through the isotropic phase towards the columnar phase. Scale: the width of the capillaryis 4 mm, while in the second part the field of view is 1.5 mm wide. The recorded time is 4 hours. Observed with a Nikon LV100 Pol polarized light microscope equipped with a QImaging MicroPublisher 5 megapixel ccd camera. A full wave retardation plate ($\lambda = 530$ nm) was used.

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