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Liquid Crystals

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Liquid crystal bridges

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The liquid crystalline material octylcyanobiphenyl was studied in the form of bridges spanning the space between two solid supports in an immiscible water bath. In the nematic phase the bridge collapses above a certain length-to-diameter ratio, consistent with the behaviour of ordinary Newtonian liquid bridges. The smectic A phase, however, exhibited the formation of very long, stable columns as a consequence of its non-Newtonian behaviour.

1. Introduction

A liquid bridge is composed of a liquid which spans the gap between two solid supports. The bridge may be in vacuum or air, or in an immiscible fluid. Although liquid bridges can be constrained by a variety of different boundary conditions, perhaps the most commonly studied bridge is bounded at the two ends by a pair of equivalent, colinear, right circular cylindrical rods. In this way a bridge in a gravity-free or gravity-compensated environment may adopt a cylindrical shape, with diameter d equal to that of the support rods and length L equal to the separation between the ends of the rods. This condition, of course, applies only to bridges whose volume $V = \pi d^2 L/4$; if material were withdrawn from the bridge, its shape would be pinched in the centre, as the length is constrained by the rod separation and the diameter is constrained by the necessity to wet the ends of the support rods. During the nineteenth century both Rayleigh and Plateau showed theoretically that a cylindrical liquid column in a gravity-free environment is stable against radial shape fluctuations as long as its length to diameter ratio R—this is sometimes known as the 'slenderness ratio'—is less than π [1–3]. For $R \geqslant \pi$ the radial shape fluctuations are unstable, and the bridge catastrophically pinches off and breaks into two pieces. In the intervening years theorists have examined the equilibrium shapes and stability of liquid bridges in the presence of gravity. Under gravity, of course, a vertically-oriented bridge would tend to sag, although it would not collapse for a sufficiently small slenderness ratio R. (For the purposes of this paper, R is defined as the ratio of the bridge length to the bridge diameter *at the two bounding support rods*, i.e. the diameter of the rods themselves.) If the gravitational force could be continuously varied, the originally cylindrical bridge would tend to deform, although both R and the volume of the bridge would remain unchanged. For sufficiently large R the bridge would collapse, although this would occur at $R < \pi$ if gravity were present [4–8]. In order to model this behaviour, it is necessary to introduce the dimensionless Bond number B, viz.

$$B \equiv \frac{g \Delta \rho d^2}{4\sigma}. \tag{1}$$

Here g is the gravitational acceleration, $\Delta\rho$ is the density difference between the bridge and the surrounding medium, d is the diameter of the two end supports, and σ is the surface tension of the bridge. The parameter B relates the relative importance of gravitational energy to the surface tension. The case $B = 0$ corresponds to a completely gravity-free or gravity-compensated environment, such as would obtain in outer space, in a 'drop tube' on Earth, or in a perfectly density-matched liquid bath. Variation of effective gravity may be accomplished utilizing the liquid bath by controlling the Bond number through $\Delta\rho$. For many years this two-immiscible-fluid approach has been the experiment of choice, and is performed in a 'Plateau tank' [6–11]. Often, however, it is necessary to study the fluid bridge in air or vacuum, which requires a space-borne environment [12] or a very tall evacuated drop tower [13] that provides several seconds of free-fall. More recently we demonstrated that a paramagnetic liquid (water doped with the paramagnetic salt $MnCl_2.4H_2O$) could be levitated against gravity in a strong magnetic field gradient [14]. For that experiment the effective Bond number was redefined as $B \equiv (g\Delta\rho - \chi H \nabla H) d^2/4\sigma$, where $\Delta\rho$ corresponds to

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the density of the bridge (since $\rho_{air} \approx 0$), χ is the magnetic susceptibility, and H is the magnetic field. The Bond number, and therefore the effective gravitational field, was controlled by varying H.

In recent years workers also have attempted to push the slenderness ratio R in a gravity-free environment beyond the theoretical Rayleigh limit of π. This typically has been accomplished by driving the bridge with ultrasound or by application of an axial electric field with an appropriate dielectric mismatch between the bridge and the surrounding medium [9, 10]. Other experiments have been performed on non-Newtonian polymers to extract viscosity versus shear rate information. In these experiments, as the cylindrical polymer bridge is stretched lengthwise, the diameter of the cylinder must be concomitantly reduced so as to keep the volume constant. In such experiments the cylinder is only metastable, and slenderness ratios $R > \pi$ have been achieved [15–17]. These experiments, however, do not examine the *stability* of non-Newtonian liquid bridges.

2. Experimental

In this paper we report on experiments involving bridges consisting of *liquid crystal* (LC) in the nematic and smectic A phases. Because of the liquid crystalline order, particularly the orientational order of the nematic and smectic A phases and the translational order of the smectic A phase, one might expect the stability and shape of LC bridges to be somewhat different from their isotropic counterparts. In our experiments we have made several observations:

(1) The stability of a cylindrical nematic bridge is *similar* to that of an ordinary isotropic Newtonian fluid, i.e. the maximum slenderness ratio $R \approx \pi$ for Bond number $B = 0$, and less than π for $|B| > 0$.
(2) Unlike isotropic fluids, cylinders in the smectic A phase may exhibit slenderness ratios considerably larger than π for $B = 0$.
(3) For $|B| > 0$ cylinders in the smectic A phase remain cylindrical (even those with slenderness ratios in excess of π), with no apparent sagging, until the Bond number exceeds some value B_{deform}. At this point the cylinder begins to sag, but does not break. As the Bond number is further increased by changing the density of the surrounding bath, the bridge eventually collapses when the Bond number increases to a value $B_{collapse}$.

2.1. *The nematic phase*

A cylindrical aluminum rod was mounted, facing upward, in a glass ‘Plateau’ tank. The end of the rod was machined as shown in figure 1 in order to prevent the liquid crystal from running along the side of the rod. The diameter of the rod at the tip was $d = 0.32$ cm. A nearly identical rod was mounted on a precision micrometer mount and arranged colinearly and facing downward toward the first rod. Liquid crystal was injected through a small axial hole in the upper rod using a 25-gauge hypodermic needle and butterfly syringe. As the viscosity and other physical parameters of the liquid crystal vary with temperature, the entire assembly, including the tank, was inserted into an aluminum jacket that was electronically temperature controlled to 0.2°C.

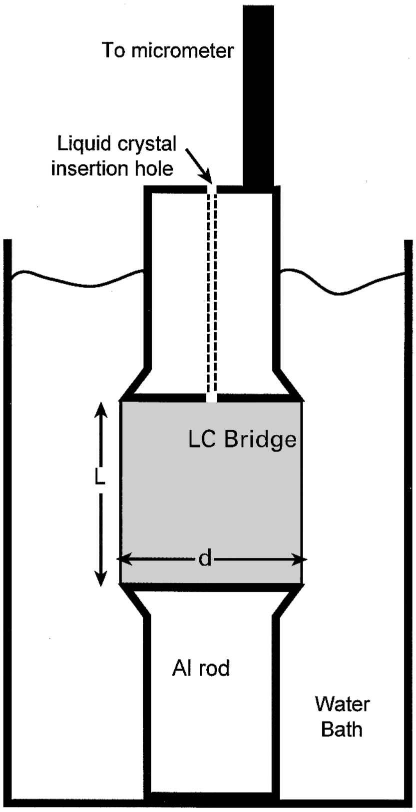

Figure 1. Schematic diagram of experimental set-up. The liquid bridge is formed between two aluminum supports of diameter $d = 0.32$ cm at the tips. The upper support has a hole for injection of liquid crystal, and is attached to a micrometer to facilitate controlled variation of R. The support assembly is in a Plateau tank filled with water.

In the absence of water, the two rods were first brought close together with a small (~0.1 cm) gap between the tips. A small amount of the liquid crystal octylcyanobiphenyl (8CB) at room temperature (in the smectic A phase) was injected into the gap between the tips, so that the liquid crystal completely wetted the ends of the two rods. As the density of the smectic A phase is $\rho_{SmA} = 0.996\,\mathrm{g\,cm^{-3}}$ [18], we chose H_2O, whose density is similar, as the density matching fluid. ($\rho_{H_2O} = 0.998\,\mathrm{g\,cm^{-3}}$ at 23°C and $0.994\,\mathrm{g\,cm^{-3}}$ at 36.5°C, the limits of the experiment.) Moreover, D_2O, whose density ρ_{D_2O} is approximately 11.1% larger than that of H_2O, could be titrated into the bath, thereby facilitating a variation of $\Delta\rho$ and thus of Bond number. After the ends of the rods had been wetted with liquid crystal, the tank was filled with H_2O. The length of the column was controlled by the micrometer, which allowed us to translate the top rod upward. As we did so, more liquid crystal was injected into the gap in order to maintain a right circular cylindrical bridge, i.e. a volume $V=\pi d^2 L/4$. Upon reaching a desired slenderness ratio R (less than π), the entire assembly was heated to 36.5°C, above the nematic–smectic A transition temperature $T_{NA}=33.5$°C. As the density of the nematic phase $\rho_N=0.985\ \mathrm{g\,cm^{-3}}$ [18] is less than the water bath ($\Delta\rho\sim 0.009\ \mathrm{g\,cm^{-3}}$), the bridge began to deform (figure 2), but did not collapse. D_2O of density $\rho_{D_2O}=1.102$ at $T=36.5$°C was then titrated into the tank in order to further increase $\Delta\rho$. For sufficiently large $\Delta\rho$ the bridge finally collapsed. Note that during this process the volume of the bridge remained constant, even as it became deformed by the addition of D_2O to the bath.

Coriell, *et al.* calculated the maximum value of the initial slenderness ratio R for an isotropic liquid that corresponds to the limit of stability for a given Bond number B [4]. Throughout the region $B \leqslant 0.1$, an excellent approximation to their result is the empirical form

$$R=a+b_1\exp\left(\frac{-B}{c_1}\right)+b_2\exp\left(\frac{-B}{c_2}\right) \qquad (2)$$

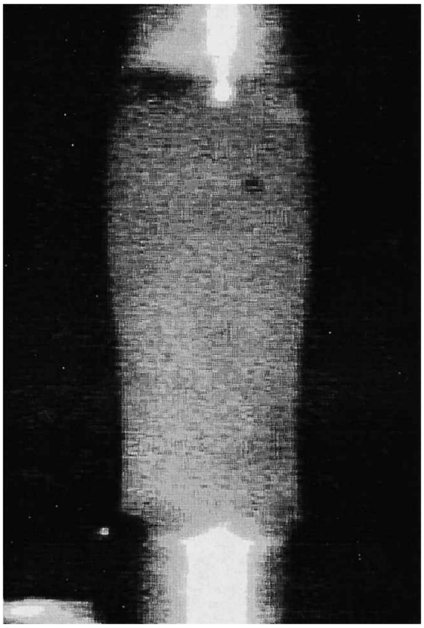

Figure 2. Deformed, but stable, nematic bridge. The density of the bath is higher than the density of the nematic, resulting in an upward bulge of the liquid crystal.

where the five dimensionless parameters are: $a=2.21$, $b_1=0.81$, $b_2=0.12$, $c_1=0.071$, and $c_2=0.0038$. Additionally, Meseguer found an analytic asymptotic form, viz. $R=\pi[1-3/2(3/2B^2)^{1/3}]$ [19], although this is valid only in the limit of $B\to 0$ and breaks down in the larger Bond number region of our experiment.

Assuming that the nematic phase behaves as an ordinary Newtonian fluid, we substituted equation (1) into equation (2). This gives R at the stability limit in terms of $\Delta\rho$ (which is the experimentally-controlled variable) and σ at the nematic–water interface (which is the unknown parameter). The stability of the nematic bridge was examined for a number of different slenderness ratios, all of which gave consistent results. From the data we extracted a surface tension $\sigma=(16\pm 1)\ \mathrm{erg\,cm^{-2}}$. Figure 3 shows R versus the density mismatch $\Delta\rho$ at which the bridge collapses. The curve, cf. equation (2), is a best fit to the data with the fitting parameter $\sigma=16\ \mathrm{erg\,cm^{-2}}$. The behaviour of the nematic bridge is clearly similar to that of an isotropic Newtonian liquid bridge [4]. This is not really surprising, as the only significant differences between the two are the orientational order of the nematic phase and the associated elastic energy associated with director distortions. In order to estimate the role of orientational elasticity, we imagine the elastic energy associated with a pinching of the bridge; this would approximately correspond to the energy of a hemispherical cap of radius $r=d/2$. Neglecting the energy associated with disclinations, we may estimate the elastic energy of a nematic cap as $F_{\mathrm{elastic}}\sim 1/2Kr^{-2}\times 2/3\pi r^3$, where K is a typical elastic constant of order 10^{-6} dyn [20]. Taking r as the radius of the end of the bridge ($r=0.16$ cm), we find $F_{\mathrm{elastic}}\sim 2\times 10^{-7}$ ergs. On the other hand, the energy associated with the surface tension for the hemispherical cap is $F_{\mathrm{surface}}\sim 2\pi r^2\sigma$, or approximately 3 ergs. Owing to the large radius of curvature, the surface term is clearly orders of magnitude larger than the elastic term. In consequence we would expect the elasticity of the nematic phase to play only a negligible role in the behaviour of the bridge, and the nematic bridge would respond in a manner similar to that of an isotropic liquid bridge.

2.2. *The smectic A phase*

Let us now turn to bridges in the smectic A phase. The smectic A phase is known to exhibit significant 'shear thinning', wherein the viscosity decreases markedly with increasing strain rate. The viscosity versus shear strain rate was measured for 8CB more than a dozen years ago, and was found to exhibit extreme thinning behaviour [21]. In addition to the non-Newtonian shear thinning, bulk smectic A samples tend to exhibit viscoelastic behaviour with a shear yield stress Y. For shear

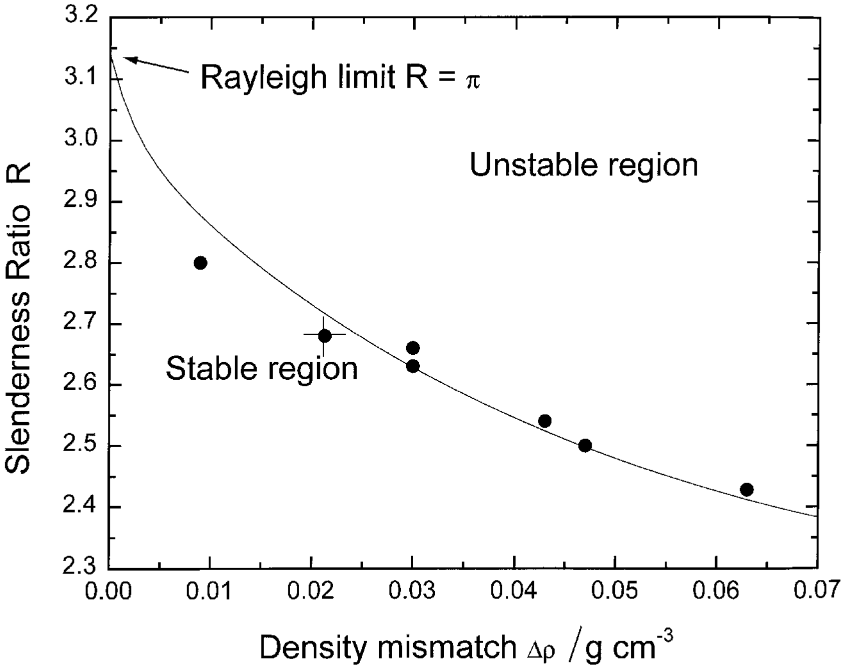

Figure 3. Slenderness ratio R versus density mismatch $\Delta\rho$ at the limit of stability; the maximum value on the x-axis, $\Delta\rho = 0.07\ \mathrm{g\,cm^{-3}}$, corresponds to a Bond number $B = 0.108$. The bridge is stable below the curve; the solid line is a fit to the data with $\sigma = 16 \pm 1\ \mathrm{erg\,cm^{-2}}$.

stress below Y only elastic behaviour is observed. Above Y one observes an onset of plastic flow, and the material is referred to as a Bingham fluid. In smectic A liquid crystals this behaviour arises from the domain structure of an unaligned sample, i.e. randomly oriented regions of characteristic length ~ a few μm, with each region having parallel smectic layers. To examine the effects of this highly non-Newtonian behaviour on bridge stability, we first formed a cylindrical smectic A column of slenderness ratio $R = 3.88$ (figure 4) at room temperature, approximately 23°C. This is well in excess of the Rayleigh limit of π for Newtonian liquids [1, 2], and it was observed to remain unchanged in shape for well over 50 h. A column of $R = 4.2$ was similarly stabilized. It is clear that the smectic A phase behaves differently from the nematic phase. We next examined the stability of the cylindrical smectic A columns on changing the density of the bath. A cylindrical column of slenderness ratio $R = 3.88$ was initially drawn in a density-matched H_2O bath. D_2O was then titrated into the water bath in order to increase the (temperature-dependent) density mismatch $\Delta\rho$ up to $\Delta\rho = 0.037\ \mathrm{g\,cm^{-3}}$. The temperature was then slowly increased. At lower temperatures there was no observable change in the column, indicating the absence of flow. However, when the temperature reached $T_{NA} - 1$°C (approximately 32.5°C), the smectic A cylinder began to deform (but did not collapse) with the density mismatch $\Delta\rho_{\mathrm{deform}}$ at this temperature equal to $0.034\ \mathrm{g\,cm^{-3}}$. If we make the excellent approximation that the surface tension of the smectic A–water interface is the same as nematic–water interface [22, 23], $\Delta\rho_{\mathrm{deform}}$

Figure 4. Cylindrical smectic A bridge with $R = 3.88$ in pure H_2O bath.

would correspond to a Bond number $B_{\text{deform}} = 0.053$. However, we note that because of the smectic's apparent yield stress, the surface tension does not play the same crucial role as it does for the isotropic and nematic liquids. Thus, the Bond number at the onset of deformation is given only for completeness, and does not play an important role in the physics. Because of the extremely high viscosity at small flow rates, the smectic A bridge took several hours before its sagging shape equilibrated. More D_2O was then added to the bath and the shape of the bridge deformed further, but did not collapse. Finally, at $\Delta\rho_{\text{collapse}} = 0.065\ \text{g cm}^{-3}$, the bridge pinched off and collapsed into pieces at each of the two rods.

To understand the behaviour of a bulk smectic A under stress, we examined the creep flow for 8CB using a cone and plate rheometer [24]. A constant stress in the range of 1 to 50 dyn cm^{-2} was applied for 2 min, and then another 2 min was permitted for elastic recovery. (Nearly identical results were obtained for much shorter duration measurements of 10 s.) This was done for several different stress values, at each of the three temperatures 25.5, 28.5, and 31.5°C. Note that the nematic–smectic A transition temperature is 33.5°C. Figure 5 shows the strain versus shear stress at the end of the shear cycle. At low stress the strain was very small and completely elastic. Above some critical yield stress Y we observed the onset of inelastic flow, where it is clear from figure 5 that Y is a strong function of temperature T. At the nematic–smectic A phase transition temperature the yield stress vanished. The yield stress versus temperature in degrees Celsius is shown in the inset of figure 5.

The maximum value $\tau_{\max}$ of the shear stress in a vertical cylinder of isotropic material that is held from above and below may be estimated by applying elasticity theory [25]. One finds that $\tau_{\max}$ is independent of the height of the cylinder and obeys a relation of the form $\tau_{\max} \sim rg\Delta\rho/6$. Calculating the surface shear stress for the value $\Delta\rho_{\text{deform}} = 0.034\ \text{g cm}^{-3}$ at which the cylinder began to deform at 32.5°C, we find $\tau_{\max} \sim 1\ \text{dyn cm}^{-2}$. This is smaller than, but of the same magnitude as the measured yield stress $Y = (4 \pm 2)\ \text{dyn cm}^{-2}$ at this temperature (cf. figure 5). The discrepancy may be due to temperature gradients in the Plateau tank (with some regions of the tank closer to the nematic–smectic A phase transition temperature), as well as to built-in stresses on formation of the cylinder. Additionally, we note that in the region $\Delta\rho_{\text{deform}} < \Delta\rho < \Delta\rho_{\text{collapse}}$, the smectic liquid crystal flows for a time, then stops. This strongly suggests that the liquid crystal deformation reduces the stress sufficiently to a value comparable to Y. This results in a significant slowing and ultimate cessation of flow.

Finally, it is important to comment on the length scales in the experiment. The behaviour of the smectic A phase often differs from that of an isotropic liquid. However, as the smectic domains are empirically of length scales of a few microns, the smectic would behave as a granular isotropic solid as long as the bridge has dimensions much larger than the domain size. This certainly is the case in our experiment, with one exception: when the bridge is just beyond the stability point, such that the (already pinched) collapsing bridge is just about to break into two smectic droplets. This

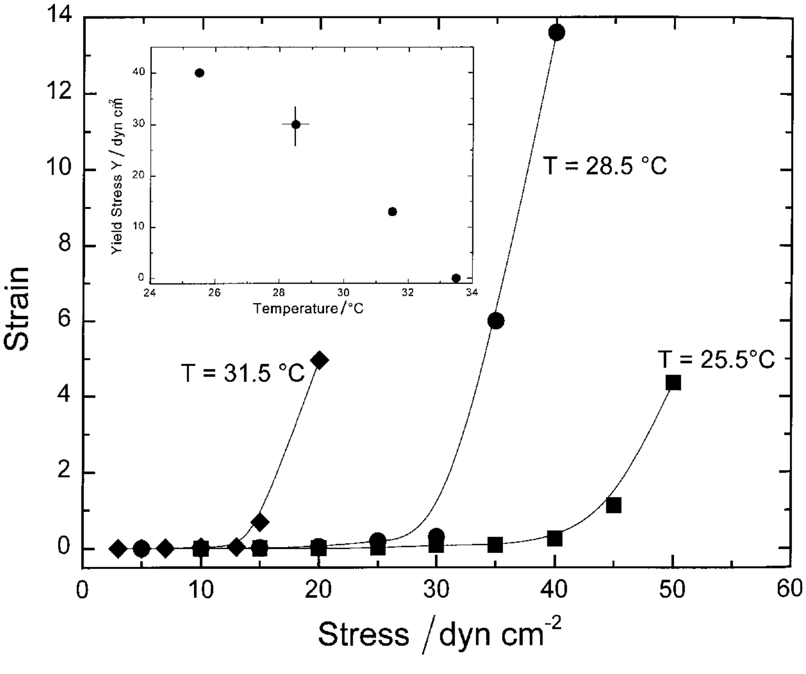

Figure 5. Strain versus shear stress at three different temperatures. The yield stress Y is the point at which flow begins, and corresponds to the sharp increase in slope. Inset corresponds to Y versus T.

problem in dynamics is beyond the scope of this work, however.

3. Conclusion

Overall, our results indicate that the orientational order of the nematic liquid crystal is insufficient to have an appreciable effect on the stability of an initially cylindrical nematic bridge. For all practical purposes, the nematic bridge is similar in behaviour to an isotropic Newtonian liquid bridge. On the other hand, owing to the shear thinning of the smectic, and more importantly to its viscoelastic behaviour, the smectic A bridge can be stabilized at neutral buoyancy even for very large slenderness ratios $R > \pi$. Moreover, if there is a density mismatch between the smectic A and the bath, deformation does not occur until the mismatch induces a shear stress greater than the yield stress of the smectic.

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