Orientational capillary pressure on a nematic point defect

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The analysis of some experimental data for the coalescence of two point defects in a capillary tube filled with a lyotropic liquid crystal shows that before the very interaction starts, defects move under the effect of a constant force. We suggest that this effect could be due to a *capillary pressure* and show how it can be justified by only assuming that there is a slight misalignment from the homeotropic anchoring on the boundary. These predictions are in good agreement with the data and open the way to further experiments.
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I. INTRODUCTION

Nematic liquid crystals are optically uniaxial fluids. The orientation of the optic axis, which may vary from point to point, is usually denoted by a unit vector $\boldsymbol{n}$, also called the *director*. Points of discontinuity for $\boldsymbol{n}$ appear as *defects* under crossed polarizers; they are classified by a topological theory, which assigns each of them a relative integer, often called the *charge* (see, e.g., [1] for a specific reference and [2] for a review). Roughly, the absolute value of the topological charge says how many times the director field surrounding a defect wraps about the unit sphere, while the sign of the charge indicates the orientation of this wrapping.

Throughout this paper we employ the one-constant approximation to Frank's free-energy functional:

$$E[\boldsymbol{n}]:=\frac{1}{2}\int_{\mathcal{B}}K|\nabla\boldsymbol{n}|^2dv, \tag{1}$$

where $\mathcal{B}$ is a cylindrical tile. If the lateral boundary of the tube enforces *homeotropic anchoring*, the energy minimizers do not exhibit point defects: they are regular fields that escape along the axis of the tube in either direction. They are often referred to as *escaped* or *fluted* orientations and were discovered by Cladis and Kléman [3] and Meyer [4]. Since the two escaped fields are symmetric with respect to a change of orientation along the axis, they store the same energy per unit height.

Two differently escaped fields cannot be continuously connected: thus, when they both appear inside the tube the field must exhibit at least one point defect. Actually, arrays of point defects are frequently observed along the axis of capillary tubes: they alternate in charge and interact with each other. Often two of them attract, coalesce, and finally annihilate one another.

In this paper we report some experimental data concerning the approaching of a pair of defects with opposite topological charges along the axis of a capillary tube filled with lyotropic liquid crystal. The experiment is described in [5]: it was performed to explore the coalescence between two point defects, but it also showed another interesting phenomenon that is the object of this paper. The actual coalescence between two point defects will be modelled and interpreted in [6].

Here we focus attention on the fact that at the beginning of the phenomenon the distance between the defects decreases linearly in time. We will show that this is the sign that no mutual force is at work: defects do not interact with one other, but they move independently under the effect of a *capillary pressure*. To prove this conjecture, in Sec. III we propose a model that builds upon a slight loss of symmetry in the homeotropic anchoring, possibly related to the direction of filling. In Sec. IV, the outcome of this analysis is compared with the data mentioned above: this comparison yields good agreement and thus proves the soundness of our conjecture. Finally, in Sec. V, we propose some directions of further experimental research.

II. EXPERIMENT

During the past year, Hillig and Saupe performed several experiments with the aim of observing the coalescence of two point defects that slide along the axis of a capillary tube enforcing homeotropic anchoring: these experiments are described in [5]. Here we report some of the data for a binary micellar nematic, containing 49 wt % of the anionic surfactant cesium perfluorooctanoate (CsPFO) and 51 wt % water, which shows a lamellar phase up to 31.5 °C, a nematic phase with disklike micelles up to 40.1 °C, and which transforms into an isotropic micellar solution above 40.1 °C [7]. For

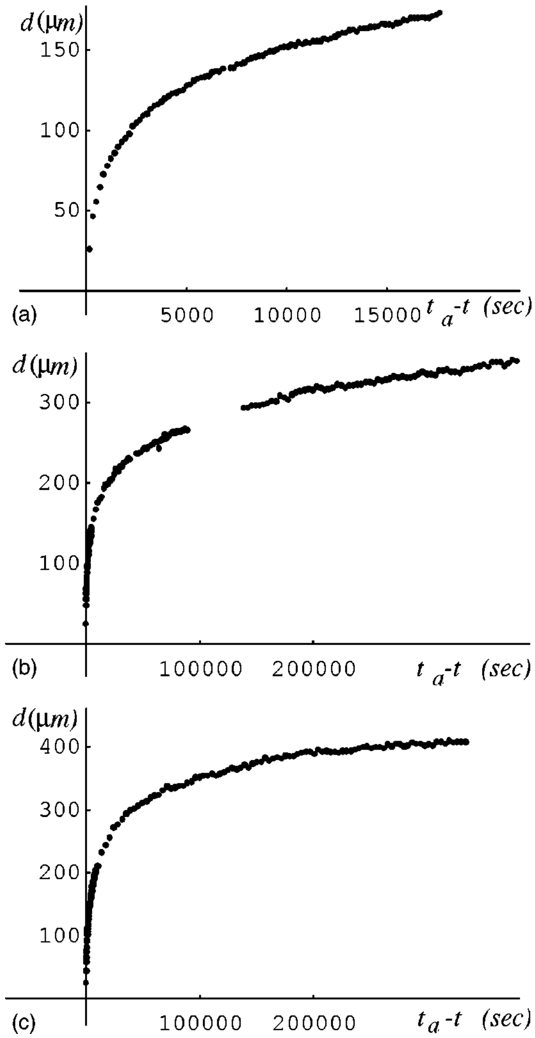

FIG. 1. Experimental data in a tube with diameter (a) 130 μm, (b) 300 μm, and (c) 400 μm, showing the distance d (μm) between two defects vs. the time to annihilation t_a-t (sec).

these measurements, small amounts of the mixture were sealed in cylindrical capillaries (Vitro Dynamics, Inc.: $D=400$, 300 μm; Fa. Hilgenberg: $D=130$ μm) to prevent water evaporation; the capillaries were used without further surface treatment and placed into a dish filled with immersion oil A of Nikon ($nd_{23\,°\mathrm{C}}=1.515$) in order to match the refractive indices. Sequences of micrographs were collected at certain time intervals controlled by a computer. In order to obtain well-defined textures, the samples were heated to the isotropic phase and then cooled to 39 °C. There was a two-phase range of 0.2 °C at the isotropic-nematic transition, in which nematic droplets developed which fused soon to a Schlieren texture. This texture relaxes within half a day into the escaped field described in Sec. I and defects arise so as to join two differently escaped fields. The time dependence of the distances between defects was determined within an experimental error of ± 2 μm.

For three tubes with diameter 130, 300, and 400 μm, Fig. 1 shows the distance d between the defects versus the *time to annihilation* t_a-t; here t_a represents the *annihilation time*. These data clearly show that at the beginning of the phenomenon the two defects feel a constant force: there is an evident linear decay of the distance in time. We show that it does not

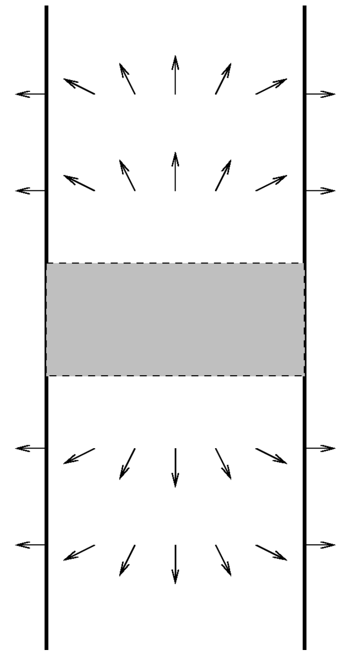

FIG. 2. The unspecified structure of a $+1$ defect serves to join two differently escaped fields.

result from the interaction of the two defects, which cannot produce a constant force: in this range the interaction forces are screened and defects move independently, each unaware of the presence of the other. Our purpose here is to understand why a single defect can move along the axis of the tube: as far as the authors know, this phenomenon has not been observed before; in the following sections we propose an explanation for it.

III. THEORETICAL CONJECTURE

In this section we turn attention to a single defect inside a cylindrical tube and we explore the possibility that it moves. In fact, we believe that the uniform motion showed by the data reported in the preceding section is just the superposition of the independent motions of the two defects.

Let $r=0$ be the equation of the axis of the tube in the frame of cylindrical coordinates $(\boldsymbol{e}_r, \boldsymbol{e}_\vartheta, \boldsymbol{e}_z)$. Consider a $+1$ defect lying on the axis at $z=0$. We now show that if the anchoring is homeotropic, then the defect cannot move. In fact, we can disregard the problem of modeling the director field around the point defect (which is studied, for instance, in [8] and [9]): for sufficiently high tubes, the structure of this field just serves to join two escaped fields with different directions of bend, precisely as shown in Fig. 2. We imagine that this structure roughly remains the same along all possible motions, so that the change in the elastic energy can only come from the growth or depletion of the regions with uniform fields. Since the two differently escaped fields store the same energy density, the elastic energy would not vary if the defect moves along with its structure. Hence, no spontaneous motion is possible within this setting, since the defect feels no force.

To account for the motion of a single defect, one has to assume that the above symmetry somehow breaks down. We were led to conjecture that the filling of the capillary determines a favored direction and produces a slight deviation from the homeotropic anchoring.

A very naive model is the following. Assume that the lateral boundary prescribes the following anchoring on the director field:

$$\boldsymbol{n}(R,\vartheta,z)=\cos\,\varphi_0\boldsymbol{e}_r+\sin\,\varphi_0\boldsymbol{e}_z, \tag{2}$$

where R is the radius of the capillary. It can be proved (see [10]) that there are exactly two stationary fields for the functional in Eq. (1) that obey this boundary condition. We call them $\boldsymbol{n}_-$ and $\boldsymbol{n}_+$: they are obtained by substituting for φ in

$$\boldsymbol{n}(r,\vartheta,z)=\cos\,\varphi(r,z)\boldsymbol{e}_r+\sin\,\varphi(r,z)\boldsymbol{e}_z, \tag{3}$$

either

$$\varphi_-(r,z)=\arcsin\left(\frac{R^2\cos^2\varphi_0-(1-\sin\,\varphi_0)^2r^2}{R^2\cos^2\varphi_0+(1-\sin\,\varphi_0)^2r^2}\right) \tag{4}$$

or

$$\varphi_+(r,z)=-\arcsin\left(\frac{R^2\cos^2\varphi_0-(1+\sin\,\varphi_0)^2r^2}{R^2\cos^2\varphi_0+(1+\sin\,\varphi_0)^2r^2}\right), \tag{5}$$

respectively. Figure 3 illustrates both $\boldsymbol{n}_-$ and $\boldsymbol{n}_+$ for positive φ_0.

It follows from Eq. (1) that the elastic free energy per unit height is

$$E_-=2\pi K(1-\sin\,\varphi_0) \tag{6}$$

for $\boldsymbol{n}_-$, and

$$E_+=2\pi K(1+\sin\,\varphi_0) \tag{7}$$

for $\boldsymbol{n}_+$. Imagine now a single defect on the axis of a sufficiently high tube: around it the director field has a certain unspecified structure that remains essentially unchanged in time, while far away it escapes like $\boldsymbol{n}_-$ and $\boldsymbol{n}_+$. Thus the force driving the defect can easily be computed as the difference between E_+ and E_-:

$$\boldsymbol{f}=4\pi K\,\sin\,\varphi_0\boldsymbol{e}_z, \tag{8}$$

which is independent of the size of the capillary.

To ascertain whether our conjecture on the loss of homeotropic anchoring is able to explain the data in the preceding section, we now need an appropriate dynamical model leading to the equation of motion for the system of two defects. Here, as in [6], we employ the dissipation principle recently proposed by Leslie [11] to rederive the classical theory of nematic flows. When the hydrodynamic flow is negligible, the rate of change in the elastic free energy is balanced by the dissipation $\mathcal{W}$ due to the viscous torques acting on the director field:

$$\dot{\mathcal{E}}+\mathcal{W}=0; \tag{9}$$

here no account is taken for the kinetic energy since the molecular inertia is regarded as negligible.

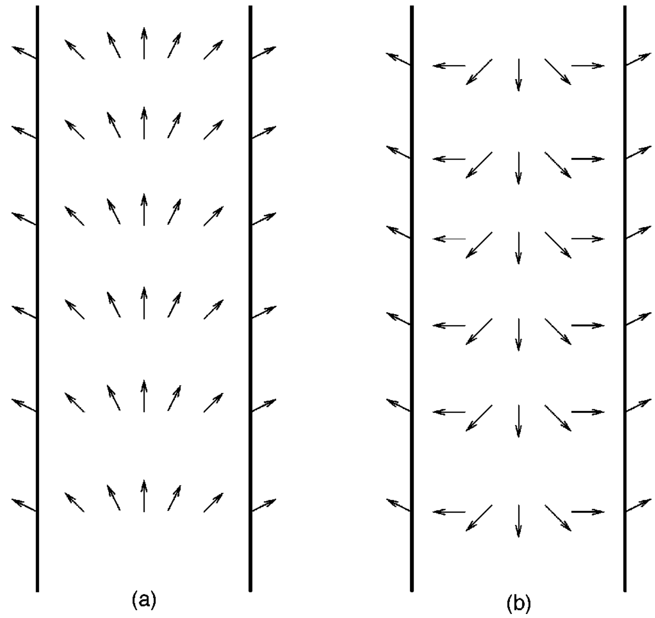

FIG. 3. (a) and (b) show $\boldsymbol{n}_+$ and $\boldsymbol{n}_-$, the two minimizers of the elastic free energy when the anchoring is not homeotropic.

The dissipation $\mathcal{W}$ has been computed in [10] by relying on a model for the field around the defect. For two-point defects at the distance d in a tube with radius R, $\mathcal{W}$ is given by

$$\mathcal{W}=\frac{\pi^{3/2}}{2}\gamma_1R\,\cos\,\varphi_0\{\sqrt{A(\varphi_0)}+\sqrt{A(-\varphi_0)}\}\dot{d}^2, \tag{10}$$

where $\gamma_1>0$ is the rotational viscosity (cf., e.g., Chap. 5 of [12]), φ_0 is the anchoring angle, and

$$A(\varphi_0):=\frac{1+\sin\,\varphi_0}{1-\sin\,\varphi_0}\{2\,\ln\,2-1-2\,\ln(1+\sin\,\varphi_0)+\sin\,\varphi_0\}. \tag{11}$$

Inserting Eq. (10) into Eq. (9) and using the fact that

$$\dot{\mathcal{E}}=4\pi K\,\sin\,\varphi_0\dot{d}, \tag{12}$$

we arrive at the following equation of motion:

$$\dot{d}=-\frac{2R}{\tau}\frac{4\,\tan\,\varphi_0}{\sqrt{\pi}(\sqrt{A(-\varphi_0)}+\sqrt{A(\varphi_0)})}, \tag{13}$$

where $\tau:=\gamma_1R^2/K$ is a characteristic time that depends on the capillary radius. Hence we obtain the time t as a function of d:

$$t(d)=\frac{\tau}{2R}\sqrt{\pi}\frac{\sqrt{A(-\varphi_0)}+\sqrt{A(\varphi_0)}}{4\,\tan\,\varphi_0}(d_i-d), \tag{14}$$

where d_i is the *initial distance* between the defects.

Equation (14) holds when the two defects are sufficiently far apart, that is, when their specific structures do not interact with one another. It is shown in [10] that the interaction between them vanishes when d exceeds the critical distance

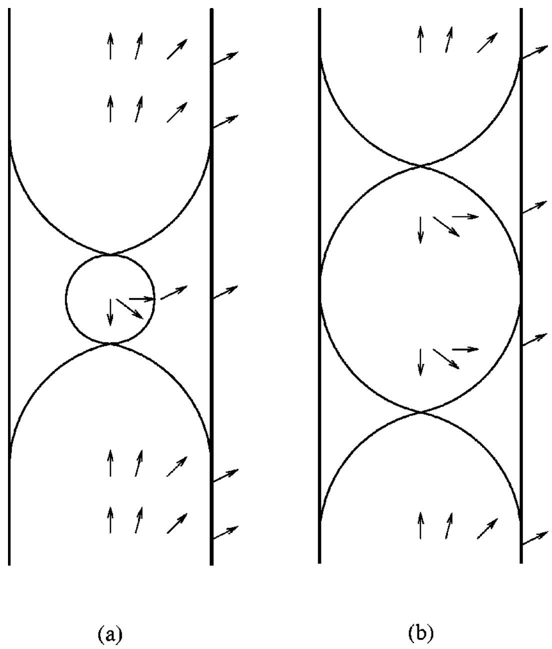

FIG. 4. Cross section of the optimal surface employed to construct a model director field between two defects with opposite topological charges: (a) $d<d_c$; (b) $d>d_c$.

$$d_c := 2\frac{\sqrt{A(-\varphi_0)\pi}R}{\cos\varphi_0}. \tag{15}$$

This conclusion rests on the construction of a model field devised in [10] to describe the director $\boldsymbol{n}$ in the region between two defects with opposite topological charges. In other words, this construction resorts to an axisymmetric closed surface connecting the defects, which joins a rescaled escaped field $\boldsymbol{n}_+$ and a radial tilted field that agrees with Eq. (2), respectively inside and outside the surface [see Fig. 4(a)]. The optimal shape of this surface is determined by minimizing the elastic free energy stored in the cylinder when the distance d between the defects is prescribed. For $d>d_c$ the optimal surface reaches the lateral boundary of the cylinder, and the total elastic free energy grows linearly in d. When the defects are moved apart, the director field changes all over the cylinder, but there is no energetic change, apart from the one described in this paper, because regions with the escaped fields $\boldsymbol{n}_+$ and $\boldsymbol{n}_-$ simply get intercharged [see Fig. 4(b)].

According to Eq. (14), when $d>d_c$ the time is a linear function of the distance, and the slope is a function of φ_0, R, and γ_1/K. Notice that any deviation from the homeotropic anchoring is crucial here: were φ_0 is equal to zero, no motion would be possible. On the other hand, when a deviation is present, the linear relation between the distance and the time also depends on the material constants K and γ_1, and on the radius of the tube. It is shown in the next section how these quantities must be chosen so as to fit the data.

IV. COMPARISON WITH THE DATA

Here we interpret the data of Sec. II by the law (14). Since the anchoring angle φ_0 is supposed to be very small, we

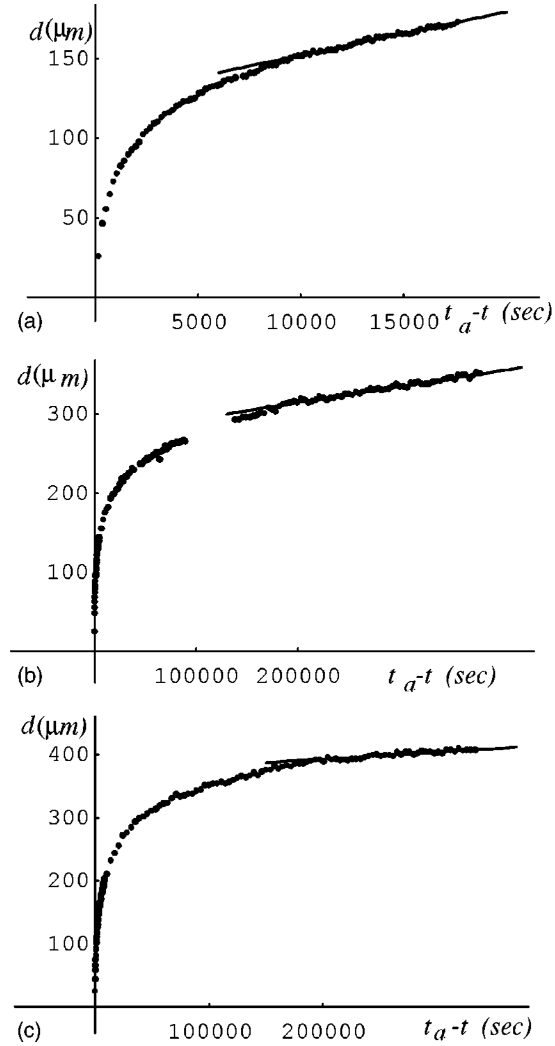

FIG. 5. Best fits of the early data in Fig. 1 with the theoretical law which gives the distance d (μm) between two defects vs. the time to annihilation $t_a - t$ (sec).

write Eq. (14) up to the first-order approximation in φ_0, thus obtaining

$$d=\frac{4K\varphi_0}{\sqrt{\alpha\pi}\gamma_1 R}t+d_i, \tag{16}$$

where we have set $\alpha := 2\ln 2 - 1$. The slope of this line increases when the radius of the tube decreases, thus showing a typical effect of capillarity.

We now perform a least-square fit of the early data in Fig. 1 with a linear function, so as to determine the slope in Eq. (16). The outcome of the fit is as follows:

$$R=65\ \ \mu\text{m}:\quad \frac{4K\varphi_0}{\sqrt{\alpha\pi}\gamma_1}=1.769\times10^{-9}\ \ \text{cm}^2\ \text{s}^{-1},$$

$$\sigma^2=6.755\times10^{-9}\ \ \text{cm}^2,$$

$$R=150\ \ \mu\text{m}:\quad \frac{4K\varphi_0}{\sqrt{\alpha\pi}\gamma_1}=3.038\times10^{-10}\ \ \text{cm}^2\ \text{s}^{-1},$$

$$\sigma^2=4.117\times10^{-8}\ \ \text{cm}^2, \tag{17}$$

$$R = 200 \ \mu\text{m:} \quad \frac{4K\varphi_0}{\sqrt{\alpha\pi}\gamma_1} = 2.246\times 10^{-10} \ \text{cm}^2 \ \text{s}^{-1},$$

$$\sigma^2 = 3.157\times 10^{-8} \ \text{cm}^2,$$

where σ is the standard deviation. The agreement thus obtained with the data in Sec. II is shown in Fig. 5. For lyotropic liquid crystals it is sensible to assume that K/γ_1 is on the order of 10^{-8} cm^2 s^{-1}. Taking it exactly as 10^{-8} cm^2 s^{-1}, from Eq. (18) we arrive at the following measurements of the anchoring angle:

$$R = 65 \ \mu\text{m:} \quad \varphi_0 = 0.048,$$

$$R = 150 \ \mu\text{m:} \quad \varphi_0 = 0.008, \tag{18}$$

$$R = 200 \ \mu\text{m:} \quad \varphi_0 = 0.006.$$

It is worth noting that the constant pressure acting on both defects cannot possibly be interpreted as a long-range electric effect, since in lyotropic liquid crystals these effects are completely screened.

V. PERSPECTIVES

The above analysis opens the way to some further experiments. Actually, the experiment described above was originally set up to observe the coalescence of two point defects with opposite topological charges, and so no attention was paid to the role of the anchoring, which afterwards turned out to be crucial. Thus, some questions remain open.

First, with refined optical equipment, it is perhaps possible to measure the tilt angle φ_0, at least for the smallest capillary. One could then compare a direct measurement of the deviation from homeotropic anchoring with the theoretical prediction made in Eq. (16). Once the validity of Eq. (16) is proved for liquid crystals with well-known material constants, this law could also be employed to compute the rotational viscosity of less known liquid crystals.

Second, two defects with opposite charges attract each other only if the angle φ_0 has the appropriate sign, that is, the one which makes the more distorted field $\boldsymbol{n}_+$ fill the region between them. Otherwise, the pressure acting on both defects changes sign, thus forcing them to repel each other. If attention is paid to the direction of filling, it could be related to the sign of φ_0. Maybe a more difficult task would be to find a connection between the velocity of filling and the magnitude of φ_0.

Third, it should be proved experimentally that after a defect has traversed a certain region of the tube no other defect traverses it, since the field must have relaxed to the absolute minimizer.

Finally, for liquid crystals with negative diamagnetic anisotropy, applying a magnetic field parallel to the capillary axis could decrease the elastic energy stored in the field $\boldsymbol{n}_+$ and increase that stored in $\boldsymbol{n}_-$, so as to fill the gap between them. There should be a critical magnetic field able to stop the motion of a defect: above this threshold, the defect should then start moving again, but in the opposite direction.

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