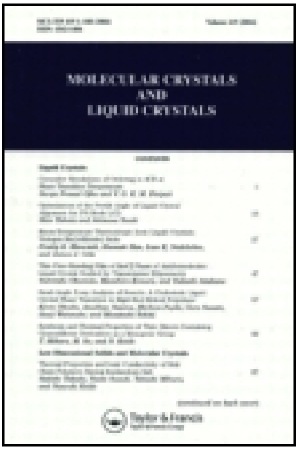

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Axially Symmetric Director Configurations of Inverted Neiiiatic Emulsions

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Director configurations for inverted nematic emulsions with axial symmetry are evaluated. In particular, a chain of two water droplets in a spherically confined nematic solvent is investigated by employing the finite element technique. We confirm an experimental observation that the distance d of the point defect from the surface of a water droplet scales with the radius γ of the droplet like $d \approx 0.3\ \gamma$. When the water droplets are moved apart, we find that the point defect does not stay in the middle between the droplets, but rather forms a dipole with one of them.

Keywords: complex geometries; topological defects

INTRODUCTION

For several years now, an extensive study has been devoted to liquid crystals confined to complex geometries, like drops in a polymer matrix or a random porous network in silica aerogel.[1] This article presents a numerical investigation of the inverse problem, which is posed by particles suspended in a nematic solvent.[2–4] We address recent work on inverted nematic emulsions where surfactant-coated water droplets are dispersed in a nematic solvent.[2,5] The advantage of such a system is that the particles are easily observable by polarizing microscopy since the size of the water droplets is of the order of a micron. Furthermore, the anchoring of the liquid crystal molecules on the surfaces of the droplets is controllable via the surfactant. The most striking feature, from a theoretical point of view, is that inverted emulsions provide an ideal laboratory for the investigation of topological defects. In inverted emulsions point defects, also called hedgehogs, occur. When a water droplet with homeotropic, *i.e.*, perpendicular anchoring of the director at its surface is placed into a uniformly aligned nematic liquid crystal, a hyperbolic hedgehog is nucleated. The droplet and its tightly bound companion defect provide a key unit for our understanding of inverted nematic emulsions. We will call it a dipole for short because of its dipolar symmetry. A phenomenological theory predicts that the dipole couples to a splay deformation in the director field.[2,6] Two parallel dipoles are attracted via a long-range dipolar interaction which explains the observed chaining of water droplets.

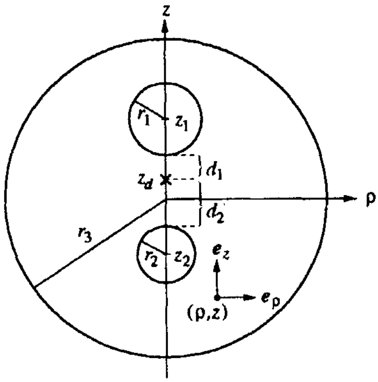

FIGURE 1 Coordinate frame and geometry parameters for two water droplets in a large nematic drop.

In our numerical investigation we consider multiple nematic emulsions where the nematic host fluid containing the water droplets is confined to large nematic drops, which, in turn, are surrounded by water. The water droplets form chains with a hyperbolic hedgehog situated between two droplets. In the experiment it was found that the distance d of the point defect from the surface of a water droplet scales with the radius r of the droplet like $d \approx 0.3\,r$.[3,6] In the following we will call this relation the scaling law. A possible explanation for the chaining, as in a uniformly aligned sample, could be the presence of a dipole. One water droplet fits perfectly into the center of a large nematic drop. Any additional water droplet has to be accompanied by a hyperbolic hedgehog for topological reasons. If the dipole forms, it is attracted by the strong splay deformation in the center, until a short-range repulsion mediated by the defect sets in.

GEOMETRY AND NUMERICAL METHOD

In our article we numerically investigate a particular geometry of axial symmetry. According to Fig. 1 we consider two spherical water droplets with respective radii r_1 and r_2 in a large nematic drop with radius r_3. Due to the axial symmetry both the water droplets and the hyperbolic hedgehog are always located on the z axis. We employ a cylindrical coordinate system. The coordinates z_1, z_2, and z_d denote, respectively, the positions of the centers of the droplets and of the hyperbolic hedgehog on the z axis. The distances of the hedgehog from the surfaces of the two water droplets are, respectively, d_1 and d_2.
We, furthermore, restrict the nematic director to the (ρ, z) plane, which means that we do not allow for twist deformations. The director is expressed in the local coordinate basis $(\hat{\boldsymbol{\rho}}, \hat{\mathbf{z}})$,

$$\mathbf{n}(\rho, z) = \sin\Theta(\rho, z)\,\hat{\boldsymbol{\rho}} + \cos\Theta(\rho, z)\,\hat{\mathbf{z}} \ , \tag{1}$$

where we introduced the *tilt angle* Θ. At all the boundaries we assume a rigid homeotropic anchoring of the director, which allows us to omit any surface term in the free energy.

In order to find the equilibrium director field the reduced Oseen-Zöcher-Frank free energy[7]

$$\overline{F} = \int d^3x\,\overline{f}(\Theta) \ , \tag{2}$$

with the reduced free energy density

$$\begin{aligned} \overline{f}(\Theta) &= \frac{\overline{K}_{11}}{2}\left(\frac{\sin\Theta}{\rho} + \Theta_\rho \cos\Theta - \Theta_z \sin\Theta\right)^2 \\ &\quad + \frac{1}{2}\left(\Theta_z \cos\Theta + \Theta_\rho \sin\Theta\right)^2 \ , \end{aligned} \tag{3}$$

is minimized. In Eqs. (2) and (3) all lengths are given in units of a characteristic scale a, typically several microns, and $\overline{K}_{11} = K_{11}/K_{33}$ is the ratio of the splay and the bend elastic constant. Accordingly, the reduced free energy $\overline{F}$ is measured in units of $K_{33}\,a$. The indices ρ and z indicate, respectively, partial derivatives $\partial/\partial\rho$ and $\partial/\partial z$. Because of the nontrivial geometry of our problem we decided to employ the method of finite elements[8], where the integration area is covered with triangles. In the area between the small spheres we chose a refined grid to account for the strong director deformations close to the point defect.[9] To find a minimum of the free energy we start with a configuration where the hyperbolic point defect is located at z_d and let it relax via the standard Newton-Gauss-Seidel method. During the relaxation the position z_d of the point defect does not change.

All our calculations are performed for the nematic liquid crystal pentylcyanobiphenyl (5CB), for which the experiments were done[4,5]. It has a bend elastic constant $K_{33} = 0.53 \times 10^{-6}$ dyn and a scaled splay elastic constant $\overline{K}_{11} = K_{11}/K_{33} = 0.79$. We choose $r_3 = 7$, $r_{1/2} = 0.5 \ldots 2$ for the radii, and $b = 0.195$ for the lattice constant of the grid. With such parameters we obtain a lattice with 2200-2500 vertices.

RESULTS AND DISCUSSION

In this section we present some selected results from our numerical investigations.[9] We start with the search for the minimum energy configuration. In Fig. 2 we plot the reduced free energy $\overline{F}$ as a function of the distance $d_1 + d_2$ between the surfaces of the small spheres, which are placed symmetrically about the center, *i.e.*, $z_2 = -z_1$. Their radii are $r_1 = r_2 = 1$.

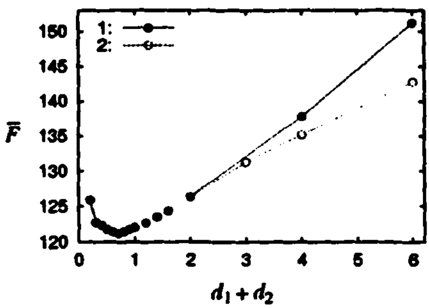

FIGURE 2 The free energy $\overline{F}$ as a function of the distance $d_1 + d_2$ between the small spheres which are placed symmetrically about $z = 0$ ($r_1 = r_2 = 1$). Curve 1: $z_d = 0$, curve 2: position z_d of the defect can relax along the z axis.

Curve 1 shows a clear minimum at $d_1 + d_2 \approx 0.7$, the defect stays in the middle between the two spheres at $z_d = 0$. In curve 2 we move the defect along the z axis and plot the minimum free energy for each fixed distance $d_1 + d_2$. It is obvious that beyond $d_1 + d_2 = 2$ the defect moves to one of the small spheres. Before we investigate this result in more detail we first address the scaling law.

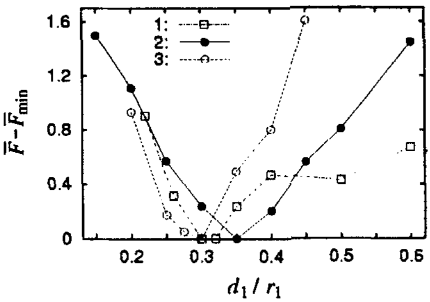

FIGURE 3 The free energy $\overline{F} - \overline{F}_{min}$ as a function of $d_1/r_1 = d_2/r_2$. The small spheres are placed symmetrically about $z = 0$. Curve 1: $r_1 = r_2 = 0.5$, curve 2: $r_1 = r_2 = 1$, and curve 3: $r_1 = r_2 = 2$.

In Fig. 3 we take three different radii for the small spheres, $r_1 = r_2 = 0.5, 1, 2$, and plot the free energy versus d_1/r_1 close to the minimum. (Recall that d_1 is the distance of the hedgehog from the surface of sphere 1.) Since for such small distances $d_1 + d_2$ the defect always stays at $z_d = 0$, *i.e.*, in the middle between the two spheres, we have $d_1/r_1 = d_2/r_2$. The quantity $\overline{F}_{min}$ refers to the minimum free energy of each curve. For each ratio we obtain an energetically preferred distance d_1/r_1 in the range of [0.3, 0.35], which agrees well with the experimental value of 0.3.

When we move the two spheres with radii $r_{1/2} = 1$ together in the same direction along the z axis, the defect always stays in the middle between the droplets and obeys the scaling law. We have tested its validity within the range [0,3] for the defect position z_d. Of course, the absolut minimum of the free energy occurs in the symmetric position of the two droplets, $z_2 = -z_1$.

We further check the scaling law for $r_1 \neq r_2$. We investigate two cases. When we choose $r_1 = 2$ and $r_2 = 0.6$, we obtain $d_{1/2} \approx 0.3\, r_{1/2}$. In the second case, $r_1 = 2$ and $r_2 = 1$, we find $d_1 \approx 0.37\, r_1$ and $d_2 \approx 0.3\, r_2$. As observed in the experiment the defect sits always closer to the smaller sphere. There is no strong deviation from the scaling law

$d_{1/2} = (0.325 \pm 0.025)\, r_{1/2}$, although we would allow for it, since $r_1 \neq r_2$.

Now we turn to the question if the dipole formed by one water droplet and a companion hyperbolic point defect has a meaning in our geometry. To answer this question we place sphere 2 with radius $r_2 = 1$ in the center of the nematic drop at $z_2 = 0$. Then, we determine the energetically preferred position of the point defect for different locations z_1 of sphere 1 ($r_1 = 1$). The position of the hedgehog is indicated by $\Delta = (d_2 - d_1)/(d_1 + d_2)$. If the defect is located in the middle between the two spheres, Δ is zero since $d_1 = d_2$. On the other hand, if it sits at the surface of sphere 1, Δ is one since $d_1 = 0$. In Fig. 4 we plot the free energy $\overline{F}$ versus Δ. In curve 1, where the small spheres are farthest apart from each other ($z_1 = 5$), we clearly find the defect close to sphere 1. This verifies that the dipole is existing. It is thermally stable, since a rough estimate of the thermally induced mean displacement of the defect yields 0.01[6]. When sphere 1 is approaching the center (curves 2 and 3), the defect moves away from the droplet until it nearly reaches the middle between both spheres (curve 4). This means, the dipole vanishes gradually until the hyperbolic hedgehog is shared by both water droplets.

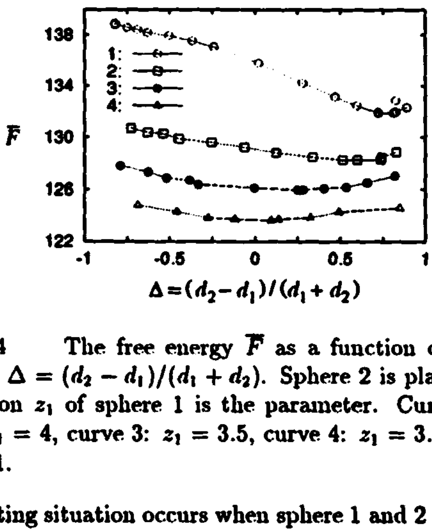

FIGURE 4 The free energy $\overline{F}$ as a function of the reduced coordinate $\Delta = (d_2 - d_1)/(d_1 + d_2)$. Sphere 2 is placed at $z_2 = 0$. The position z_1 of sphere 1 is the parameter. Curve 1: $z_1 = 5$, curve 2: $z_1 = 4$, curve 3: $z_1 = 3.5$, curve 4: $z_1 = 3$. The radii are $r_1 = r_2 = 1$.

An interesting situation occurs when sphere 1 and 2 are placed symmetrically about $z = 0$. Then, the defect has two equivalent positions on the positive and negative part of the z axis. In Fig. 5 we plot again the free energy $\overline{F}$ versus the position Δ of the defect. From curve 1 to 3 the minimum in $\overline{F}$ becomes broader and more shallow. The defect moves closer towards the center until at around $z_1 = -z_2 = 2.3$ (curve 4) it reaches $\Delta = 0$. This is reminiscent to a symmetry-breaking second order phase transition[10] which occurs when, in the course of moving the water droplets apart, the dipole starts to form. We take Δ as an order parameter, where $\Delta = 0$ and $\Delta \neq 0$ describe, respectively, the high- and the low-symmetry phase. A Landau expansion of the free energy yields

$$\overline{F}(\Delta) = \overline{F}_0(z_1) + a_0[2.3 - z_1]\Delta^2 + c(z_1)\Delta^4 \ , \qquad (4)$$

where $z_1 = -z_2$ plays the role of the temperature. Odd powers in Δ are not allowed because of the required symmetry $\overline{F}(\Delta) = \overline{F}(-\Delta)$. This free energy qualitatively describes the curves in Fig. 4.

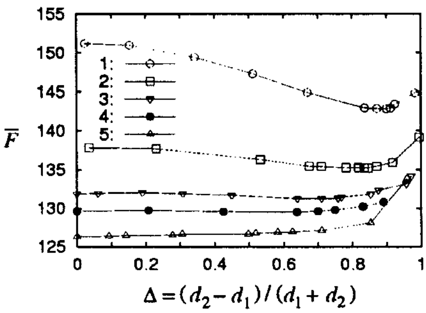

FIGURE 5 The free energy $\overline{F}$ as a function of the reduced coordinate $\Delta = (d_2 - d_1)/(d_1 + d_2)$. The small spheres are placed symmetrically about $z = 0$. Curve 1: $z_1 = -z_2 = 4$, curve 2: $z_1 = -z_2 = 3$, curve 3: $z_1 = -z_2 = 2.5$, curve 4: $z_1 = -z_2 = 2.3$, curve 5: $z_1 = -z_2 = 2$. The radii are $r_1 = r_2 = 1$.

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