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Twist transition in nematic droplets: a stability analysis

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The twist transition of the radial configuration between two concentric spheres with rigid perpendicular anchoring at the surfaces is examined. We perform a stability analysis and derive a sufficient condition for the twist transition to take place. We show that even a small sphere inside a large one can stabilize the radial configuration which confirms recent experiments. The light transmission of the twisted director field in a nematic drop is calculated by means of Jones matrices. The result agrees qualitatively with experimental images.

1. Introduction

Nematic configurations within confined geometries have attracted a lot of interest in liquid crystal research during the last decade [1]. They are of considerable technological importance in polymer dispersed liquid crystals (PDLCs), which are used as light shutters. On the other hand, they represent appealing systems whereby topological defects and the influence of confining geometries on bulk phase transitions can be studied. It has been shown, both experimentally and theoretically, that the director configuration strongly depends on the type of surface anchoring, on the strength of an applied magnetic field, and on the bulk and surface elastic constants.

In this paper we will mainly concentrate on nematic configurations within spherical droplets which occur either in nematic emulsions [2] or in a polymer matrix in PDLCs [3]. Nearly 30 years ago two main director configurations depending on the surface anchoring were predicted: a radial one with a point defect at the centre for perpendicular orientation of the molecules at the droplet surface and a bipolar one with two surface defects called boojums for tangential anchoring [2]. This simple picture had to be modified when it was found that nematic droplets in both cases may also exhibit a twisted structure [4]. For the bipolar configuration a stability analysis for the twist transition was performed [5].

From the theoretical side, it was pointed out that the perpendicular anchoring at the surface induces a defect structure in the interior of the droplet which is called a hedgehog and which carries a topological charge 1. There are two main classes of these hedgehogs, a radial one and a hyperbolic one, the latter being energetically favoured for a ratio of bend to splay constants smaller than six [6, 7]†. It has also been proposed that the twisted radial director configuration in a nematic drop is given by a combination of a hyperbolic hedgehog at the centre of the drop and a radial one at the periphery [8]. This configuration, which involves a twist in the director field, has been analysed by means of an ansatz function, and a criterion has been given for the twist transition [8].

In this paper we focus on the director field between two concentric spheres with perpendicular anchoring at both the surfaces and present a stability analysis for the radial configuration against axially symmetric deformations. In particular, we will derive a criterion for the twist transition and we will show that even small spheres inside a large one are sufficent to avoid twisted configurations. This has been recently observed in experiments on multiple nematic emulsions [9].

Throughout the paper we assume a rigid surface anchoring of the molecules. For completeness we note that in a single droplet for sufficiently weak anchoring strength an axial structure with an equatorial disclination ring appears [10].

The rest of this paper is organized as follows: In §2 we write the Frank free energy in terms of small deviations from the radial configuration. Then, in §3, we formulate and solve the corresponding eigenvalue equation. The lowest eigenvalue leads to the criterion for the twist transition. Finally, the article closes with a discussion of our results in §4.

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† We disregard here the influence of the saddle-splay term with the elastic constant K_{24} which alters this criterion [7].

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2. Expansion of the elastic energy

We consider the defect-free radial director configuration between two concentric spheres of radii $r_{\min}$ and $r_{\max}$. Such a geometry is, for example, realized in multiple nematic emulsions, where surfactant-coated water droplets are dispersed in large nematic drops, which, in turn, are surrounded by the water phase [9]. Rigid perpendicular anchoring of the director at both the surfaces can be achieved by a special choice of the surfactant [9, 11]. If the smaller sphere with radius $r_{\min}$ is missing, the radial director configuration exhibits a point defect at the centre. We will argue below that this situation, $r_{\min} = 0$, is included in our treatment.

The twist transition reduces the $SO(3)$-symmetry of the radial director configuration to an axial C_{∞}-symmetry. In order to investigate the stability of the radial configuration $\mathbf{n}_0 = \mathbf{e}_r$ against a twist transition, we write the local director in a spherical coordinate basis allowing for small deviations along the polar (θ) and the azimuthal (ϕ) direction:

$$\mathbf{n}(r,\theta) = \left(1 - \frac{1}{2}b^2 f^2 - \frac{1}{2}a^2 g^2\right)\mathbf{e}_r + ag\mathbf{e}_\theta + bf\mathbf{e}_\phi. \quad (1)$$

$f(r,\theta)$ and $g(r,\theta)$ are general functions, which do not depend on ϕ due to our assumption of axial symmetry. The amplitudes a and b describe the magnitude of the polar and azimuthal deviation of the director field from the radial configuration. The second order terms in b and a result from the normalization of the director.

The Frank free energy of a nematic liquid crystal is given as a sum of three elastic terms:

$$F = \int d^3r\{K_{11}(\nabla\cdot\mathbf{n})^2 + K_{22}(\mathbf{n}\cdot\nabla\times\mathbf{n})^2 + K_{33}[\mathbf{n}\times(\nabla\times\mathbf{n})]^2\}. \quad (2)$$

Since we consider strong anchoring at all the surfaces, we do not take into account any surface terms. We calculate the splay, twist, and bend contribution of the elastic energy up to second order in a, b and find:

$$(\nabla\cdot\mathbf{n})^2 = \frac{1}{r^2}[4 - 4b^2(f^2 + rf_rf) - 4a^2(g^2 + rg_rg) + a^2(\cot\theta g + g_\theta)^2 + 4a(\cot\theta g + g_\theta)] \quad (3)$$

$$(\mathbf{n}\cdot\nabla\times\mathbf{n})^2 = \frac{1}{r^2}b^2(\cot\theta f + f_\theta)^2 \quad (4)$$

$$[\mathbf{n}\times(\nabla\times\mathbf{n})]^2 = \frac{1}{r^2}[b^2(f + rf_r)^2 + a^2(g + rg_r)^2] \quad (5)$$

where an index denotes a partial derivative; e.g. $f_r = \partial f/\partial r$. For the untwisted configuration ($a = b = 0$) only the splay term of the Frank free energy contributes and its total free energy is given by

$$\int_{r_{\min}}^{r_{\max}} r^2\,dr \int_{S^2} d\cos\theta\, d\phi K_{11}(\nabla\cdot\mathbf{n})^2 = 8\pi K_{11}(r_{\max} - r_{\min}). \quad (6)$$

If an azimuthal ($b \neq 0$) or a polar component ($a \neq 0$) of the director is introduced, the splay energy can be reduced at the cost of non-zero twist and bend contributions depending on the values of the Frank elastic constants K_{11}, K_{22}, and K_{33}.

With the help of equations (3), (4), and (5), the free energy ΔF of the director field of equation (1) with respect to the radial configuration can be written as:

$$\begin{aligned}\Delta F = {} & 2\pi b^2 \int dr \int d\cos\theta[-4K_{11}(f^2 + rf_rf) \\ & + K_{22}(\cot\theta f + f_\theta)^2 + K_{33}(f + rf_r)^2] \\ & + 2\pi a^2 \int dr \int d\cos\theta[-4K_{11}(g^2 + rg_rg) \\ & + K_{11}(\cot\theta g + g_\theta)^2 + K_{33}(g + rg_r)^2].\end{aligned} \quad (7)$$

Note that the linear term in a in the splay term of equation (3) vanishes when integrated over θ.

For any function $f(r,\theta)$ leading to a negative value of the first integral in equation (7), the radial configuration ($a = b = 0$) is unstable with respect to a small azimuthal deformation ($b \neq 0$), which introduces a twist into the radial director field. Therefore, in what follows we will call it the twist deformation. An analogous statement holds for $g(r,\theta)$ which introduces a pure bend into the radial director field. We are now looking for the condition which the elastic constants have to fulfil in order to allow for such functions $f(r,\theta)$ and $g(r,\theta)$. As we will demonstrate in the next section, the solution of this problem is equivalent to solving an eigenvalue problem.

3. Formulating and solving the eigenvalue problem

In a first step we focus on the twist deformation ($b \neq 0$). We are facing the problem of knowing for which values of K_{11}, K_{22}, K_{33} the functional inequality

$$\begin{aligned}\int dr \int dx\{ & K_{22}(1 - x^2)[xf/(1 - x^2) - f_x]^2 \\ & + (K_{33} - 4K_{11})f^2 + (2K_{33} - 4K_{11})rf_rf \\ & + K_{33}r^2 f_r^2\} < 0\end{aligned} \quad (8)$$

has solutions $f(r, x)$. The integral in this inequality is the first integral of equation (7) after substituting $x = \cos\theta$.

Inserting the identities

$$r^2 f_r^2 = \frac{\partial}{\partial r}(r^2 f_r f) - f\frac{\partial}{\partial r}(r^2 f_r)$$

$$r f_r f = \frac{1}{2}\frac{\partial}{\partial r}(r f^2) - \frac{1}{2} f^2$$

$$-2x f_x f = -\frac{\partial}{\partial x}(f^2 x) + f^2$$

$$f_x^2(1-x^2) = \frac{\partial}{\partial x}[(1-x^2) f_x f] + 2x f_x f - (1-x^2) f_{xx} f$$

into equation (8) we obtain surface terms which are zero because of our boundary conditions. Finally, we arrive at

$$\frac{\int \mathrm{d}r \int \mathrm{d}x (K_{22} f D^{(x)} f + K_{33} f D^{(r)} f)}{\int \mathrm{d}r \int \mathrm{d}x\, f^2} < 2K_{11} \qquad (9)$$

where the second order differential operators $D^{(x)}$ and $D^{(r)}$ are given by

$$D^{(x)} = (1-x^2)\frac{\partial^2}{\partial x^2} + 2x\frac{\partial}{\partial x} + \frac{1}{1-x^2} \qquad (10)$$

$$D^{(r)} = -r^2\frac{\partial^2}{\partial r^2} - 2r\frac{\partial}{\partial r}. \qquad (11)$$

The inequality in equation (9) is best fulfilled when the left hand side assumes a minimum. According to the Ritz principle in quantum mechanics this minimum is given by the lowest eigenvalue of the operator

$$K_{22} D^{(x)} + K_{33} D^{(r)} \qquad (12)$$

on the space of square-integrable functions with $f(r_{\min}, \theta) = f(r_{\max}, \theta) = 0$ for $0 \leqslant \theta \leqslant \pi$ (fixed boundary condition) and $f(r, 0) = f(r, \pi) = 0$ for $r_{\min} \leqslant r \leqslant r_{\max}$.

The eigenvalue equation of the operator $K_{22} D^{(x)} + K_{33} D^{(r)}$ separates into a radial and an angular part. The radial part is a Eulerian differential equation [12] with the lowest eigenvalue

$$\lambda_0^{(r)} = \frac{1}{4} + \left[\frac{\pi}{\ln(r_{\max}/r_{\min})}\right]^2 \qquad (13)$$

and the corresponding eigenfunction

$$f^{(r)}(r) = \frac{1}{\sqrt{r}} \sin\left[\pi \frac{\ln(r/r_{\min})}{\ln(r_{\max}/r_{\min})}\right]. \qquad (14)$$

The angular part of the eigenvalue equation is solved by the associated Legendre functions $P_n^{m=1}$. The lowest eigenvalue is $\lambda_0^{(x)} = 2$ and the corresponding eigenfunction is $f^{(x)}(\theta) = P_1^1(\theta) = \sin\theta$.

With both these results we obtain the instability condition for a twist deformation:

$$\frac{1}{2}\frac{K_{33}}{K_{11}}\left\{\frac{1}{4} + \left[\frac{\pi}{\ln(r_{\max}/r_{\min})}\right]^2\right\} + \frac{K_{22}}{K_{11}} < 1. \qquad (15)$$

This inequality is the main result of the paper. If it is fulfilled, the radial director field no longer minimizes the Frank free energy. Therefore it is a sufficient condition for the radial configuration to be unstable against a twist deformation. It is not a necessary condition since we have restricted ourselves to second order terms in the free energy, not allowing for large deformations of the radial director field. Hence we cannot exclude the existence of further configurations which besides the radial produce local minima of the free energy.

To clarify our last statement we take another view. The stability problem can be viewed as a phase transition. Let us take K_{33} as the 'temperature'. Then condition (15) tells us that for large K_{33} the radial state is the (linearly) stable one. If the phase transition is second order-like, the radial state loses its stability exactly at the linear stability boundary, while for a first order-like transition the system can jump to the new state (due to non-linear fluctuations) even well inside the linear stability region. Thus, as long as the nature of the transition is not clear, linear stability analysis cannot predict with certainty that the radial state will occur in the linear stability region. Furthermore, if the transition line is crossed, the linear stability analysis breaks down, and there could be a transition from the twisted to a new configuration. However, there is no experimental indication of such a new structure. With this in mind we will discuss the instability condition (15) in the next section.

We finish this section by noting that the elastic energy for a bend deformation ($a \neq 0$) has the same form as that for the twist deformation ($b \neq 0$), but with K_{22} replaced by K_{11}, cf. equation (7). Therefore, we immediately conclude from equation (15) that the instability condition for a polar component ($a \neq 0$) in the director field (1) cannot be fulfilled for positive elastic constants. A director field with vanishing polar component is always stable in the second order.

4. Discussion

The instability condition (15) indicates for which values of the elastic constants K_{11}, K_{22}, and K_{33} the radial configuration is expected to be unstable with respect to a twist deformation. The instability domain is largest for $r_{\max}/r_{\min} \to \infty$ and decreases with decreasing ratio $r_{\max}/r_{\min}$; that is, a water droplet inside a nematic drop can stabilize the radial configuration.

In figure 1 the instability condition (15) is shown. If the ratios of the Frank elastic constants define a point in the grey triangles, the radial configuration can be unstable depending on the ratio $r_{\max}/r_{\min}$. The dark grey area gives the range of the elastic constants where a twisted structure occurs for $r_{\max}/r_{\min} = 50$. With increasing ratio $r_{\max}/r_{\min}$ the instability domain enlarges until it is limited by $K_{33}/(8K_{11}) + K_{22}/K_{11} = 1$ for $r_{\max}/r_{\min} \to \infty$. The light grey triangle is the region where the radial configuration is unstable for $r_{\max}/r_{\min} > 50$, but where it is stable for $r_{\max}/r_{\min} < 50$.

The circles in figure 1 represent the elastic constants for MBBA, 5CB, and PAA. For 5CB the elastic constants are in the light grey domain, i.e. a twisted structure is expected for $r_{\max}/r_{\min} \to \infty$ (no inner sphere), but not for $r_{\max}/r_{\min} < 50$. Such a behaviour has been recently observed in multiple nematic emulsions [9]. It has been found that a small water droplet inside a large nematic drop prevents the radial configuration from twisting.

Two examples of nematic drops observed under the microscope between crossed polarizers can be seen in figure 2. In the upper image the director configuration is pure radial, in the lower one it is twisted. The upper drop contains a small water droplet that stabilizes the radial configuration according to equation (15). The water droplet is not visible in this image because of the limited resolution. A better image is presented in [9].

We have calculated the polarizing microscope picture of the twisted configuration by means of the 2×2 Jones matrix formalism [1]. We took the director field of equation (1) and used the eigenfunction of equation (14) with an amplitude $b = 0.15$. The result shown in figure 3 is in qualitative agreement with the lower experimental image in figure 2.

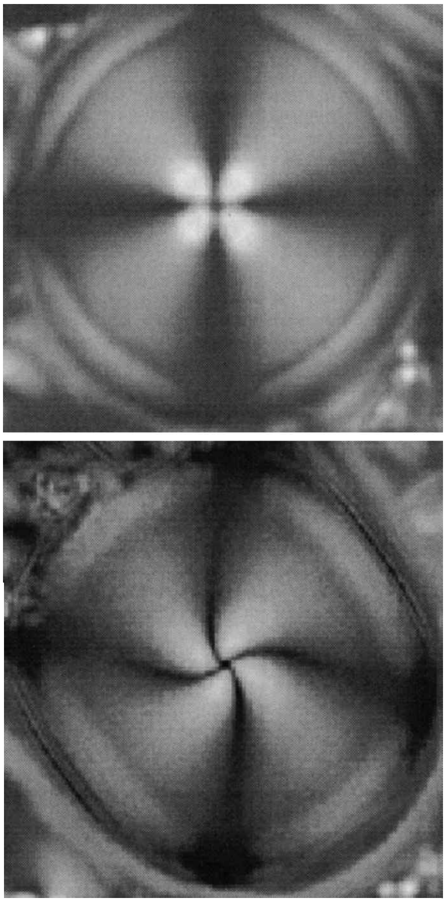

Figure 2. Radial (above) and twisted (below) configuration of the director field in a nematic drop (diameter $\approx 20\,\mu$m) of 5CB observed in the microscope between crossed polarizers. In the radial configuration there is a small isotropic liquid droplet in the centre of the nematic drop (invisible in this image). Courtesy of P. Poulin.

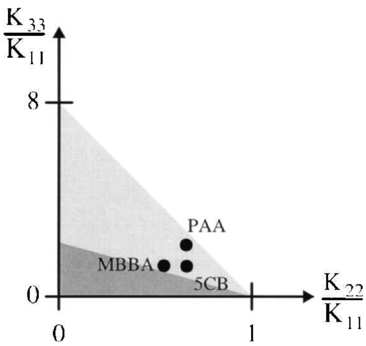

Figure 1. Stability diagram for the twist transition, cf. equation (15). The dark grey triangle corresponds to the ratios of Frank constants where the radial configuration is unstable for a ratio $r_{\max}/r_{\min} = 50$. The light grey triangle is the region where the radial configuration is unstable for $r_{\max}/r_{\min} > 50$. The circles represent the elastic constants for MBBA, 5CB and PAA.

In figure 4 we plot the radial part $f^{(r)}(r)$ [see equation (14)] of the eigenfunction $f(r, \theta) = f^{(r)}(r) f^{(x)}(\theta)$ governing the twist deformations. For large values of $r_{\max}/r_{\min}$ it is strongly peaked near $r_{\min}$. The maximum of $f^{(r)}(r)$ occurs at a radius r_0 which is given by

$$\ln \frac{r_0}{r_{\min}} = \frac{\ln(r_{\max}/r_{\min})}{\pi} \arctan \frac{2\pi}{\ln(r_{\max}/r_{\min})}. \qquad (16)$$

Hence, for $r_{\max}/r_{\min} \gg 1$ the maximal azimuthal component $bf(r_0, \theta)$ of the director field is located at $r_0/r_{\min} = e^2 \approx 7.39$, i.e. close to the inner sphere. From the polarizing miscroscope pictures it can be readily seen that the twist deformation is largest near the centre of the nematic drop. In the opposite limit, $r_{\max}/r_{\min} \approx 1$, the position of maximal twist is at the geometric mean of $r_{\min}$ and $r_{\max}$: $r_0 = (r_{\min} r_{\max})^{1/2}$.

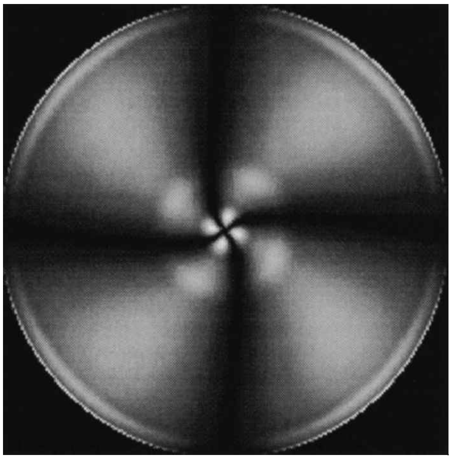

Figure 3. Calculated transmission for the twisted configuration of the director field in a nematic drop whose diameter is 20 µm. The transmission amplitude was obtained by summing over 20 wavelengths between 400 and 800 nm. The amplitude b of the twist deformation was set to 0.15. This figure has to be compared with the lower image in figure 2.

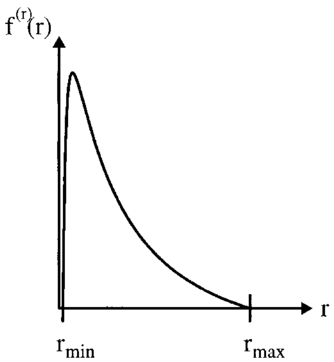

Figure 4. Radial dependence of $f^{(r)}(r)$ [cf. equation (14)] for $r_{max}/r_{min}=50$. The function is strongly peaked close to r_{min}.

In the limit $r_{min}\to 0$, where the inner sphere is not present, a point defect with a core radius r_c is located at $r=0$. In this case our boundary condition, $f^{(r)}(r_{min})=0$, makes no sense, since the director is not defined for $r<r_c$. Fortunately, for $r_{min}\to 0$ the lowest eigenvalue of the operator (12) and therefore the instability condition is insensitive to a change of the boundary condition. Furthermore, the shape of the eigenfunction is independent of the boundary condition; in particular its maximum is always located close to r_{min}.

A last comment concerns the work of Lavrentovich and Terentjev [8]. In figure 5 we plot as a dashed line the criterion, $K_{33}/(4K_{11})+K_{22}/(2K_{11})=1$, which the authors of [8] derived for the twist transition in the case $r_{max}/r_{min}\to\infty$. They constructed an ansatz function which connects a hyperbolic hedgehog at the

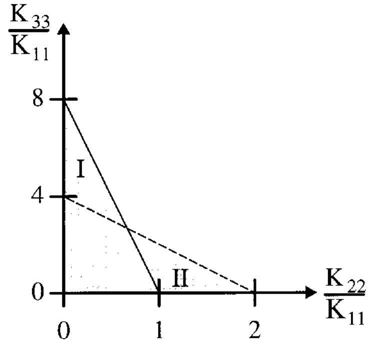

Figure 5. A comparision between the regions of instability for a radial director field against twisting derived in this article (full line) and by Lavrentovich and Terentjev (dashed line) for $r_{max}/r_{min}\to\infty$. The regions differ by the areas I and II.

centre via a twist deformation to a radial director field at the periphery of a nematic drop. Then they performed a stability analysis for an appropriately chosen order parameter. The region of instability calculated in this article and their result differ by the areas I and II. This is due to the complementarity of the two approaches. While the authors of [8] allow for large deviations with respect to the radial configuration at the cost of fixing an ansatz function, we allow the system to search the optimal configuration (i.e. eigenfunction) for small deformations. We conclude that both results together give a good approximation of the region of instability for the radial configuration against twisting. However, we cannot exclude that a full non-linear analysis of the problem leads to a change in the stability boundaries.

In conclusion, we have performed a linear stability analysis of the radial configuration in nematic drops with respect to a twist deformation. Assuming strong perpendicular anchoring at the surfaces we have derived an instability condition in terms of the elastic constants. We could show that a small water droplet inside the nematic drop stabilizes the radial configuration.

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