Proceedings of the Steklov Institute of Mathematics, Vol. 223, 1998, pp. 168—174.
Translated from Trudy Matematicheskogo Instituta imeni V.A. Steklova, Vol. 223, 1998, pp. 171—177.


# Equilibrium Shape of a Nematic Liquid-Crystal Droplet

A.G. Kalugin and A.N. Golubyatnikov

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In this paper, we carry out an analytical and numerical analysis of the equilibrium problem for a nematic liquid-crystal droplet suspended in an ordinary isotropic fluid. We demonstrate how the orientation of the easy axis affects the droplet shape. When the direction of the easy axis is close to the normal, the droplet is typically oblate, whereas, when the axis is close to the tangent to the surface, the droplet is extended along the symmetry axis. For intermediate values of the surface-orientation angles. the droplet exhibits conical peaks on its poles.

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## 1. INTRODUCTION

Nematic liquid crystals (or oriented fluids) represent the media that are characterized by the elasticity of the orientation of the long axes of molecules. The surface tension at the interface between such media also exhibits certain anisotropic properties associated with the internal orientation of the media. In this case, the surface tension may distort the parallel orientation of lines inside a volume even in the free (equilibrium) state of the oriented fluid. A review of theoretical models of the surface tension in liquid crystals and certain experimental results was presented in [1].

In this paper, we carry out an analytical and numerical analysis of the equilibrium problem for a nematic liquid-crystal droplet suspended in an ordinary isotropic fluid. We demonstrate how the orientation of the easy axis affects the droplet shape. When the direction of the easy axis is close to the normal, the droplet is typically oblate, whereas, when the axis is close to the tangent to the surface, the droplet is extended along the symmetry axis. For intermediate values of the surface-orientation angles, the droplet exhibits conical peaks on its poles.

## 2. THE BASIC VARIATIONAL EQUATION

Within the Oseen–Frank model, the equilibrium of a liquid-crystal droplet can be described on the basis of the L.I. Sedov variational equation [2].

Suppose that $\mathbf{A}(x^i)$ is a unit vector field of the orientation of a medium and $x^i$ are the Eulerian coordinates of points, $i = 1, 2, 3$. Supposing that the state of the continuum is isothermal, we assume that the volume and surface densities of the free energy are given by the following scalar functions:

$$F_V(g_{ij}, A^i, \nabla_i A^j), \qquad F_S(A^i, n_i),$$

where $g_{ij}$, $A^i$, and $\nabla_i A^j$ are the components of the metric tensor, vector $\mathbf{A}$, and its covariant derivative, respectively, and $\mathbf{n}$ is a unit outward normal to the surface. Due to the scalar nature of the functions $F_V$ and $F_S$, the essential arguments of these functions are scalars that consist of the above components. In particular, $F_S$ depends only on $A_n = A^i n_i$.

Consider the equilibrium of an oriented fluid droplet that occupies a certain Lagrangian volume $V$ bounded by the surface $S$ and is immersed into an isotropic fluid. Both fluids are assumed to be incompressible, have identical constant densities $\rho$, and be situated in the gravitational field with the potential $U(x^i)$. Then the following relation holds:

$$\delta\left(\int\limits_{V_0}(F_V+p(1-\Delta))\,d\tau_0+\int\limits_S F_S\,d\sigma\right)=\delta\int\limits_{V_0}\rho U\,d\tau_0+\int\limits_S p_e\delta x_n\,d\sigma\,,$$

where $p$ is the internal pressure used as a Lagrange multiplier in the incompressibility condition, $\Delta=\det(\partial x^i/\partial\xi^q)$, $p_e=\rho U+\mathrm{const}$ is the external pressure, and $V_0$ is the invariable volume.

Independently varying the positions $x^i(\xi^p)$ of the particles of the medium under constant Lagrangian variables $\xi^p$, the orientation filed $\mathbf{A}$ of the medium under the condition $|\mathbf{A}|=1$, and the pressure, we obtain the following system of equilibrium equations and boundary conditions:

$$\nabla_i(p+F_V-\rho U)=0\,,\qquad(\delta_k^j-A^jA_k)\left(\frac{\partial F_V}{\partial A^j}-\nabla_i\left(\frac{\partial F_V}{\partial\nabla_iA^j}\right)\right)=0\,,\tag{1}$$

in the volume $V$ and

$$p_i^jn_j=\nabla_\alpha\sigma_i{}^\alpha-p_en_i\,,\qquad(\delta_k^j-A^jA_k)\left(\frac{\partial F_V}{\partial\nabla_iA^j}n_i+\frac{dF_S}{dA_n}n_j\right)=0\,,\tag{2}$$

$$p_j{}^i=-p\delta_j{}^i-\nabla_jA^l\frac{\partial F_V}{\partial\nabla_iA^l}\,,\qquad\sigma^i{}_\alpha=x^i_\alpha F_S-A_\alpha n^i\frac{dF_S}{dA_n}\,,\tag{3}$$

on the surface $S$; here, $p_j{}^i$ is the stress tensor introduced by Ericksen, and $\sigma^i{}_\alpha$ is the tensor of surface tensions. The surface covariant derivative $\nabla_\alpha$, $\alpha=1,2$, is calculated by the metric $a_{\alpha\beta}=g_{ij}x^i_\alpha x^j_\beta$, where $x^i_\alpha=\partial x^i/\partial u^\alpha$ are the tangent vectors and $u^\alpha$ are the Lagrangian coordinates on the surface.

By virtue of equations (1), the following standard equilibrium equation of continuum mechanics is valid [2]:

$$\nabla_ip_j{}^i+\rho\nabla_jU=0\,.$$

The first equation in (1) defines the pressure: $p=\rho U-F_V+\mathrm{const}$. Later, the pressure can be eliminated in order to give an independent formulation of the problem of orientation distribution and the droplet shape.

When there are edges (or peaks) on the droplet surface that are not specially formed by the suspension- or support-type external conditions, we should adopt the balance condition for the linear forces,

$$(\sigma_i{}^\alpha m_\alpha)_++(\sigma_i{}^\alpha m_\alpha)_-=0\,,$$

that are calculated on both sides of the edge; here $\mathbf{m}_\pm$ are the corresponding external normals to the edge that lie on the surface. Since there is a term containing a normal to the surface in (3), in the general case, the linear force contains a tearing component, which is not characteristic of the surface tension of isotropic fluids.

By the second vector condition in (2), the first condition reduces to the single scalar relation

$$A^\alpha\nabla_\alpha A_n\frac{d^2F_S}{dA_n^2}+\nabla_iA^i\frac{dF_S}{dA_n}=b^\alpha_\alpha\left(F_S-A_n\frac{dF_S}{dA_n}\right)+p-p_e\,,$$

where $b_{\alpha\beta}=n_i\nabla_\alpha x^i_\beta$ is the second fundamental tensor of the surface. This result is associated with the fact that the theory does not depend on the choice of the surface coordinates $u^\alpha$ and allows one vary actually only one function describing the surface configuration, for example, in Eulerian coordinates.

If we take a vector product of the second equation in (1) multiplied by $\mathbf{A}$, we obtain the equation that is equivalent to the equation for the internal angular momentum of the medium [3].

Frequently, it is assumed that $F_S$ depends on $A_B$ rather than on $A_n$, where $\mathbf{B}$ is a unit vector that specifies the direction of the so-called easy-orientation axis, which makes an angle $\omega$ with the normal to the surface. Typically, $F_S$ is a decreasing function of ${A_B}^2$, and we can assume that $0 \le \omega \le \pi/2$. Varying the orientation of $\mathbf{B}$ for given $\mathbf{A}$ and $\mathbf{n}$, we obtain that $|A_B|$ attains its maximum when the vector $\mathbf{B}$ lies in the plane of vectors $\mathbf{A}$ and $\mathbf{n}$ and closer to the straight line directed along $\mathbf{A}$. In this case,

$$|A_B| = \sin\omega\sqrt{1 - A_n^2} + \cos\omega|A_n| \,. \tag{4}$$

## 3. THE EQUILIBRIUM OF A DROPLET

Consider the functional

$$E = \int\limits_V F_V \, d\tau + \int\limits_S F_S \, d\sigma \,, \tag{5}$$

where $V$ is the variable volume ($d\tau = \Delta d\tau_0$).

The variation of functional (5) in the class of functions $A^i(x^k)$ satisfying the second equation in (1), as well as in the class of shapes of the surface $S$ for a given volume $V$ and under the boundary conditions (2) (after eliminating the pressures $p$ and $p_e$), is equal to zero. The preservation of the volume given by $V = \int_S r_n \, d\sigma = V_0$, where $\mathbf{r}$ is a radius vector, can be added to functional (5) with the Lagrange multiplier $\lambda$. In addition, we should take into account the preservation of the coordinate of the droplet volume center given by $\int_S r^2 \, \mathbf{n} \, d\sigma = 0$.

Consider a simple version of the theory, the so-called one-constant approximation, when

$$F_V = \frac{1}{2} K \nabla_i A_j \nabla^i A^j \,, \qquad F_S = \alpha + \frac{1}{2}\beta(1 - {A_B}^2) \,,$$

where $K$, $\alpha$, and $\beta$ are positive constants.

Depending on the physical and chemical treatment of the contact surface, the orders of magnitudes of the above constants (for example, for the MBBA [4]) are as follows: $K \sim 6\,\mathrm{pN}$, $\alpha \sim 0.04\,\mathrm{N/m}$, and $\beta$ ranges from $10^{-8}$ to $10^{-3}\,\mathrm{N/m}$. The characteristic radius of the liquid-crystal droplet (a radius when a droplet can be interpreted as a single crystal) is $R_0 \sim 10^{-7}\,\mathrm{m}$. Thus, we can construct two dimensionless parameters, $\varepsilon_1 = \beta/\alpha \sim 10^{-1}$–$10^{-7}$ and $\varepsilon_2 = \beta R_0/K \sim 10$–$10^{-4}$, and employ these parameters to simplify the equations.

When $\beta = 0$, the isoperimetric minimum problem for the functional $E$ for a given $V$ has a unique solution, $\mathbf{A} = \mathrm{const}$, and the droplet has a spherical shape. When $\beta > 0$, because $\varepsilon_1$ is small, the problem of the droplet shape can always be linearized with respect to the deviations from sphere. In this case, if $\varepsilon_2 \ll 1$ (weak anisotropy of the surface tension), the equations for the droplet shape and the orientation distribution become completely linear; therefore, they can be solved virtually independently; one just should check the agreement of signs that is necessary to minimize functional (5).

In the case of strong anisotropy, when $\varepsilon_2 \gg 1$, one has to solve nonlinear equations (1) (but with a given value of $|A_n| = \cos\omega$ on the sphere [5]) in order to determine the volume distribution of the orientation to the zero-order approximation, which, typically, is a final result of the solution. Then, the droplet shape is determined from the known values of the Ericksen stress tensor on the sphere. In this paper, we consider the case of weak anisotropy, which allows direct determination of the droplet shape.

We will seek the solution in the class of functions possessing the symmetry of the fundamental state. The value of $E$ increases as the order of symmetry decreases. Then, in the spherical coordinates $r$, $\theta$, $\varphi$, the contravariant components of the orientation vector and the normal vector to the droplet surface $r = R(\theta)$ are expressed as

$$\mathbf{A} = \left(\cos\chi, \frac{\sin\chi}{r}, 0\right), \qquad \mathbf{n} = \left(\frac{R, -R'/R, 0}{\sqrt{R^2 + R'^2}}\right).$$

Introduce the function $u = \theta + \chi(r,\theta)$ defined as the angle between the symmetry axis of the droplet and the vector $\mathbf{A}$. Then, by the symmetry of the basic solution with respect to the equatorial plane, the function $R(\theta)$ is even and $u(r,\theta)$ is odd with respect to the variable $x = \pi/2 - \theta$.

Taking into account (4), we can rewrite the functional $E + \lambda V$ as

$$E + \lambda V = \pi K \int_0^\pi \int_0^R \left(r^2 u_r^2 + u_\theta^2 + \frac{\sin^2 u}{\sin^2\theta}\right) \sin\theta \, dr \, d\theta$$

$$+ 2\pi \int_0^\pi R\sqrt{R^2 + R'^2}\left(\alpha + \frac{\beta}{2}\sin^2(\omega - h(\zeta))\right)\sin\theta\, d\theta + \frac{2\pi\lambda}{3}\int_0^\pi R^3 \sin\theta \, d\theta, \tag{6}$$

$$\zeta = \theta - u - \arctan(R'/R),$$

where $u_r$ and $u_\theta$ are the partial derivatives with respect to $r$ and $\theta$, respectively; here, we introduced the periodic sawtooth function $h = \arccos|A_n|$: $h(\zeta) = |\zeta|$ for $\zeta \in [-\pi/2; \pi/2]$ with the period $\pi$.

To take into account the nondifferentiability of the function $h(\zeta)$ in the neighborhood of arbitrary points $\zeta = \theta$ (that are not close to 0, $\pi/2$, or $\pi$) during subsequent linearization of the term containing this function, we should formulate certain boundary conditions at the salient points, such as the absence of linear or concentrated external forces on the droplet surface. In accordance with the symmetry properties of the desired solution, it is sufficient to solve the linearized problem only in the domain $0 \le r \le R_0$, $0 \le \theta \le \pi/2$.

Suppose that the basic solution corresponds to $u = 0$ and $R = R_0$. Introduce the following dimensionless variables (that are of order unity in magnitude):

$$r_1 = \frac{r}{R_0}, \qquad y(x) = \frac{R - R_0}{\varepsilon_1 R_0}, \qquad w(r,x) = \frac{u}{\varepsilon_2}.$$

Then, retaining in (6) the quadratic terms in $\varepsilon_1$ and $\varepsilon_2$ as the highest terms and varying this functional, we obtain the following equation for the orientation angle for $0 < x < \pi/2$:

$$(r^2 w_{r_1})_{r_1} + \frac{(w_x \cos x)_x}{\cos x} - \frac{w}{\cos^2 x} = 0, \tag{7}$$

$$w_{r_1}(1,x) = \frac{1}{2}\sin 2(x+\omega), \qquad w(r_1, 0) = w\left(r_1, \frac{\pi}{2}\right) = 0.$$

Redefining $\lambda$, we have the following equation for the shape of the droplet surface:

$$(y' \cos x)' = (2y + \lambda_1) \cos x - \frac{1}{2} \cos(3x + 2\omega). \tag{8}$$

Using the linearized isoperimetric condition $\int_0^{\pi/2} y \cos x \, dx = 0$, one can determine $\lambda_1$ from equation (8). For the smooth droplet surface, we have

$$\lambda_1 = -\frac{1}{6}(\sin 2\omega + \cos 2\omega).$$

When there are edges on the surface, the following quantity should be continuous:

$$\left(y' + \frac{1}{2} \sin 2(x + \omega)\right) \cos x;$$

this quantity is proportional to the radial component of the total linear force on a given side of the edge. Hence, when the parameter $\omega$ is constant, the droplet surface is everywhere smooth, except, possibly, the poles. When the function $y(x)$ is bounded and there are no concentrated forces on the poles of the droplet, the analysis of the asymptotic behavior of the solution to (8) as $x \to \pi/2$ shows that there exist conical peaks with $y' = \frac{1}{2} \sin 2\omega$ for almost every $\omega$.

Let us present the exact solutions to equations (7) and (8). Let $\omega = 0$; i.e., let the axis of easy orientation coincide with the normal. Then,

$$w = w_1 \equiv \frac{1}{4} r_1^2 \sin 2x, \qquad y = y_1 \equiv \frac{1}{24}(3 \cos 2x - 1). \tag{9}$$

Thus, the shape of the droplet represents an oblate ellipsoid of revolution. The minimum of functional (6) is guaranteed by the fact that the variables $w$ and $y'(x)$ have correlated (opposite) signs on the circle $r_1 = 1$. A similar solution exists for $\omega = \pi/2$. In this case, the signs of $w$ and $y$ in (9) are simply reversed. The surface shape represents an oblong ellipsoid.

It is convenient to determine the distribution of orientation lines (interpreted as the lines of flow of the vector field $\mathbf{A}(x^i)$) inside the droplet in the meridian plane $\varphi = 0$ by using the cylindrical coordinates $r_c = r \sin \theta$, $z = r \cos \theta$:

$$\frac{dr_c}{dz} = \tan u \approx u = \varepsilon_2 w. \tag{10}$$

Using solution (9), we obtain

$$r_c = C \exp\left(\pm \frac{\varepsilon_2 z^2}{4R_0^2}\right)$$

for $\omega = 0$ and $\omega = \pi/2$, respectively; here, the constant $C \in [0, 1]$. In the first case, the orientation lines move away from the symmetry axis as $z$ increases, whereas in the second, they approach the symmetry axis.

In the general case, the solution is sought in the following form:

$$w = w_1 \cos 2\omega + w_2 \sin 2\omega, \qquad y = y_1 \cos 2\omega + y_2 \sin 2\omega;$$

thus, the determination of the functions $w_2$ and $y_2$ reduces to the solution of equations (7) and (8) for $\omega = \pi/4$.

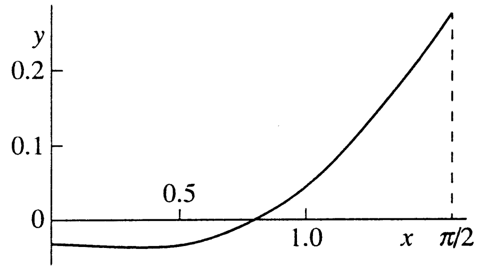


**Fig. 1**

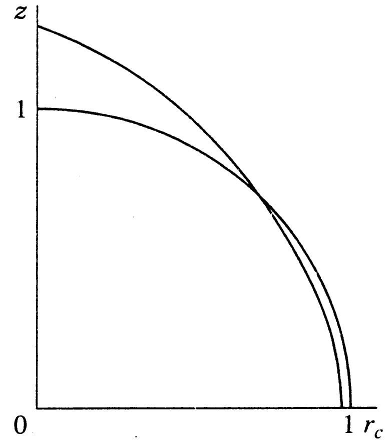


**Fig. 2**

## 4. DETERMINATION OF THE DROPLET SHAPE AND ORIENTATION FOR $\omega = \pi/4$

When $\omega = \pi/4$, equation (8) is rewritten as

$$(y' \cos x)' = \left(2y - \frac{1}{6}\right) \cos x + \frac{1}{2} \sin 3x\,;$$

we solved this equation numerically for the following boundary conditions at the ends of the integration interval: $y'(0) = 0$ and $y'(\pi/2) = 1/2$.

Calculations were performed by the standard sweep method. The accuracy of calculations was checked by verifying the volume-preservation condition, which was fulfilled with a relative error of $10^{-4}$. The results of calculating the droplet shape for $\omega = \pi/4$ in the cylindrical coordinates normalized by $R_0$ are illustrated in Fig. 1. For clearness, the graph is represented for $\varepsilon_1 = 1$. Figure 2 demonstrates the droplet shape in the meridian section. Because of the symmetry, we reproduce only a quarter of the section. For comparison, Fig. 2 also demonstrates the graph of a circle corresponding to the case of isotropic surface tension.

To determine the orientation of the medium for $\omega = \pi/4$, equation (7) is solved subject to the boundary conditions $w_{r_1}(1, x) = \frac{1}{2} \cos 2x$ and $w(r_1, 0) = w(r_1, \pi/2) = 0$. Let $w = -W_x(r_1, x)$. Then, the solution is reduced to the axially symmetric solution of the Neumann problem inside the ball $r_1 < 1$, $|x| \le \pi/2$ for the Laplace equation (in the function $W$) subject to the boundary condition $W_{r_1}(1, x) = \frac{1}{6} - \frac{1}{4}|\sin 2x|$. Using the expansion of $W$ in terms of spherical functions of even order and retaining the first two terms, we obtain

$$w = \frac{1}{48} r_1^2 \sin 2x + \frac{15}{1024} r_1^4 (-2 \sin 2x + 7 \sin 4x) + \dots\,. \tag{11}$$

To determine the lines of flow of the field $\mathbf{A}(x^i)$ in the plane $\varphi = 0$, we apply equation (10), which, in view of the smallness of $\varepsilon_2$, can be rewritten as

$$\frac{dr_c}{dx} = \frac{R_0 C \varepsilon_2}{\cos^2 x} w \left(\frac{C}{\cos x}, x\right), \qquad z = R_0 C \tan x\,, \qquad r_c(0) = R_0 C\,. \tag{12}$$

Substituting expression (11) into equations (12), performing the integration, and eliminating $x$, we obtain

$$r_c = R_0 C \left(1 + \frac{15\varepsilon_2}{256}\left(\frac{16}{45} + 5C^2 - 2z_1^2\right) z_1^2\right), \qquad z_1 = \frac{z}{R_0}.$$

The corresponding orientation lines represent W-shaped curves that have minima at $z = 0$ and two maxima at $z_1^2 = \frac{5}{4}C^2 + \frac{4}{45}$. The maxima lie inside the droplet when $C < 0.846$, which corresponds to the angles given by $|x| > 32.2°$.

Thus, the obtained results on the determination of the droplet shape and the distribution of internal orientation for $\omega = \pi/4$, together with formulas (9), corresponding to the case $\omega = 0$, allow us to give the complete solution of the problem for any value of $\omega$. In particular, for all $\omega \in (0, \pi/2)$, the droplet has conical peaks on its poles in the approximation considered.

## ACKNOWLEDGMENTS

This work was supported by the Russian Foundation for Basic Research, project nos. 97-01-00570 and 96-01-00376.

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*Translated by I. Nikitin*