

# Monopole structures and shape of drops of smectics-C

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It is demonstrated experimentally for the first time that in C-type smectic liquid crystals (SLC C) there exist monopole structures in the form of a concentric spherical system of SLC C layers, from the center of which there emerges either one  $(0,1)$  disclination or two  $(0,a^2)$  disclinations. It is shown theoretically that the existence of monopoles in SLC C is the consequence of the non-integrability of the phase of the wave function that describes the order parameter of the SLC C. An analytic expression is obtained for the gradient of this phase and coincides with the form of the vector-potential of the Dirac monopole. It is confirmed experimentally that a transformation from a 't Hooft-Polyakov monopole to a Dirac monopole is feasible in an SLC-A to SLC-C transition. A new phenomenon was observed in experiment, namely a change of the equilibrium form of SLC C drops from spherically symmetric to axisymmetric in the form of two connected spherical segments. The observed droplet shapes are the first example of an equilibrium form of an anisotropic body with two unequal values of the surface tension. From an examination of the functional of the surface energy of such droplets it is concluded that they correspond to a stable state of the system.

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## INTRODUCTION

Interest has considerably increased of late in the investigation of the so-called monopole structures. Their existence was first suggested by Dirac in a description of isolated magnetic charges.<sup>1</sup> Pointlike magnetic charges, according to Dirac's theory, are singularities of the electromagnetic field, and of dual character. On the one hand, the Dirac monopole is a point singularity in the distribution of the magnetic field  $H$ , which is directed along radius vectors drawn from the center of the charge; on the other hand, this point defect is connected with a linear singularity ("filament") of the field of the vector-potential  $A$ . The filament goes off from the center of the monopole to infinity.

In modern theory of gauge fields one considers one more possible magnetic-charge structure—without a linear singularity—the so-called 't Hooft-Polyakov monopole. Both the Dirac and the 't Hooft-Polyakov monopoles have quantized charges and are topological in nature—their structures are connected with definite topological invariants (see Refs. 3 and 4, respectively).

Notwithstanding the many arguments favoring the real existence of monopoles, experimental searches for these objects (see, e.g., Ref. 5) yielded no unambiguous positive result. Particular interest attaches therefore to experimental investigations of the possibility of formation of structural analogs of monopoles in different condensed media. The basis for the assumption of the existence of such structure is the general topological description of charge-singularities in field theory and defect-singularities of the ordering in condensed media (see, e.g., Ref. 6). Thus, for the superfluid anisotropic  $^3\text{He-A}$  structure called vorton was predicted in Ref. 7, and its mathematical description coincides in essence with the description of the Dirac monopole. It turned out later, however, that the vorton in  $^3\text{He-A}$  is unstable to relaxation into an energetically preferred configuration called "boojum" by Mermin.<sup>8</sup>

The first rigorous mathematical description of energetically stable monopole structures in condensed media was given by Volovik<sup>7</sup> within the framework a continual large-scale theory of cholesteric liquid crystals (CLC). In each case, for scales much larger than the pitch of the cholesteric helix, the distribution of the CLC distortion field coincides with the distribution of the Dirac-monopole vector potential, and the vector normal to the CLC layers plays the role of the magnetic field intensity in the structure of the magnetic charge—the CLC crystals form a spherical concentric system from the center of which emerges a  $\chi(+2)$  disclination—the analog of the filament. As indicated in Ref. 9, the  $\chi(+2)$  line can break up here into two  $\chi(+1)$  lines. The stability of the monopole in the CLC is due to the fact that the CLC layers are equidistant in the continual approximation (there is no such condition for the vorton in  $^3\text{He-A}$ , and this is why it is unstable and relaxes to a boojum.)<sup>9</sup>

The conclusions of Ref. 9 were confirmed<sup>9,10</sup> by experiment and monopole structures were indeed observed in CLC provided the large-scale condition  $P/R \sim 10^{-2}$  ( $P$  is the thickness of the CLC layer and  $R$  is the structure radius) was satisfied. Monopoles were observed both with one  $\chi(+2)$  disclination and with two  $\chi(+1)$  ones. We note that the structures with one  $\chi(+2)$  disclination in a concentric spherical system of layers were observed also for lyotropic cholesterics<sup>12</sup> and for nematic-cholesteric mixtures,<sup>13</sup> but only for the case  $P/R \sim 10^{-1}$ . At such (and larger) values of  $P/R$ , as shown experimentally in Refs. 11 and 14 and as follows from the theory,<sup>9</sup> the monopoles in CLC can become unstable.

Existence of monopole structures in smectic liquid crystals (SLC) was suggested in Ref. 15. One can expect the experimental realization of the large-scale requirement and of stable monopole structures in SLC to be simpler than for CLC, since the thickness  $d \approx 3 \times 10^{-9}$  m of the SLC layer is considerably smaller than the thickness  $P \approx 3 \times 10^{-7}$  m of the CLC layer. In addition, interest in experimental investi-

gations of such structures is due also to the fact that, as indicated in Ref. 15, in the SLC A to SLC C phase transition one can observe a transformation of the same type as that in the 't Hooft-Polyakov to Dirac monopole transition, with appearance of linear defects—disclinations of the types  $(0, I)$  and  $(0, a^2)$ . This circumstance can have a direct bearing on the notions developed recently concerning the disclination-disclination mechanism of phase transitions (see, e.g., Ref. 16). In Ref. 15, however, they did not exclude the possibility that this transition is not realized in SLC because of distortions of the stacking of the SLC layers.

Experimental investigations of the possibility of formation of monopole structures in SLC C and of a transition such as from the 't Hooft-Polyakov monopole to the Dirac monopole have not been made to this day. Such an investigation is reported in this paper. In addition, our purpose was also to study the structures of the cores of the disclinations  $(0, I)$  and  $(0, a^2)$ , as well as the possible distortions in the stacking of the SLC C layers in monopole structures.

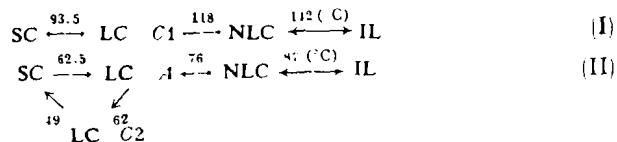
### EXPERIMENTAL TECHNIQUE

To investigate the possibility of formation of monopole structures in smectics it is necessary to create conditions under which the SLC layers have concentric spherical stacking, e.g., to produce liquid-crystal droplets that have a nearly normal boundary orientation of the molecules and assume a spherical shape under the action of surface-tension forces.

To produce such smectic droplets we dispersed the investigated material in an isotropic liquid matrix having a density close to that of the liquid crystal, but such that the two did not mix. This liquid was chosen to be glycerin to which small amounts (up to 1 wt. %) of lecithin solution were added.

The droplets investigated had a radius 5–30  $\mu\text{m}$ . The sample, in a quartz or glass cell, as placed in a thermostatically controlled (accurate to 0.1  $^{\circ}\text{C}$ ) heater. The texture of the droplets was investigated with an NU-2E polarization microscope in transmitted light.

Two mesomorphic substances were investigated: *n*-nonyloxy-benzoic acid (substance I) and butoxyphenyl ester of this acid (substance II), with the following phase diagrams:



Both substances have a smectic C phase, but the character of the temperature dependence of the angle  $\omega$  between the long axes of the molecules and the direction of the normal  $\mathbf{n}$  to the layers is different for them. For substance I the angle  $\omega \approx 45^{\circ}$  and is practically independent of temperature<sup>17</sup>; substance II shows a strong dependence of  $\omega$  on the temperature  $\omega = \omega_0(T_0 - T)^{0.4}$  (Ref. 18) ( $T_0$  is the temperature of the SLC A – SLC C transition). With increasing temperature, SLC C 1 and SLC C 2 go over respectively into the nematic and smectic A phase.

### EXPERIMENTAL RESULTS

The results of the investigation of the droplets of substances I and II are shown in Figs. 1 and 2.

*Substance I* (Fig. 1). When solid crystals of substance were heated in an isotropic matrix, they melted to form SLC C 1 droplets with radii  $R \approx 5\text{--}30 \mu\text{m}$ . The texture of the droplets as observed in polarized light has one or two defect lines emerging from the center of the drop to its surface (Figs. 1a, b, c). No other defects were observed in the droplets, thus attesting to a spherical stacking of the SLC C layers. This conclusion was confirmed also by observation through crossed Nicol prisms with the sample rotated in the horizontal plane.

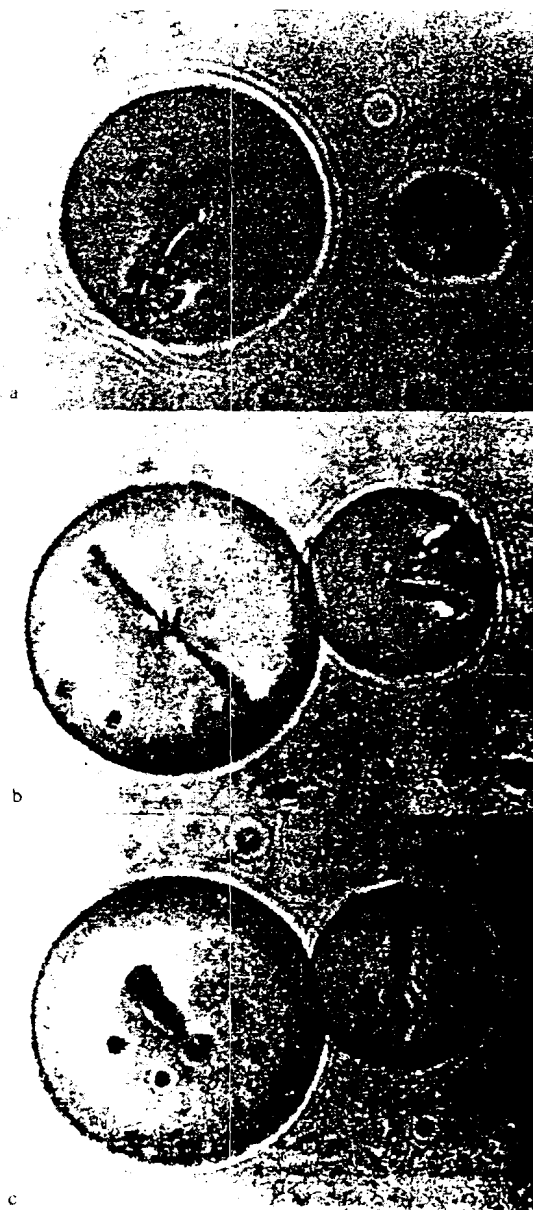


FIG. 1. Droplets of SLC C 1 in an isotropic glycerin matrix with lecithin added. Droplet radius  $R \approx 15\text{--}30 \mu\text{m}$ . There are no Nicol prisms, a—formation of droplet with one  $(0, I)$  disclination in the solid crystal to SLC C transition; b, c—droplet with two  $(0, a^2)$  disclinations whose spatial orientation changes.

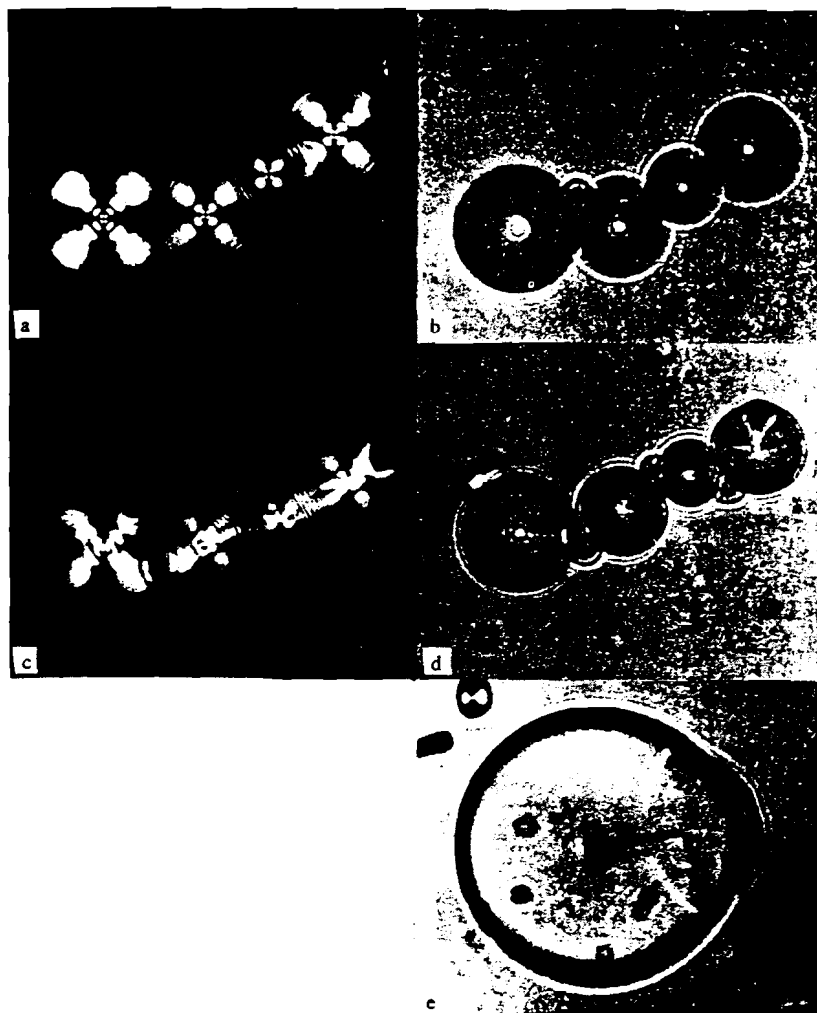


FIG. 2. Droplets of substance II in an isotropic matrix. Droplet radius  $R = 15-30 \mu\text{m}$ ; a, c) observation in crossed Nicols; b, c, e) without Nicols; a, b) SLC A droplets with point defects; c, d) the same droplets in the SLC C 2 phase with linear  $(0, a^2)$  disclinations; e) SLC C 2 droplet in the form of two spherical segments joined along the direction of the  $(0, I)$  disclination.

The orientation of the defect lines can change, and their visible length is a maximum if the lines are located in a horizontal plane and a minimum for orientation along the vertical (Figs. 1b, c). This circumstance is evidence that the defect is located inside the volume of the drop along the radius. We note that reorientation as well as slight bending of the lines is connected with viscous flow of the substance inside the droplets as a result of temperature gradients.

The cores of the defect lines, as can be seen from Fig. 1, have dimensions of the order of  $2-3 \mu\text{m}$ , much larger than the thickness of the CLC C layer.

The textures shown in Fig. 2 were produced both upon melting of the solid crystal, and following the NLC-SLC C 1 transition. The only difference was in the frequency at which the particular structure appeared in the drops; in the latter case two disclinations were produced approximately  $10^2-10^3$  times more frequently than one; in the former case this ratio was smaller by a decade. This difference is connected with the kinetics of the structure formation and with the actual conditions on the droplet surfaces. Thus, in the case of melting, one defect line is produced if a solid-crystal fragment is preserved in the droplets for some time (Fig. 1a). After the end of the melting process this disclination can break up into two, but if the disclination emerges to the sur-

face at a point where there is a foreign particle (another droplet, an air bubble, etc), the line remains stable.

It must also be noted that the shape of the SLC C 1 droplets, no matter how obtained, hardly differs from spherically symmetric, except for small distortions near the emergence of the defect lines to the surface.

*Substance II* (Fig. 2). The SLC C 2 is formed only by cooling the smectic A phase. The SLC A droplets are strictly spherical. When observed in crossed Nicols their texture has four extinction branches emerging from the droplet centers and coinciding with the directions of the Nicol polarizations (Fig. 2a), as well as annular isochromic lines. When a quartz edge is inserted, with the thin end forward, between the polarizer and the sample, the isochromic lines are shifted in the textures, namely, towards the droplet centers in quadrants I and III made up by the extinction branches, and in the opposite directions in quadrants II and IV. Since the SLC A are optically uniaxial and positive, this attests to radial orientation of the molecules in the droplets.<sup>19</sup> The SLC A layer stacking corresponding to this orientation is spherical concentric. Thus, a point defect of the field normal to the smectic layers is located at the center of the droplets. It is clearly seen when observed in unpolarized light (Fig. 2b).

In the SLC A-SLC C 2 transition, the textures in the

droplets are changed—the directions of the extinction branches become different (Fig. 2c) since the inclination of the molecules in the layers makes the smectic *C* optically biaxial. In addition, defect lines are produced in the droplets, two as a rule (Figs. 2c, d). These lines are analogous to the described defects in the SLC *C* 1 droplets, except that the size of their cores is  $\lesssim 1 \mu\text{m}$  and increases with increasing distance from the SLC *A*–SLC *C* 2 transition point.

When the sample is cooled 2–5 °C below the SLC *A*–SLC *C* 2 transition point the spherical shape of the droplets becomes distorted: emergence of the substance from the region of the core, one of the defect lines causes the droplets to assume the form of two spherical segments joined along the direction of this line. This form of the droplets remains stable at fixed temperature (Fig. 2a).

## DISCUSSION OF RESULTS

### 1. Monopole structures in SLC *C*

The microphotographs shown in Figs. 1a, b, c and 2c, d are evidence of formation of monopole structures in SLC *C* droplets in the form of concentric spherical systems of layers of the smectic *C*, from whose center emerge defect lines, either one of strength  $m = 2$  or two of strength  $m = 1$  each. We shall consider the mechanism whereby they are formed using as the example the SLC *C* 2 phase produced when SLC *A* is cooled.

The order parameter of SLC *A* is specified by one vector  $\mathbf{n}$  normal to the smectic layers, with states described by  $\mathbf{n}$  and  $-\mathbf{n}$  physically equivalent. In a spherical drop with normal boundary conditions there will be topologically allowed and energetically preferred the presence of one field defect  $\mathbf{n}$  whose topological charge  $q$  can be easily determined from the relation<sup>20</sup>

$$\sum_i q_i = E/2, \quad (1)$$

which connects the sum  $\sum_i q_i$  of the topological charges of the system of point defects with the Euler characteristic  $E$  of the closed surface where the condition  $\mathbf{n} = \pm \mathbf{v}$  is satisfied ( $\mathbf{v}$  is the vector normal to this surface). Since  $E = 2$  for a sphere and there is one point defect inside a spherical drop, its charge is

$$q = 1. \quad (2)$$

This singularity is located in the center of the droplet, for in view of the condition that the SLC *A* layers must be equidistant ( $\text{curl } \mathbf{n} = 0$ ) any displacement of the singularity from this position leads to the appearance of additional distortions in the system.

Only the scalar field of the projections of the ends of the molecules is specified on the projection of each SLC *A* layer. In SLC *C* this field is replaced by the vector field  $\boldsymbol{\tau}$  of the projections of the long axes of the molecules. The field  $\boldsymbol{\tau}$  is specified everywhere to be tangential to the SLC *C* layers. Besides the vector  $\boldsymbol{\tau}$ , a complete description of the order parameter of SLC *C* calls for the introduction of the vector  $\mathbf{n}$  normal to the layers, and their vector multiproduct  $\boldsymbol{\tau} \times \mathbf{n}$ .<sup>21</sup>

If the smectic layers retain their spherical stacking in

the course of the phase transition, there exists in the field  $\mathbf{n}$ , just as earlier in SLC *A*, a point defect  $q = 1$ . This defect, however, is no longer isolated—it is connected with the disclinations in the field  $\boldsymbol{\tau}$  (or  $\boldsymbol{\tau} \times \mathbf{n}$ ). The appearance of the latter is due to satisfaction of the Poincaré-Hopf topological theorem (see, e.g., Ref. 22) concerning the indices of the curls of a vector field that is continuous everywhere except at a finite number of points, and specified to be tangential to a closed surface.

$$\sum_i m_i = E, \quad (3)$$

where  $m_i$  is the index of the  $i$ -th curl. From relation (3) it follows that in a spherical droplet there must be produced linear defects in the field  $\boldsymbol{\tau}$  (or  $\boldsymbol{\tau} \times \mathbf{n}$ ) tangent to the closed spherical surfaces of the SLC *C* layers, for example on disclination of force  $m = 2$  or two disclinations with  $m = 1$  ( $(0, I)$  and  $(0, a^2)$ , respectively<sup>15</sup>). The produced structures are spherical concentric systems of SLC *C* layers, from the centers of which emerge either  $(0, I)$  disclinations of  $(0, a^2)$  disclination, and a monopole exists in SLC *C*.

Consequently the mechanism that produces monopole structures in SLC *C* is topologically equivalent to the production of Dirac monopoles—magnetic-field charges (see, e.g., Ref. 22)—and monopoles in CLC.<sup>11,23</sup>

Just as in the case of cholesterics,<sup>9</sup> for monole structures in SLC *C* it is possible to introduce a variable that characterizes directly the distribution of the molecules and its analytic expression is of the same form as the vector-potential field for a magnetic charge. Let us demonstrate this.

The order parameter of SLC *C* can be specified not only in the form of the triad  $\mathbf{n}, \boldsymbol{\tau}, \boldsymbol{\tau} \times \mathbf{n}$  but also with the aid of the wave function  $\psi = \omega \exp(i\phi)$ , where  $\omega$  is the angle of inclination of the molecules in the layers and  $\phi$  is the azimuthal direction of this inclination (Fig. 3).<sup>24</sup> The latter representation allows us to introduce an analogy with the description with superfluid helium-4, inasmuch as in both systems the total change of the phase does not change the free energy (the gauge-invariance property).<sup>24</sup> An even more complete analogy, however, is observed in the description of SLC *C*, superfluid anisotropic helium 3 (the <sup>3</sup>He-*A* phase), and the states of particles in quantum mechanics.

The change of phase  $\phi \rightarrow \phi + \alpha$  of the wave function that describes the SLC *C* has the meaning of rotation of the wave vectors  $\boldsymbol{\tau}$  and  $\boldsymbol{\tau} \times \mathbf{n}$  relative to  $\mathbf{n}$ . For the quantity  $\alpha$  at each point of the system we can specify its spatial derivatives

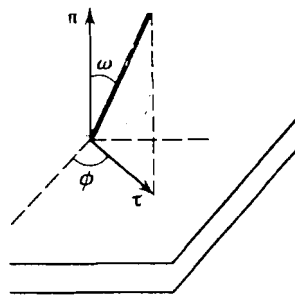


FIG. 3. The ordering of molecules in SLC *C* layers is described by specifying the inclination angle between the long axis of the molecule and the normal to the layer, as well as the azimuthal angle  $\phi$  of the inclination.

$$\kappa_x = \partial\alpha/\partial x, \quad \kappa_y = \partial\alpha/\partial y, \quad \kappa_z = \partial\alpha/\partial z. \quad (4)$$

Since the quantity  $\alpha$  is not a total differential of some function (rotations of the triad  $\mathbf{n}$ ,  $\boldsymbol{\tau}$ ,  $\boldsymbol{\tau} \times \mathbf{n}$  do not commute  $\partial^2\alpha/\partial x\partial y \neq \partial^2\alpha/\partial y\partial x$ , etc), in the general case  $\text{curl } \boldsymbol{\kappa} \neq 0$ . A similar result for  $^3\text{He-A}$  shows that the superfluid flow is not potential, and for the description of particles in quantum mechanics it shows that the magnetic field is not zero.<sup>1</sup> For SLC  $C$  the quantity  $\boldsymbol{\kappa}$  is the field of the distortions of the orientations of the molecule projections on the plane of the layers. If the latter constitute a spherical concentric system (its concentricity is ensured by the condition  $\text{curl } \mathbf{n} = 0$ ) we can write in the spherical coordinate system  $(r, \theta, \varphi)$

$$\text{rot } \boldsymbol{\kappa} = \boldsymbol{\tau}/r^2. \quad (5)$$

Solutions of the last equation are, for example,

$$\kappa_r = \kappa_\theta = 0, \quad \kappa_\varphi = 1/2 \text{tg}(\theta/2), \quad (6)$$

and

$$\kappa_r = \kappa_\theta = 0, \quad \kappa_\varphi = -1/2 \text{ctg}(\theta/2). \quad (7)$$

Expressions (6) and (7), which contain linear singularities directed respectively along  $r = -z$  and  $r = z$  coincide with the solutions for the vector potential of the Dirac monopole,<sup>1</sup> as well as with the expressions for the field of the distortions of the monopoles in CLC.<sup>9</sup> The field  $\mathbf{n}$  is radial in this case.

The appearance of structures of the Dirac-monopole type in SLC  $C$  is thus the consequence of non-integrability of the phase of the wave function ( $\text{curl } \boldsymbol{\kappa} \neq 0$ ) that describes the SLC  $C$  order parameter. For this reason the analogy 24 in the description of helium-4 and SLC  $C$  is incomplete, since there is no non-integrability condition for helium-4.

For SLC  $A$ , the phase  $\phi$  of the wave function becomes meaningless and there are no disclinations in the monopole structure: the only defect is a point defect of the field  $\mathbf{n}$  (a structure of the type of the 't Hooft-Polyakov monopole. The transition SLC  $A$ -SLC  $C$  2 in a spherical drop is indeed an analog, from the topological point of view, of the transformation of 't Hooft-Polyakov monopole into a Dirac monopole. When the droplets are heated, the reverse transition is also possible, and has been observed in experiment.

## 2. Linear $(0, l)$ and $(0, a^2)$ disclinations

The linear defects contained in the SLC  $C$  monopole structures are, as shown above,  $(0, l)$  and  $(0, a^2)$  disclinations. The latter are frequently encountered in smectic Schlieren textures,<sup>26</sup> but their study in these textures is made difficult by the circumstance that the lines are oriented perpendicular to the sample plane and are observed from the end. There is no such restriction in the case of monopole structures and oriented  $(0, a^2)$  disclinations in different planes, including the horizontal one, make possible a detailed study of their construction. The foregoing pertains to both the  $(0, l)$  disclinations in monopole structures, which were not described to this day for Schlieren textures. Thus, the monopole structures are convenient objects for the study of  $(0, l)$  and  $(0, a^2)$  disclinations.

We shall discuss the properties of these lines within the framework of the theory of homotopic groups according to

the classification of the defects in SLC.<sup>15,21</sup>

It is known that linear singularities in a three-dimensional condensed medium are described by the fundamental group  $\pi_1(V)$  of the degeneracy space  $V$  of a given medium (see, e.g., Ref. 27). For SLC  $A$  and SLC  $C$ , according to Ref. 15,

$$\pi_1(V_A) = Z \square Z_2, \quad (8)$$

$$\pi_1(V_C) = Z \square Z_4, \quad (9)$$

where  $V_A$  and  $V_C$  are the degeneracy spaces of SLC  $A$  and SLC  $C$ ,  $Z$  is the integer group, and  $Z_2$  and  $Z_4$  are the groups of residues in module 2 and 4, respectively. An element of the group  $\pi_1(V_C)$  can be written in the form  $(b, p)$ , where  $b$  is an integer that characterizes the dislocation part of the defect,  $p$  is one of the elements of the group  $Z_4 = (I, a, a^2, a^3)$  with unity element  $I$  and characterizes the disclination part. For defects that do not violate the translational invariance of the system (of the disclinations proper) we have  $b = 0$ . The elements  $(0, a_2)$  and  $(0, I) = (0, a^2)^2$  can be set in correspondence with a disclination in the field of molecule projections on the plane of the SLC  $C$  layers; the axis of such a disclination is directed along the normal  $\mathbf{n}$  to the layers. The disclinations  $(0, a_2)$  and  $(0, I)$  have respectively strengths  $m = 1$  and  $m = 2$  (Figs. 4a, b). It must be noted here that since  $I$  is the unity element of the group  $Z_4$ , the disclinations  $(0, I)$  can be continuously transformed into nonsingular configurations (Fig. 4c) or split into two  $(0, a^2)$  lines. The latter is energetically preferred, since the elastic energy of the distortions near the disclination is  $f \sim m^2 l$ , where  $l$  is the length of the defect.

On the face of it, the experimental observation of stable  $(0, l)$  lines in monopole structures indicates that the premises developed are contradictory (a similar conclusion was drawn in a discussion<sup>28</sup> of the properties of  $\chi(+2)$  lines in CLC—the analogs of  $(0, l)$  disclinations in SLC  $C$ . Actually, however, there is no contradiction, and observation of stable  $(0, l)$  lines under the conditions of our experiments is due, first, to the fact that their transition into an absolutely defect-free state is impossible by virtue of the nonzero value of the Euler characteristic of the droplet surface; second, the splitting of the  $(0, l)$  disclination into two  $(0, a^2)$  disclinations can be hindered by pinning of the end of the line by extraneous particles.

The elements  $(0, a^2)$  and  $(0, I)$  of the group  $\pi_1(V_C)$  constitute a subgroup that is isomorphic to  $Z_2$  and is the core of the holomorphism  $\pi_1(V_C) \rightarrow \pi_1(V_A)$ . A similar statement holds also for the homomorphism  $\pi_1(V_C) \rightarrow \pi_1(V_N)$ , where  $V_N$  is the degeneracy space of NLC.<sup>29</sup> This means vanishing of the corresponding disclinations in SLC  $C$ -SLC  $A$  and SLC  $C$ -

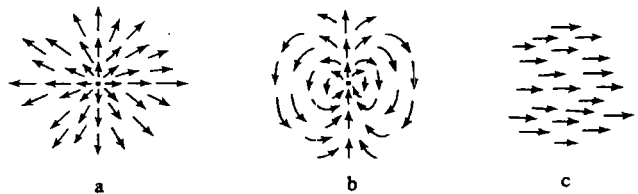


FIG. 4. Distributions of the field  $\boldsymbol{\tau}$  of the projections of SLC  $C$  on the layer plane for  $(0, l)$  and  $(0, I)$  disclinations and the defect-free state (a, b, and c respectively).

NLC transitions, as is indeed observed in experiment. One more consequence of these premises is that the  $(0, I)$  and  $(0, a^2)$  lines must have nonsingular cores of either SLC  $A$  or NLC. Other possible situations such as an isotropic core are also topologically allowed, but the formation of such a core calls for higher energy consumption than in the nematic or smectic  $A$  cores; on the other hand, the assumption that an empty core exists for defects in SLC  $C$ , which are in essence liquids, has no physical meaning.

That the cores of the  $(0, I)$  and  $(0, a^2)$  lines are not singular can be easily seen from Figs. 1a, b, c and 2c, d, since their thickness is several micron. As for the very structure of the core, the nematic model seems preferable. We shall demonstrate this by considering both the smectic  $A$  and the nematic models and comparing them with the experimental data.

**Smectic  $A$  model of core.** This model, as seen from Fig. 5a, is characterized by the presence of a singular cylindrical surface that separates the core of the defect (SLC  $A$ ) from its surrounding SLC  $C$  phase. On this interface the molecule orientation changes abruptly by an angle  $\omega$ . It is therefore logical to introduce the effective value of the surface energy with a tension coefficient  $\sigma_c$ , which is a function of the disorientation angle:  $\sigma_c(\omega) \sim \sigma_c^0 \sin^2 \omega$ , where  $\sigma_c^0$  is a constant (a similar approach was first undertaken in Ref. 30 to describe disclination cores in NLC). The energy per unit disclination length is then given by

$$F = m^2 K \sin^2 \omega \ln(R/r_c) + 2\pi r_c \sigma_c^0 \sin^2 \omega, \quad (10)$$

where  $K$  is the Frank elastic constant for transverse bending and  $r_c$  is the radius of the disclination core. Minimizing  $F$  with respect to  $r_c$  we obtain an estimate of the equilibrium value of the core dimension.

$$r_c = m^2 K / 4\pi \sigma_c^0. \quad (11)$$

Since  $K \sim U/d$  and  $\sigma_c^0 \lesssim U/d^2$ , where  $U$  is the energy of the intermolecular interaction in the SLC, and  $d$  is the characteristic molecular dimension ( $d \approx 20\text{--}30 \text{ \AA}$ ), it follows from (11) that

$$r_c \approx 3 \cdot 10^{-9} \text{ m}. \quad (12)$$

The estimate (12) of the dimension of the smectic  $A$  disclination core, as seen from Figs. 1a, b, c and 2c, d, does not agree with the experimental data ( $r_c \approx 10^{-6} \text{ m}$ ). Thus, the presence of a smectic  $A$  core in  $(0, I)$  and  $(0, a^2)$  disclinations is unlikely.

**Nematic model of core.** The need for the existence of a singular cylindrical surface surrounding the disclination core is obviated for the nematic model, since the difference between the orientations of molecules on opposite sides of the defects attunes itself smoothly to the deformations of the lateral bending (Fig. 5b). The situation is similar to the effect

of "escape to the third dimension" of disclinations in NLC with integer values of  $m$ ,<sup>31</sup> except that what escapes is not a planar but a bulk disclination,<sup>32</sup> since the molecules near a disclination in SLC  $C$  do not lie in one plane normal to the defect axis. The width of the core is governed in this case by the elastic constant and by the angle  $\omega$ . The qualitative dependence of the core width on  $\omega$  can be seen from a comparison of Figs. 1 and Figs. 2c, d for SLC  $C$  1 ( $\omega \approx 45^\circ$ ) and SLC  $C$  2 ( $0^\circ \leq 26^\circ$ ): in SLC  $C$  1 droplets the disclination cores are larger. In addition, when  $\omega$  changes with temperature the disclinations in SLC  $C$  2 become thicker with increasing  $\omega$ . Attention must be paid here to the fact that the cores of the two  $(0, a^2)$  lines in SLC  $C$  droplets differ somewhat in thickness (Figs. 2b, c and 2d). In all likelihood, the cause of the effect is that lines are not under identical conditions—the bending of the layers in the drop increases the misorientation angle of the molecules near the core for one of them, and decreases for the other.

It is known that the effect of escape of planar disclinations in NLC can lead to the appearance of discontinuities of the director field in the region of the smeared core,<sup>31,32</sup> since escape "up" and "down" along the disclination line is equally probable. In the case of disclinations in SLC  $C$  2 with small values of  $\omega$  escape is possible in only one of the two directions (up in Fig. 5b), and no singularities appear in the region of the cores (Fig. 2c, d). At the same time for SLC  $C$  1 ( $\omega \approx 45^\circ$ ) various distortions are observed along the disclination cores (Fig. 1). Recognizing that the angle  $\omega$  can be noticeably increased by the presence of Laplace pressure, this effect is attributed to the possibility of escape in both directions along the disclinations.

It can thus be concluded from the foregoing that the nematic model of the cores of the  $(0, I)$  and  $(0, a^2)$  disclinations is best capable of explaining the experimentally observed singularities of these lines.

### 3. Shape of smectic $C$ droplets

It follows from an experimental investigation of the SLC  $C$  2 droplets in a wide temperature interval that their shape can be appreciably changed (Fig. 2e) when the substance is cooled to a certain temperature  $T_1$  lower by 2–5  $^\circ\text{C}$  than the point  $T_0$  of the SLC  $A$ –SLC  $C$  2 transition (depending on the lecithin concentration in the matrix). The effect

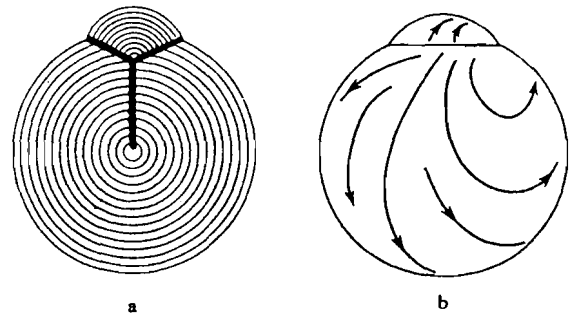


FIG. 6. Structure of droplets in the form of two spherical segments: a—distribution of SLC  $C$  layers, b—one of the possible distributions of the field  $\tau$  on the surface of the droplet, corresponding to the presence of a  $(0, I)$  disclination directed upwards, in the lower segment (not indicated in Fig. 6b) and to interface segments with defects between the segments.

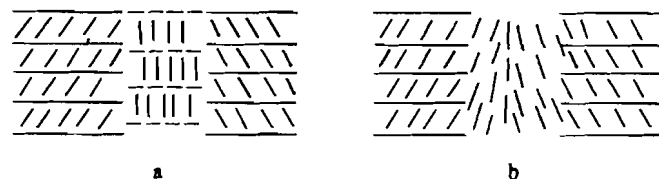


FIG. 5. Model of the structure of the  $(0, a^2)$  disclination cores a—smectic  $A$  core, b—nematic core.

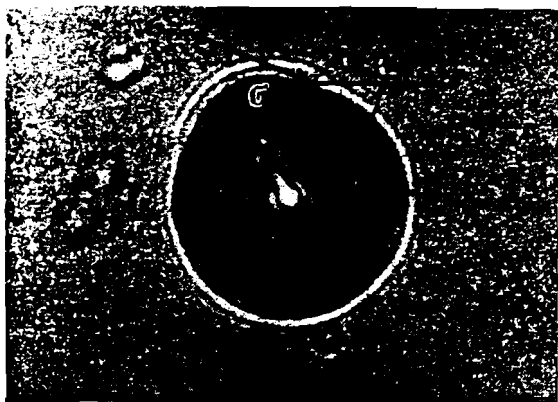


FIG. 7. Illustrating the condition for mechanical equilibrium of the forces  $\sigma_1$ ,  $\sigma_2$ , and  $\sigma_{12}$  on the contour of the interface between segments 1 and 2.  $\theta_1$  and  $\theta_2$  are the angles between the directions of the forces  $\sigma_1$  and  $\sigma_2$  and the normal to the  $PP'$  line, which has the same direction as  $\sigma_{12}$ .

consists of departure of the substance from the region of the core of one of the  $(0, a^2)$  disclinations of the monopole spherical structure (or from the core of the  $(0, J)$  disclination) and formation of an additional bulge on the droplet, as a result of which the latter takes the form of two spherical segments joined along the direction of the defect line.

It follows from observation in crossed Nicols, the stacking of the layers is concentric in both segments (Fig. 6a). The junction surface of the segments is concave towards the larger segment and the break of the linear disclination emerging from the center of the large segment takes place at the center of this interface. From the last fact, as well as from simple topological considerations, it follows that the segment-junction surface is a defect wall with the molecule orientations discontinuous on its opposite sides (Fig. 6b): absence of a disclination in the outer layers of the droplet is possible only if an annular linear defect is produced in each of them. It is the system of these lines which makes up the defect wall (a similar situation arises when a magnetic charge is described in the model of Su and Yang<sup>34</sup>).

Since the molecule orientations on the two sides of the segment interface are different, it is possible to introduce, as before, the concept of effective surface tension  $\sigma_{12}$  (the subscripts 1 and 2 pertain to the smaller and larger segments, respectively). Let us demonstrate the need for introducing the quantity  $\sigma_{12}$  and estimate its value.

As already noted, the droplets retain the form of two spherical segments at fixed temperature for an arbitrarily long time—tens of hours. Thus, this form corresponds to an equilibrium state of the droplet. It follows hence that the surface-tension forces  $\sigma_1$  and  $\sigma_2$  acting on the interface between the droplet and the matrix and applied to the boundary of the defect wall are balanced by the force  $\sigma_{12}$  (due to the surface tension  $\sigma_{12}$ ), which is also applied to the outer boundary of the defect wall, but is directed into the interior of the droplet (Fig. 7). In other words, the condition of mechanical equilibrium is satisfied (see, e.g., Ref. 35):

$$\sigma_1 + \sigma_2 + \sigma_{12} = 0. \quad (13)$$

It follows even from the sole fact that the forces  $\sigma_1$  and  $\sigma_2$  are not collinear that  $\sigma_{12}$  is not zero. Moreover, from (13) we can

quantitatively estimate the relation between  $\sigma_{12}$  and  $\sigma_1$  or  $\sigma_2$ . We consider for this purpose the condition (13) for one of the experimentally observed SLC C2 droplet shapes (Fig. 7). From an analysis of Fig. 7 and relation (13) we readily see that

$$\sigma_{12}/\sigma_2 = \sin \theta_2 (1 - \operatorname{tg} \theta_1 \operatorname{ctg} \theta_2) \quad (14)$$

(the angles  $\theta_1$  and  $\theta_2$  are defined in Fig. 7). Since  $\sigma_2 \sim \sigma_1 \sim 10^{-2}$  N/m (Refs. 36 and 24), the estimate  $\sigma_{12} \lesssim 10^{-3}$  N/m is valid (experiment yields  $1 - \tan \theta_1 \cot \theta_2 \sim 10^{-1}$ ).

We can estimate similarly the ration  $\sigma_1/\sigma_2$

$$\sigma_1/\sigma_2 = \cos \theta_2 / \cos \theta_1, \quad (15)$$

which yields  $\sigma_1 < \sigma_2$ , whereas in experiment we always have  $\theta_1 < \theta_2$ .

Since the phase composition of segments 1 and 2 is the same, the only reason why  $\sigma_1$  and  $\sigma_2$  are unequal is the difference between the inclination angles  $\omega_1$  and  $\omega_2$  of the molecules in the layers of the corresponding segments, while  $\omega_1$  and  $\omega_2$  are determined in turn, at a fixed temperature, by the Laplace pressure  $P$ . An analysis of the droplet shape shows that  $P_1 > P_2$  and hence  $\omega_1 > \omega_2$ . The last result, when juxtaposed with the inequality  $\sigma_1 < \sigma_2$ , indicates that the anisotropy  $\Delta\sigma$  of the surface tension for substance II in the temperature region  $T_1$  is positive, i.e., larger molecule inclination angles correspond to smaller values of the surface tension:

$$\Delta\sigma = \sigma_{\parallel} - \sigma_{\perp} > 0, \quad (16)$$

where  $\sigma_{\parallel}$  and  $\sigma_{\perp}$  are the values of the surface tension for molecule orientations at angles 0 and  $\pi/2$  to the normal to the interface. Since the condition  $\Delta\sigma < 0$  (which ensures the existence of spherical monopole structures of SLC C2), the inequality (16) leads to the conclusion that the sign of  $\Delta\sigma$  is reversed within the interval  $T_1 \lesssim T \lesssim T_0$ .

It is known<sup>35</sup> that a distinction must be made between the stable and metastable states. The experimental results indicate that in the region  $T \approx T_1$  the spherical droplet shape becomes metastable and its distortion means a transition of the system to a stable state. To ascertain when the shape of an SLC C2 droplet consisting of two spherical segments can indeed correspond to a stable state it is not enough to consider the condition (13), but it is necessary to find the minima of the droplet surface energy

$$E_s = \sigma_1 S_1 + \sigma_2 S_2 + \sigma_{12} S_{12},$$

where  $S_1$  and  $S_2$  are the areas of the surface segments and  $S_{12}$  is the area of the junction surface. We note that the contribution of the volume energy  $E_V$ , which is the energy of the elastic distortions, can be neglected compared with  $E_s$  since  $E_V/E_s \sim K/\sigma R \sim d/R \ll 1$ . The problem must be solved with account taken of a number of additional conditions: constancy of the droplet volume, dependence of the surface tension of the molecule inclination angle, and the dependence of the latter on the Laplace pressure. It is convenient here to use a toroidal coordinate frame in which the droplet surface coincides with the coordinate surfaces.<sup>37</sup> The condition that the volume  $v$  be constant takes then the form

$$\rho = 2R \left[ \sum_{i=1}^2 (2 + \sin^{-1}(\xi_i/2)) \operatorname{ctg}(\xi_i/2) \right]^{-1}, \quad (17)$$

where  $\rho$  is the radius of the boundary of the junction surface of the spherical segments,  $\xi_1$  and  $\xi_2$  are the angles at which the diameter of this circle is seen from the surfaces of the segments (Fig. 8), and  $R = (3\nu/4\pi)^{1/3}$  is the radius the drop would have if it were a sphere. We discuss now in greater detail the last two conditions.

The very fact that the surface tension depends on the inclination angle of the LC relative to the interface is well known (see, e.g., Ref. 38), but there are not experimental data to date on the exact character of this dependence for the SLC C. We shall therefore follow the approximation<sup>38</sup>

$$\sigma = \Delta\sigma \cos^2 \omega + \sigma',$$

where  $\Delta\sigma$  and  $\sigma'$  are constants (at  $T \lesssim T_1$ ) and  $\omega$  is the equilibrium value of the inclination angle of the molecules in the layers at a given temperature and zero pressure. We have then for  $\sigma_1$  and  $\sigma_2$

$$\sigma_1 = \Delta\sigma \cos^2 \omega_1 + \sigma', \quad \sigma_2 = \Delta\sigma \cos^2 \omega_2 + \sigma'. \quad (18)$$

The dependences of  $\omega_1$  and  $\omega_2$  on the Laplace pressure can be obtained by using Hooke's law:

$$\begin{aligned} \cos \omega_1 &= \cos \omega (1 - 2\sigma_1 \sin \xi_1 / B\rho), \\ \cos \omega_2 &= \cos \omega (1 - 2\sigma_2 \sin \xi_2 / B\rho), \end{aligned} \quad (19)$$

where  $B$  is Young's compression modulus of the SLC C 2 layers.

The problem reduces thus to finding the minima of the function

$$E_s = 2\pi\rho^2 \left( \frac{\sigma_1}{1 - \cos \xi_1} + \frac{\sigma_2}{1 - \cos \xi_2} \right) + \pi\rho^2 \sigma_{12} \quad (20)$$

(we assume  $S^{12} = \pi\rho^2$ ) subject to the additional conditions (17)–(19).

The problem is difficult to solve analytically, and a computer was used. The calculation  $E_s$  as a function of  $\xi_1$  and  $\xi_2$  (with  $\sigma$ ,  $\Delta\sigma$ ,  $\sigma_{12}$ ,  $B$ , and  $R$  as parameters) indicates that the appearance of droplets in the form of two spherical segments ( $\pi/2 < \xi_1 < \pi$ ,  $0 < \xi_2 < \pi/2$ ) can indeed correspond

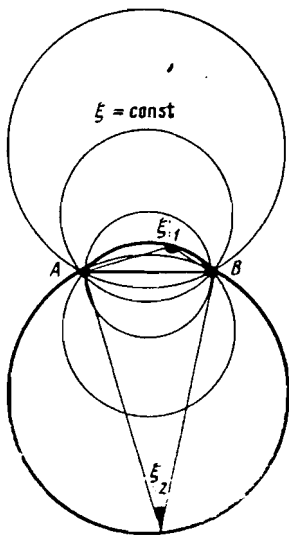


FIG. 8. Coordinate surfaces  $\xi = \text{const}$  in a toroidal coordinate frame.  $AB = 2\rho$ . The surfaces of the spherical segments of the SLC C coincide with the two surfaces  $\xi_1 = \text{const}$  and  $\xi_2 = \text{const}$ ; they are marked by thick lines.

to an equilibrium stable state and be energetically preferred to the existence of a spherical drop ( $\xi_1 = \pi$ ,  $\xi_2 = 0$ ) (see Fig. 9). This possibility is realized at the parameter values  $\sigma \sim 10^{-2}$  (Refs. 36 and 24),  $\Delta\sigma \sim (10^{-2} - 10^{-3})$  (Ref. 36),  $\sigma_{12} \lesssim 10^{-3}$  (in N/m,  $B = 2 \times 10^4$  N/m<sup>2</sup> (Ref. 39), and  $R = 5$  to  $50 \mu\text{m}$ , which agree with the parameters previously known from other investigations, and agree also with the experimental conditions in the present study.

## CONCLUSION

The results of an investigation of droplets of smectic liquid crystals of type C 1 and C 2, carried out in the present study, allow us to draw the following conclusions.

There can exist in SLC C monopole structures constituting systems of spherical concentric layers of SLC C with one (0, I) or two (0, a<sup>2</sup>) disclinations emerging from the center of the system. It is possible to introduce for such structures a variable that characterizes directly the ordering of the molecules, with a distribution that coincides with the form of the vector-potential field of the Dirac monopole. The onset of monopole structures is due to the non-integrability of the phase of the wave function that describes the order parameter of SLC C. This condition must be taken into account in the general case when SLC C is described.

The disclinations (0, a<sup>2</sup>) and (0, I) in SLC C have nonsingular cores with dimensions  $r_c \sim 1$  to  $3 \mu\text{m}$ ; the core thickness increases with increasing incidence angle of the molecules in the layers. The most suitable model for the description of the cores of the (0, a<sup>2</sup>) and (0, I) disclination is the nematic-core model.

A structural CLC A–CLC C phase transition comprising the onset of a (0, a<sup>2</sup>) or (0, I) disclination and preservation of one isolated point with topological charge  $q = 1$  is possible. It is of the same type as the 't Hooft–Dirac monopole transition.

Within the limits of the temperature interval in which the smectic C phase exists, reversal is possible of the sign of

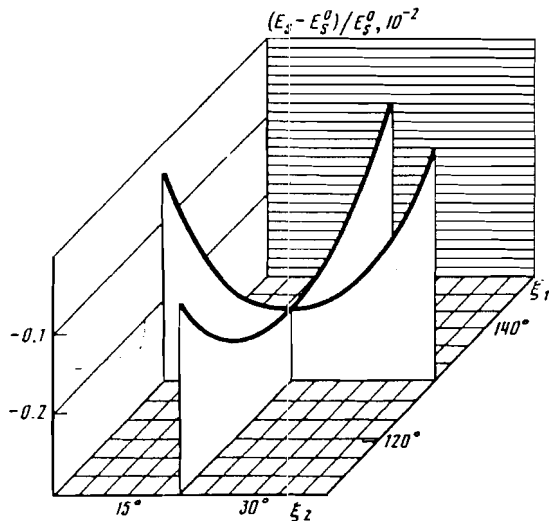


FIG. 9. Dependence of  $(E_s - E_s^0) / E_s^0 \cdot 10^{-2}$  on  $\xi_1$  and  $\xi_2$  for the following parameter values:  $\sigma = 2 \times 10^{-2}$ ,  $\Delta\sigma = 10^{-2}$ ,  $\sigma_{12} = 0.5 \times 10^{-3}$  (N/m),  $B = 2 \times 10^4$  N/m<sup>2</sup>, and  $R = 10 \mu\text{m}$ .  $E_s^0$  is the surface energy the drop would have if it were strictly spherical.



the anisotropy of the surface tension on the interface between the SLC *C* and the isotropic matrix (glycerin with lecithin added).

The SLC *C* droplet shape corresponding to the minimum of the surface energy may be not spherical, but have the shape of two joined spherical segments. Such droplets are the first example of an equilibrium stable shape of an anisotropic body with two unequal values of the surface tension on the outer boundary.

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