Threshold of structural hedgehog–ring transition in drops of a nematic in an alternating electric field

V. G. Bondar, O. D. Lavrentovich, and V. M. Pergamenshchik

Physics Institute, Ukrainian Academy of Sciences
(Submitted 17 July 1991)
Zh. Eksp. Teor. Fiz. **101**, 111–125 (January 1992)

A polarization microscope was used to investigate in detail the main types of restructuring in dispersed drops of a nematic, under normal boundary conditions in an external electric field. The basis of this restructuring is the loss of stability of a point topological defect (hedgehog) at the center of the drop, and its transformation into another defect (ring) on the surface of the drop. Depending on the regime, which is set by the field frequency f, two types of hedgehog–ring transformations are observed: in the conductivity regime (small f) the hedgehog can initially move from the center to the surface of the drop and only then be jumpwise transformed into a ring; in the dielectric regime (large f) the hedge is transformed into a ring without moving from drop center. It is shown experimentally and theoretically that in the latter regime the critical electric field E_c that causes the direct (hedgehog–ring) transition can have a nonmonotonic dependence on the drop diameter d. Namely, for small ($d \leqslant d_e$) drops it decreases with increase of d, and for large ones ($d \gg d_e$) it increases with d. Here d_e is the thickness of the domain wall, is connected with the strong director deformations localized near the equatorial plane of the drop, and can be roughly estimated as the electric coherent length. We show that elastic deformations connected mainly with this wall are responsible for the aforementioned singularities of E_c. The reverse (ring–hedgehog) transition is effected in two stages: as the field is decreased the ring initially breaks away from the surface of the drop at a certain intensity $E_d < E_c$, and then collapses into a point defect after reaching a critical value $E_{c,\mathrm{rev}}$ that decreases monotonically with increase of the drop diameter d.

1. INTRODUCTION

Drops of nematic liquid crystals (NLC) dispersed in isotropic (say, polymer) matrices have recently become the object of active research. They are of interest because of many nontrivial properties of the drops[1] and prospects of using materials prepared on their base for information display.[2–8] Such systems are in fact new objects in the physics and chemistry of colloids; their properties are much more complicated than those of traditional "liquid in liquid" systems, since one phase, the NLC, is anisotropic in view of equal orientation of the long molecules—along the director **n**.

An important property of NLC drops is that in the absence of external orienting fields the equilibrium state corresponds to an inhomogeneous equilibrium **n** distribution determined by the balance of the anisotropic part of the surface energy and of the elastic energy of the orientational strains in the NLC. In particular, if the matrix contributes to a normal orientation of **n** on the surface, an equilibrium distribution exists in the drop, close to radially symmetric and containing at the center a topological point defect, i.e., a hedgehog.[9]

Owing to the anisotropy of the NLC, an external electric field is capable of altering the molecule orientation and with it a number of system properties, such as the effective refractive index of the drops. It is just the latter which is used for information display (see, e.g., Refs. 2–8). Its timeliness notwithstanding, the character of the drop response to an external field has been little studied. In particular, there are few and, more importantly, quite contradictory data on the dependence of the drop-changing threshold intensity E_c on the drop diameter d. It is shown thus in Refs. 3 and 4 that $E_c \propto 1/d$. In Ref. 7, on the contrary, a nonmonotonic $E_c(d)$ dependence was observed: as E increases the drops initially transformed are small ($d < 10\,\mu$m) or large ($d > 10\,\mu$m), but the intermediate ones ($d \approx 10\,\mu$m) retain a structure close to the initial one. Finally, an exactly opposite behavior is described in Ref. 8: the $E_c(d)$ dependence is likewise nonmonotonic, but has a minimum rather than a maximum in the region of medium d ($\sim 4\,\mu$m).

The present paper is devoted to a study of the $E_c(d)$ dependence and the possible factors that can influence its character, for NLC drops having positive anisotropy $\Delta\varepsilon$ of the dielectric constant and $\Delta\sigma$ of the conductivity, and dispersed in a matrix that sets a normal orientation of the NLC molecules on the interface. The reason for the choice is that for alternative, tangential, boundary conditions the character of $E_c(d)$ depends on a random factor such as the degree of nonsphericity of the drops.[4,6] We note in this connection that the just normally oriented drops were investigated in Refs. 4 and 7, but unfortunately the boundary conditions were not identified in Ref. 8.

In a study of drops with normal molecule orientation it must be taken into account that effects both linear and quadratic in the field can contribute to their electrooptical response.[4] Among the linear, in particular, are the flexoelectric and surface-polarization effects. To simplify the experiments the drops were therefore subjected to only an alternating field of frequency higher than 1 kHz, at which there was no time for the director distortions due to the above linear mechanisms to develop.[4,10] Individual apects of drop behavior in a dc field are the subject of Refs. 4 and 11.

Our first problem was to elucidate the very character of the drop restructuring in an electric field, since all that has

 0038-5646/92/010060-08$05.00

been known by now is that the distribution of n is close to radially-symmetric in the absence of a field, while at $E > E_c$ it is axisymmetric with a disclination ring on the equator.[4,7] The second problem was to determine the $E_c(d)$ dependences and compare them with the character of restructurings as functions of the applied-field frequency.

2. EXPERIMENTAL PROCEDURE

The matrix chosen to ensure normal boundary conditions was the silicone elastomer $(CH_3)_3SiO[(CH_3)_2SiO]_nSi(CH_3)_3$ (of "Pherac BERLIN" manufacture). The dielectric constant ε_m and the electric conductivity σ_m of the matrix, measured at room temperature and at an electric-field frequency $f = 20$ kHz, were respectively $\varepsilon_m = 3$ and $\sigma_m \sim 1.5\cdot 10^{-7}\,\Omega^{-1}m^{-1}$. The investigated NLC was pentylcyanobiphenyl $CH_3(CH_2)_4(C_6H_4)_2CN$, (5CB) with positive anisotropy of the dielectric constant $\Delta\varepsilon = \varepsilon_\parallel - \varepsilon_\perp$, and electric conductivity $\Delta\sigma = \sigma_\parallel - \sigma_\perp$. The subscripts $\parallel$ and $\perp$ label components along and across the director. The measured average electric conductivity for 5CB was $\sigma \sim 3\cdot 10^{-6}\,\Omega^{-1}m^{-1}$, and the anisotropy of the dielectric constant was $\Delta\varepsilon = 14.2$ (Ref. 12). We investigated 5CB drops of diameter from 3 to 30 μm and dispersed in the elastomer.

To determine the character of the drop restructuring by the field, we used two measurement-cell geometries that enabled us to track the changes in two directions, normal and parallel to the field, i.e., in the equatorial and meridional sections of the drop.

In the first case the film with dispersion was placed between glass plates coated by a transparent conducting SnO_2 layer. The film thickness was fixed in the course of its preparation by teflon liners and ranged from 20 to 150 μm. In the second geometry, the film was placed between the glass plates without a conducting coating, and the field was applied in the horizontal plane with the aid of two rectangular electrodes. The gap between the electrodes was 100–200 μm.

A voltage of frequency from 1 to 20 kHz was applied to the cells from a GZ-56/1 low-frequency generator. The texture changes were recorded with a polarization microscope at room temperature. The director distribution in the drops was established from the locations of the extinction branches and the variation of the interference colors, including the case when a quartz wedge was inserted (see, e.g., Ref. 9).

3. RESULTS OF EXPERIMENT

As will be shown below, the character of the structural transformations in drops depends both on their diameter and on the applied-field frequency. A common property is that at a certain intensity, which we designate E_c, the point defect in the drop vanishes and is replaced by a structure with an equatorial disclination ring and a defect-free almost homogeneous distribution of the director in the bulk of the drop. The initial radial structure is restored in the drop when the external field is removed. We describe below various drop-construction patterns produced when the field intensity is changed.

3.1. Structural transformation in drops

Most information on the details of change of director orientation in drops is obtained with an experimental geometry in which meridional sections of the drops are observed. The force lines of the field **E** are oriented in this case in the horizontal plane and, in addition, are inclined 45° to the Nicol polarization directions.

In the initial state, $E = 0$, the distribution of **n** in the drop has radial symmetry (Fig. 1a) with the possible exception of a small region (~ 1 μm) near the core of the point defect at the center of the drop.[9] Such a structure is typical not only of large drops, but also of small ones 2–4 μm in diameter. Note that we were unable to observe an effect described in Ref. 7, a change of the drop equilibrium structure from radially symmetric to axisymmetric as the size of the latter decreases.

A feature of the radially symmetric distribution is that the extinction branches are arranged along the polarization directions of the Nicols, since by definition the extinction prisms are localized in those texture sections where **n** lies in the polarization plane of one of the Nicols. When E increases from 0 to E_c, the molecules in the drops become reoriented along the field, as evidenced by the crowding of the extinction branches towards the equator (Figs. 1b and 1c). The first to be reoriented are the molecules that are far from both the drop boundary and the equatorial plane. By virtue of the nonzero linkage, the orientation of **n** along the radius vectors is preserved on the surface.

Since $\mathbf{n} \perp \mathbf{E}$ on the equatorial plane for all values of $0 < E < E_c$, one can arbitrarily separate near this plane a

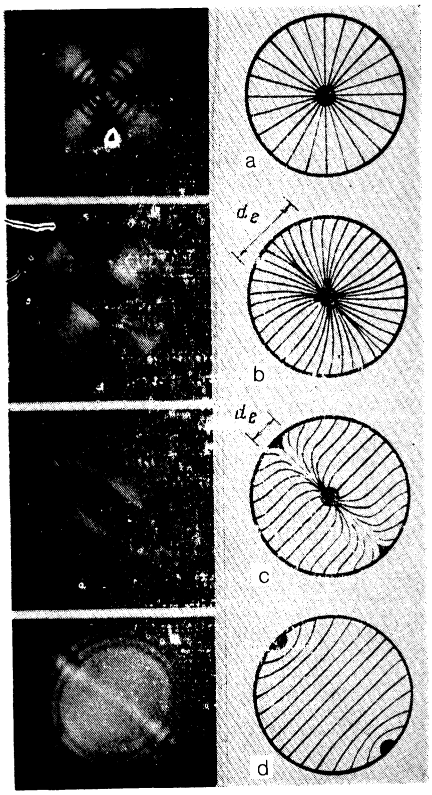

FIG. 1. Textures and meridional structures of 5CB drop ($d = 29$ μm) in an electric field hose intensity vector **E** lies in the plane of the figure and makes an angle 45° with the directions of the Nicol polarizations: (a) radial structure, $E = 0$; (b) appearance and (c) compression of a domain wall in the equatorial plane of the drop, $E > 0$; (d) axisymmetric structure, $E > E_c$.

plane-parallel layer in which the director changes orientation from its state $\mathbf{n} \perp \mathbf{E}$ at the center to the state $\mathbf{n} \| \mathbf{E}$ and the layer boundaries. This layer is a defect of the wall type. For large drops, obviously, the wall thickness d_e is determined by the competition between only two contributions to the energy, dielectric and elastic. That is to say, one can expect

$$d_e \approx 2(4\pi K/\Delta\varepsilon E^2)^{1/2},$$

where K is the mean value of the Frank elastic moduli. The $d_e \propto 1/E$ dependence for large (exceeding 8 μm) drops is qualitatively confirmed also by experiment: When E is increased the wall thickness decreases, cf. Figs. 1b and 1c. For small drops, the picture is on the whole similar, but is made more complicated by the influence of the near-surface orientation of the molecules.

A point defect in the bulk of the drop, as already indicated, is preserved all the way to values $E = E_c$. Its location, however, can depend on the frequency of the applied field. This fact, and also certain other details of the restructuring, can be well traced when the equatorial plane of the drop is observed, i.e., when **E** is oriented vertically, along the optical axis of the microscope (Fig. 2).

As seen from Fig. 2, with increase of E the straight extinction branches, corresponding to the radial structure (Fig. 2a), become bent, attesting to the onset, in the equatorial plane, of not only transverse but also longitudinal bending deformations (Fig. 2b), and in the drop as a whole—also twist deformations. Their cause is apparently the formation of a wall near the drop's equator (see Figs. 1b and 1c), the geometry of which is similar to that of the hybrid-orientation NLC layers considered in Refs. 13 and 14. Longitudinal-bending deformations in hybrid layers, as follows from analysis of the results of Ref. 13, are capable of lowering the elastic energy, and it is this which explains the twisting of the extinction branches.

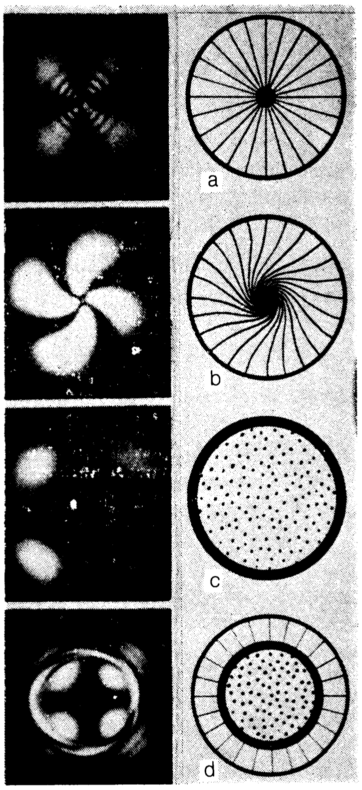

FIG. 2. Textures and equatorial structures of 5CB drop ($d = 25\ \mu$m) in an electric field with vector **E** perpendicular to the plane of the figure: (a) radial structure, $E = 0$; (b) twisting of extinction branches, $E > 0, f = 20$ kHz; (c) axisymmetric structure, $E > E_c$; (d) appearance equatorial disclination ring with decrease of field, $E < E_d < E_c$.

The onset of extinction-band twisting is accompanied by two different further drop-behavior patterns determined by the applied-field frequency.

At frequencies $f < 6$ kHz, the point defect leaves the center of the drop on its equator if $d > 8\ \mu$m (Fig. 3). In smaller drops the defect remains at the center. At frequencies $f > 6$ kHz the defect remains at the center in drops of all sizes and for arbitrary $E < E_c$ (Figs. 2a–c). The size effect is manifested in the textures only by the fact that the twist is larger the larger d.

For $E = E_c$, finally the point defect and the wall vanish near the equatorial plane of the drop, and then a disclination ring of strength $(-1/2)$ is replaced by one of strength $(+1/2)$, Figs. 1d and 2c. The behavior of the extinction branches attests to a practically complete reorientation of the molecules along the field (apart from a narrow spherical layer near the drop surface).

When the field intensity is reduced, the principal role in the structural transformations is assumed by the disclination ring, located in the case of $E > E_c$ on the equator of the drop (Fig. 2c). At a definite value of E_d, which is noticeably lower than E_c, the ring breaks away from the drop surface, but remains in its equatorial plane (Figs. 4a and 4b). The radius r can be arbitrarily varied in the range $d/4 < r_0 < d/2$ to record some arbitrary value of E. (See Figs. 4c and 4d). When the radius is decreased to $r_0 \approx d/4$, the ring becomes unstable and collapses into a point defect at the center of the drop. The structure as a whole again becomes nearly radially symmetric. Note that the field intensity $E_{c,\text{rev}}$ corresponding to reconstruction of the point defect, is substantially smaller than the value E_c causing it to vanish.

3.2. Dependences of E_c and $E_{c,\text{rev}}$ on the field frequency and on the diameter of the drops

The results of determining the $E_c(f)$ dependences for the hedgehog–ring transition and $E_{c,\text{rev}}(f)$ for the reverse transition (ring–hedgehog) at $d =$ const are shown in Fig. 5. Figure 6 shows the dependences of $E_c(d)$ and $E_{c,\text{rev}}(d)$ at a field frequency $f = 20$ kHz. It can be seen that when the frequency is varied E_c and $E_{c,\text{rev}}$ behave similarly: when the

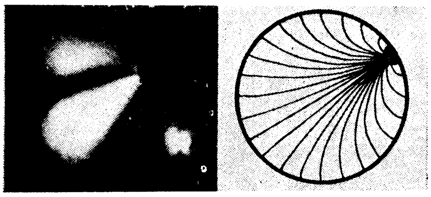

FIG. 3. Texture and equatorial structure of 5CB drop ($d = 26\ \mu$m) prior to a transition ($E < E_c$) at a field frequency $f = 2$ kHz.

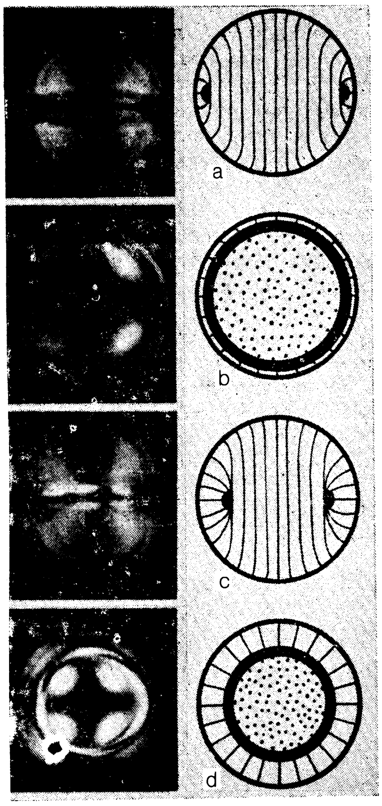

FIG. 4. Textures and structures of 5CB drops with decrease of field: (a) and (b)—detachment of ring from surface, $E = E_d$; (c) and (d)—decrease of ring radius $E < E_d$ [(a) and (c)—meridional sections through drops, (b) and (d)—equatorial].

frequency is lowered E_c increases steeply, and E_c also increases. At low frequencies one observes the usual $E_c(d)$ behavior: when the field is increased the large drops are reoriented first, and then the small ones (Fig. 5). The situation changes at $f > 6$ kHz and the $E_c(d)$ dependence becomes

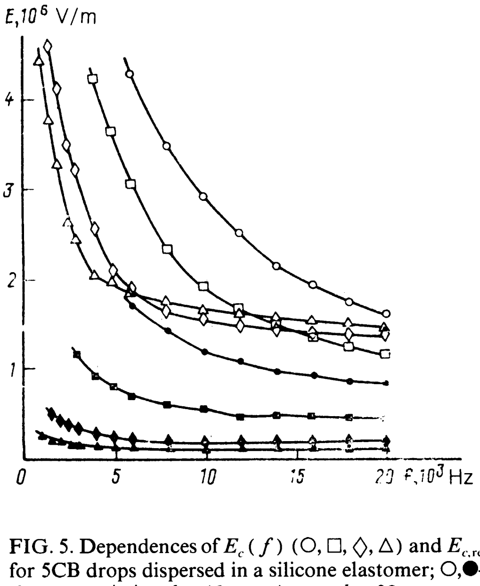

FIG. 5. Dependences of $E_c(f)$ (○, □, ◇, △) and $E_{c,\mathrm{rev}}(f)$ (●, ■, ◆, ▲) for 5CB drops dispersed in a silicone elastomer; ○,●—$d = 3\ \mu$m, □,■—$d = 8\ \mu$m, ◇,◆—$d = 19\ \mu$m, △,▲—$d = 30\ \mu$m.

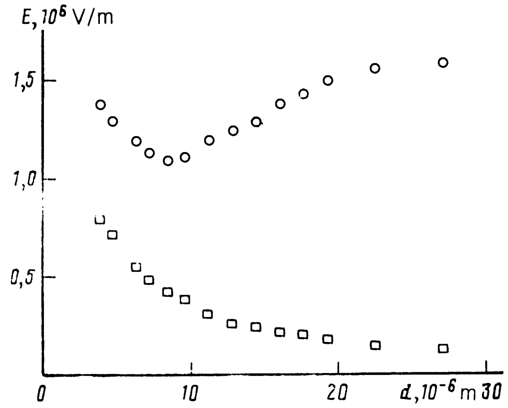

FIG. 6. Dependences of $E_c(d)$ (○) and $E_{c,\mathrm{rev}}(d)$ (□) for 5CB drops, field frequency $f = 20$ MHz.

nonmonotonic. For small drops, as before, $E_c \propto 1/d$, but for large ones ($d > 8\ \mu$m) E_c increases with d (Figs. 5 and 6). $E_{c,\mathrm{rev}}$ has no singularities whatever and decreases monotonically with increase of the drop diameter.

4. DISCUSSION OF RESULTS

In this section we attempt to examine the possible mechanism responsible for the forms of the $E_c(f,d)$ and $E_{c,\mathrm{rev}}(f,d)$ dependences, with allowance for the data on the singularities of the structural transformations in drops.

4.1. Frequency dependences of E_c and $E_{c,\mathrm{rev}}$

As seen from Fig. 5, lowering the frequency of the applied field leads to a noticeable increase of E_c under the condition $d = \mathrm{const}$. Since the material constants of the NLC and of the matrix change little in the considered frequency range from 1 to 20 kHz, it is natural to assume that the main cause of the growth of E_c can be the change of the local field E_k acting in the volume of the drop.

The value of E_k is determined not only by the intensity E_m inside the matrix, but also by the ratio of the dielectric constants of the liquid crystal (ε'_k) and the polymer (ε'_m) (see, e.g., Refs. 3 and 4):

$$E_k = E_m 3\varepsilon'_m/(2\varepsilon'_m + \varepsilon'_k), \qquad (1)$$

where ε'_k and ε'_m depend in turn on the electric conductivities σ_k and σ_m, on the dielectric constants ε_k and ε_m, and on the frequency f of the applied field,

$$\varepsilon_j' = \varepsilon_j + 2i\sigma_j/f. \qquad (2)$$

In the low-frequency limit $\varepsilon f/2\sigma \ll 1$, the value of E_k is determined mainly by the ratio of the electric conductivities σ_m/σ_k, and at high frequencies it is determined by $\varepsilon_m/\varepsilon_k$. For the systems investigated we have $\sigma_m/\sigma_k \approx 5\cdot 10^{-2}$, but $\varepsilon_m/\varepsilon_k \approx 0.3$. Consequently for a drop with constant diameter and for constant voltage applied to the cell electrodes, the value of E_k is noticeably lower at low frequencies than at high ones.

The critical frequency that determines the transition from the electric-conductivity to the dielectric regime can be estimated at $f_0 \approx 2\sigma/\varepsilon$, where σ and ε must be taken to mean the mean values for the system as a whole. As shown in Ref.

15, a good approximation in the calculation of σ and ε is the so-called logarithmic mean

$$\lg\sigma = v_m \lg\sigma_m + v_k \lg\sigma_k, \quad (3)$$

$$\lg\varepsilon = v_m \lg\varepsilon_m + v_k \lg\varepsilon_k, \quad (4)$$

where v_m and v_k are the volume fractions of the matrix and of the NLC, respectively. For the investigated mixtures we have $v_k \approx 0.3$ and we obtain as a result $f_0 \approx 3$ kHz, in good agreement with experiment. The observed $E_c(f)$ dependences at constant d (Fig. 5) can thus be qualitatively attributed to the variation of the ratios $\varepsilon'_m/\varepsilon'_k$ with frequency. Similar arguments can be advanced also for $E_{c,\mathrm{rev}}(f)$.

4.2. Dependences of E_c and $E_{c,\mathrm{rev}}$ on the drop diameter

The most unexpected of all the results shown in Figs. 5 and 6 should be taken to be the anomalous variation of $E_c(d)$ in the regions $d > 8$ μm and $f > 6$ kHz. A similar behavior of $E_c(d)$ was quite recently observed in Ref. 8, but the lack of data on the director distribution in the bulk and on the surface of the drops makes an analysis of the possible causes of the phenomena difficult.

When the behavior of the drops in an external field is considered it is necessary in general, in addition to considering the electrophysical and elastic effects, to recognize that the adhesion energy W on the NLC boundary is finite. For a large drop, with a radius exceeding the characteristic length $(K/W)^{1/2}$, the adhesion can be assumed to be strong and its contribution to the free energy can be neglected. For the boundary between 5CB and a silicone elastomer, as measured in Refs. 10 and 16, $W = 2\cdot 10^{-6}$ J/m^2 and drops with $d \geqslant 8$ μm can be regarded as large enough, i.e., the surface energy can be neglected in first approximation when the anomalous course of the $E_c(d)$ is considered.

The problem of finding $E_c(d)$ reduces thus to a determination of the balances of the elastic and dielectric moments. One must emphasize in this connection the important fact that what takes place in fact at the point $E = E_c$ is not a transition from a radial to an axisymmetric structure, as assumed until recently in a number of papers,[4,7] but a more complicated transformation with participation of a domain wall in the equatorial region of the drop. The director lines, which converge in this region to a point defect (hedgehog) at the center of the drop, are rotated from the state $\mathbf{n}\perp\mathbf{E}$ to $\mathbf{n}\|\mathbf{E}$ in a relatively narrow layer of thickness $d_e \approx 2(4\pi K/\Delta\varepsilon E^2)^{1/2}$. At the transition point itself a transformation takes place from a hedgehog into an annular disclination, inside of which the director $\mathbf{n}$ is already parallel to $\mathbf{E}$. The disclination expands, destroys the domain wall, and goes to the equator to form an axisymmetric structure. In the absence of an exact solution for the $\mathbf{n}$ distribution, we shall make, in the theoretical analysis, maximum use of the character of the restructurings considered above. The task of this analysis is to explain qualitatively the observed character of the $E_c(d)$ dependences.

Let $\xi = 2r_0/d$, where r_0 is the radius of the disclination loop into which the point defect is converted. The free energy connected with its onset, i.e., at nonzero $\xi \ll 1$, is given by

$$F = A_1\xi + A_2\xi^2 + A_3\xi^3 + \dots \quad (5)$$

We consider first a hedgehog–ring transition at the center of the drop, before the hedgehog is displaced to the surface; this corresponds to the experiment at $f > 6$ kHz (Fig. 3). Hereafter we refer to drops with $d \gg d_e$ as large and those with $d \ll d_e$ as small. It follows from experiment as well as from dimensional estimate that in the region $E \lesssim E_c$, d_e amounts to $\sim(1\text{–}5)$ μm. Let us make clear now the features of the $E_c(d)$ behavior for the two indicated drop types.

Large drops. The distributions of $\mathbf{n}$ before and after the transition are shown in Figs. 7a and 7b, respectively, in the intersection with a vertical plane. Conditionally marked on the plot is a region II of thickness $d_e/2$, in which the director is substantially deformed. Since $d \gg d_e$, the drop contains in addition to region II also a region I in which $\mathbf{n}\|\mathbf{E}$ (Fig. 7a). There is also on the periphery of the drop a region III likewise of thickness $d_e/2$; the distortions in it are due to the orienting ability of the drop surface rather than to the boundary conditions at the center as in region II. Region III will hereafter not be mentioned separately, since its contribution to F is similar to that of region II.

We examine now the changes of the free energy F that take place place in the course of the transition. At $E = E_c$ the hedgehog turns into a ring inside of which $\mathbf{n}\|\mathbf{E}$ and centered on the drop axis. Outside the ring the distribution of $\mathbf{n}$ is compressed by a factor $(1-\xi)^{-1}$, causing the term $(\partial n/\partial\rho)^2$ to be increased by $(1-\xi)^{-2}$ times, see Fig. 7b. The compression of the director lines, roughly speaking, does not influence the other terms containing no derivatives with respect to ρ. The corresponding contribution to F can therefore be estimated at

$$F_{e,l} \approx (K/8)\langle(\partial n/\partial\rho)^2\rangle(1-\xi)^{-2}\pi d^2 d_e(1-\xi^2), \quad (6)$$

where $\langle(\partial n/\partial\rho)^2\rangle$ is the average over the region II prior to the transition, and the factor $(1-\xi^2)$ is indicative of the decrease of region II when the ring is produced. The other elastic contribution to F, due to the vanishing of the hedge-

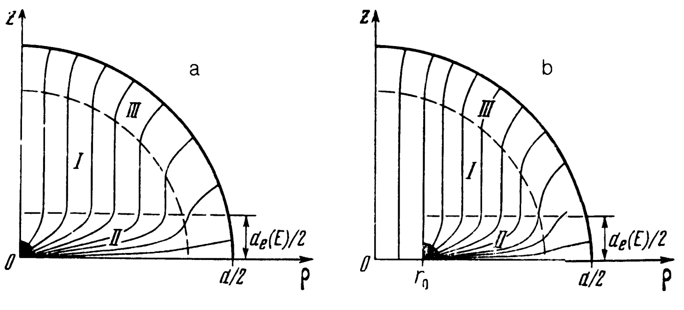

FIG. 7. Distribution of director $\mathbf{n}$ in a meridional section for large drops before (a) and during (b) the transition.

hog and its replacement by the ring, can be written in the form

$$F_{def}=p_s K d\xi\,, \tag{7}$$

where p_s is a geometric constant.[4,11]

The dielectric contribution is accumulated for large drops mainly in an equatorial region II of volume $\pi d^2 d_e/4$:

$$F_{\varepsilon,I}=(\Delta\varepsilon E^2/32\pi)\langle n_z^2\rangle \pi d^2 d_e(1-\xi^2), \tag{8}$$

where $\langle n_z^2\rangle$ is the mean value of the z-component of the director in region II.

Small drops. In this case $d \ll d_e$ and there is no region I. The diameter d determines the scale of the deformations, which are unlike (8) a volume-dependent; the dielectric contribution is therefore

$$F_{\varepsilon,I}=(\Delta\varepsilon E^2/64\pi)\langle n_z^2\rangle \pi d^3(4/3-\xi^2). \tag{9}$$

For the same reason $\langle(\partial n/\partial\rho)^2\rangle \sim 1/d^2$, and as a consequence the elastic term of type (6) does not increase in proportion to d and can be neglected compared with term (7) which is linear in d.

On the basis of the foregoing considerations we can obtain the coefficients A_i of expansion (5). We note beforehand, however, that in contrast to A_1, which contains a small number of principal terms, the coefficient A_3 can consist of a large number of terms, including some caused by rather subtle effects that make no contributions to A_1 and A_2 and have not been discussed above. For qualitative conclusions it is therefore more reliable to use A_1, and also those A_2 in which only the principal (dielectric) term is retained, and dispense entirely with A_3. If $|A_2|\xi^2 \gg A_3\xi^3$, the qualitative picture will be contained already in the expansion $F=A_1\xi+A_2\xi^2$. It must only be remembered that higher powers of ξ lead to an increase of F as $\xi\to\infty$, as shown in Fig. 8.

Grouping in (6)–(9) terms of like power we obtain the coefficients of ξ and ξ^2 in the distribution (5), in which we now retain only the decisive terms. For small drops (the condition $d \ll d_e$) we have

$$A_1=p_s K d,\quad A_2=-q_s E^2 d^3. \tag{10}$$

For large drops (the condition $d \gg d_e$) we obtain, with allowance for the form of $d_e \sim 1/E$,

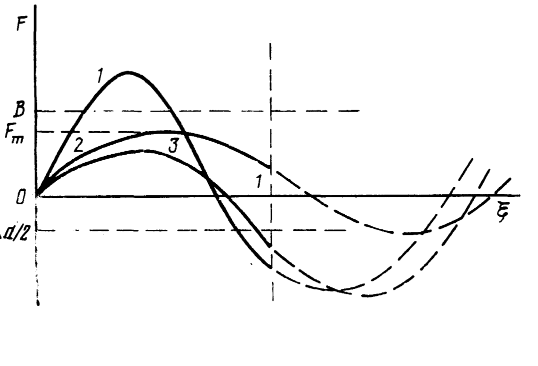

FIG. 8. Free energy $F(\xi)$ connected with formation of a disclination ring: curve 1 corresponds to large drops ($d \gg d_e$) and a field $E<E_c$; curve 2—small drops ($d \ll d_e$) and a field $E<E_c$; 3—drops of all sizes for $E>E_c$.

$$A_1=p_l K d^2 E^{-1},\quad A_2=-q_l E d^2. \tag{11}$$

The coefficients $p>0$ and $q>0$ can be easily obtained by comparing Eqs. (10) and (11) with (6)–(9).

We turn now to the critical condition for the hedgehog–ring transition. The free energy $F(\xi)$ has the form shown in Fig. 8, and $F(0)=0$ prior to the transition. At the instant of the transition the system should overcome, in a fluctuating manner, the potential barrier F_m between the states with $r_0=0$ and $r_0>0$. Next, for the transition to begin, i.e., for the fluctuation mode to set in, and also to complete the transition into an axisymmetric state, there should be produced between the latter state and the state with the hedgehog at the center a sufficient stress

$$\Delta=[-F(\xi_{\min}+F(0)]/r_0>0. \tag{12}$$

This means that on the semi-interval $(0,1]$ the function $F(\xi)$ has a sufficiently large negative minimum $F(\xi_{\min})=r_0\Delta$, which is reached at the point $\xi_{\min}$. Clearly, this point determines the radius $r_0=\xi_{\min}d/2$ of the ring after the transition: if $\xi_{\min}<1$, the ring will stop on its way to the equator at the point $\xi=\xi_{\min}$, and if the smallest value is reached on the edge of the drop, then $\xi=1$ and the ring coincides with the equator. It is in fact the last case, $\xi_{\min}=1$, which is realized in experiment, therefore

$$\Delta=[F(0)-F(1)]\,(2/d). \tag{13}$$

There are thus actually two transition conditions: 1) the presence in the $F(\xi)$ dependence of sufficiently large negative values for $0<\rho<d/2$, and 2) ability of the system to surmount a certain potential barrier, the maximum height of which we denote by B. These conditions are written analytically in the form

$$|A_2| \geqslant A_1+d\Delta/2, \tag{14}$$

$$F_m \leqslant B,\ \text{where } F_m=A_1^2/4|A_2|. \tag{15}$$

Curve 1 of Fig. 8 corresponds to $E<E_c$, since its maximum inside the drop is larger than B; curve 2 also corresponds to $E<E_c$, since it assumes its negative values outside the drop; curve 3 corresponds to $E>E_c$.

If it is assumed that B is determined by the fluctuation energy, then $B\propto V^{1/2}$, where V is the volume of the region in which **n** becomes reoriented. Since the transition of interest to us can be due to fluctuations of **n** located mainly in the equatorial plane, it follows that $V\propto d^2$, meaning $B=bd$, where $b=$ const.

With the two conditions (14) and (15) on the transition, it is obvious that E_c will be determined by the condition that is more difficult to satisfy, i.e., for large values of E. We therefore find the $E_c(d)$ dependence by assuming one of the conditions to be satisfied, finding E_c from the other, and then verify *a posteriori* the satisfaction of the first.

Let, for example, the condition (15) be satisfied for small drops at a certain d. We obtain then E_c from condition (14) and from Eq. (10):

$$E_c=(1/d)\,[(p_s K+\Delta/2)/q_s]^{1/2}, \tag{16}$$

i.e., $E_c\propto 1/d$. Then $A_1^2/4|A_2|\propto d$ and the condition (15) in the form $bd \geqslant A_1^2/4|A_2|$, being satisfied for all d, is always satisfied so long as Eqs. (10) can be used for small drops.

Thus the model, just as the experiment, leads to the conclusion that $E_c \propto 1/d$ for small drops.

The situation changes for large drops, for if the relation $E_c \propto 1/d$ holds the transition condition (15) is no longer satisfied for large drops. In fact, F_m is now determined from (11), and it can be seen that $F_m \propto d^5$. Therefore the condition (15), which takes for large d the form $bd \gg F_m \propto d^5$, is not satisfied for large d. To satisfy it we determine E_c from condition (15), and then verify the satisfaction of the second transition condition (14). From (15) and (11) we get

$$E_c = (p_i^2 K^2 d / 4 q_i b)^{1/3}. \tag{17}$$

With this $E_c(d)$ dependence, the condition (14) is satisfied all the better with increase of d:

$$|A_2| \propto d^{7/3} \gg A_1 + 2\Delta d \propto d^{5/3} + O(d).$$

From the conditions (14) and (15) for the hedgehog–ring transition it follows thus that for small ($d \ll d_e$) drops we have $E_c \propto 1/d$ and for large ones ($d \gg d_e$) we have $E_c \propto d^{1/3}$, in qualitative agreement with experiment.

We now dwell briefly on the considered situation at $f < 6$ kHz, when the hedgehog emerges to the surface of the drop before it turns into a ring (Fig. 3). The qualitative difference from the preceding picture is here the following. First, when the ring is produced the director in the drop section outside the ring is compressed along one direction of the drop, but the stresses decrease in the opposite direction. This makes, at the most, the term of (6) which is linear in ξ insignificant. Second, owing to the overlap of regions I and II in the place where the hedgehog is localized on the surface, the deformations are here three-dimensional rather than planar. Therefore F is in this case similar to the expansion (5) with coefficients (10) for small drops; this is why the $E_c(d)$ is similar in this case to the $E_c \propto 1/d$ dependence the hedge–ring transition in small drops. As for just why the hedgehog goes over to the drop surface prior to the transition, the reason is apparently that the change of the character of the regime (dielectric or resistive) when the field frequency is changed. The actual mechanism is still unclear.

The universality of the $E_{c,\mathrm{rev}} \propto 1/d$ dependence of the critical field of the reverse ring–hedgehog transition for drops of all diameters, and the character of the ring behavior when the field intensity is lower, which were described above, can also be naturally explained in the framework of the premises on which Eqs. (10)–(15) are based. It is seen from Fig. 9 that the ring lies on the equator when the field intensity is lowered from E_c to the value E_d at which the minimum of the $F(\xi)$ curve arrives from the periphery (curves 1 and 2 of Fig. 9) to the surface of the drop (curve 3); at $E_{c,\mathrm{rev}} < E < E_c$ the ring follows the position of the minimum of the $F(\xi)$ curve, and when the field lowers the potential barrier $F_{m,\mathrm{rev}}$ separating the states with $r_0 > 0$ and $r_0 = 0$ is lowered.

In the discussion of the direct hedgehog–ring transition we were interested in the $F(\xi)$ curve far from its minimum, which is located behind the drop (Fig. 8). For the reverse transition we can no longer discard the terms cubic in ξ, since the ring lies precisely at the $F(\xi)$ minimum due to the term $\sim \xi^3$, Fig. 9. The quantities $F_{m,\mathrm{rev}}$ and $F(\xi_c)$ can therefore not be determined, since there is no information on the director distribution. We can assume, however, that no

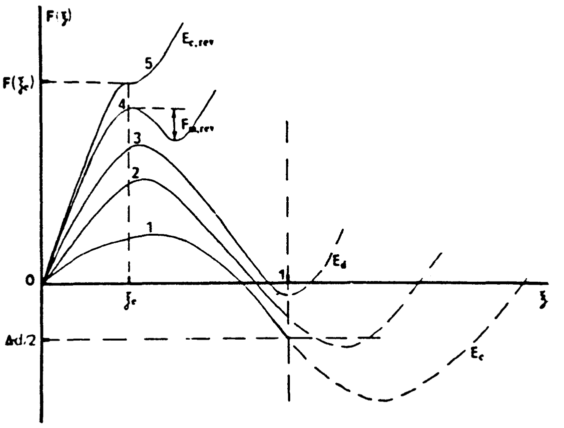

FIG. 9. Change of free energy $F(\xi)$, connected with restoration of a point defect in a drop from a ring: when the field decreases from $E = E_c$ to $E = E_d$ the position of the energy minimum arrives from the periphery (curves 1, 2) to the surface of the drop (curve 3). The ring "rests" in this case on the equator ($r_0 = d/2$). With further lowering of the field ($E_{c,\mathrm{rev}} < E < E_d$) the ring is detached from the drop surface and follows the position of the energy minimum (curve 4), $r_0 < d/2$. Finally at $E = E_{c,\mathrm{rev}}$ the ring collapses to form a hedgehog (curve 5).

strong fluctuations that throw the ring through a finite barrier can be produced in relatively weak fields $E_{c,\mathrm{rev}}$, i.e., that

$$F_{m,rev} = 0 \tag{18}$$

and the critical curve 4 on Fig. 9 has actually the form of the curve 5. In this case the criterion of the ring–hedgehog transition takes the form (18). Expanding F in powers of $(\xi_c - \xi)$ near the critical radius of the ring, we obtain

$$F_{m,rev} = A_2' (\xi_c - \xi)^2 + \ldots \tag{19}$$

(there are no linear terms, since $(dF/d\xi)_{|\xi=\xi_c} = 0$), whence $A_2' = 0$. The reverse transition in drops of arbitrary diameter is from a three-dimensionally deformed state, since the field $E_{c,\mathrm{rev}}$ is weak, $d_e \gg d$, and the director deformations are substantial in the entire volume of the drop. The elastic $A'_{2,e}$ and dielectric $A'_{2,\varepsilon}$ contributions to the coefficient A_2' have therefore the same dependence on d as for a direct transition at small d, as can be easily seen from dimensionality considerations: $A'_{2,\varepsilon} \propto E^2 d^3$, $A'_{2,e} \propto Kd$. It follows then from the condition $A_2' = 0$ that $E_{c,\mathrm{rev}} \propto 1/d$ for drops of all sizes.

5. CONCLUSION

We determined and qualitatively described the main types of restructuring in nematic drops with normal boundary conditions in an alternating field. We determined the dependences, on the frequency of the applied field and on the drop diameter, of the critical intensities E_c of the field causing a point defect in a drop to vanish, and the field $E_{c,\mathrm{rev}}$ at which this defect is restored. We have shown, first, that when the frequency is lower the fields E_c and $E_{c,\mathrm{rev}}$ increase quite strongly. This can be attributed to the change of the effective value of the field, since it is determined at low frequencies mainly by the ratio of the specific electric conductivities of the nematic and the matrix, and at high frequencies by the ratio of the dielectric constants.

The second important result is observation of a nonmonotonic $E_c(d)$ dependence: for small drops E_c decreases with increase of d, and for large one it increases. In the absence of exact solutions for the director distribution in the drops, solutions which can be obtained only with a computer, such a behavior can be understood by analyzing the geometry of the director and taking into account the nontrivial nature of the criterion for the transition. It has turned out that this behavior can be due to singularities of the structure changes in drops, namely, to formation of a defect wall of thickness $d_e \propto /E$ in the equatorial region. In large drops ($d \gg d_e$) the basic director deformations before the transition are concentrated in the wall; outside this area, $\mathbf{n} \| \mathbf{E}$ in the entire volume of the drop, with the exception of the narrow surface areas the entire volume is deformed. The difference in the transition geometry leads to a difference in the dependences of the elastic and dielectric contributions to the free energies of large and small drops, and hence to different forms of $E_c(d)$.

In conclusion, we wish to emphasize the importance of a qualitative theory that explains the main feature of the behavior of the critical field for the hedgehog–ring transition. This phenomenological theory demonstrate that these singularities cannot be determined by direct computer calculations, since such a calculation is incapable of taking into account nontrivial conditions of the transition. In fact, in computer calculations the critical condition for the transition can be defined as equality of the elastic energies of the states before and after the transition. Therefore such an effect as a smooth shift of the hedgehog from the center of the drop can be simulated with a computer, whereas a jumplike hedgehog–ring transformation connected with surmounting a finite potential barrier B cannot be simulated if the program does not contain beforehand a transition criterion of type (14) and (15). The latter cannot be obtained with a computer.

A second aspect is connected with the obvious conclusion that a nonphenomenological treatment of the hedgehog–ring transition (particularly, the determination of the quantities B, Δ, etc.) calls for going outside the framework of a description of defects with the aid of a director distribution. In fact, the fluctuations connected with surmounting the barrier between two different topological defects can be determined by the change of the internal structure of the defects. Practical importance attaches thus to the problem of studying the transformations of defects in an electric field within the framework of the Landau–de Gennes theory, which so far has seemingly been only of academic interest.[17]

[1] M. V. Kurik and O. D. Lavrentovich, Usp. Fiz. Nauk **154**, 361 (1988).
[2] J. W. Doane, N. A. Vaz, and S. Zumer, Appl. Phys. Lett. **48**, 269 (1986).
[3] P. S. Drzaic, J. Appl. Phys. **60**, No. 6, 3242 (1986).
[4] A. V. Koval'chuk, M. V. Kurik, O. D. Lavrentovich, and V. V. Sergan, Zh. Eksp. Teor. Fiz. **94**, No. 5, 350 (1988) [Sov. Phys. JETP **67**, 1065 (1988)].
[5] J. W. Doane, A. Golemne, J. L. West, J. B. Whitehead, and B.-G. Wu, Mol. Cryst. Liq. Cryst. **165**, 511 (1988).
[6] B.-G. Wu, J. H. Erdmann, and J. W. Doane, Liq. Cryst. **5**, 1453 (1989).
[7] J. H. Erdmann, S. Zumer, and J. W. Doane, Phys. Rev. Lett. **64**, 1907 (1990).
[8] H. Nomura, S. Suzuki, and Y. Atarashi, Jpn. J. Appl. Phys. **29**, No. 3, 522 (1990).
[9] O. D. Lavrentovich and E. M. Terent'ev, Zh. Eksp. Teor. Fiz. **91**, 2084 (1986) [Sov. Phys. JETP **64**, 1237 (1986)].
[10] O. D. Lavrentovich, V. M. Pergamenshchik, and V. V. Sergan, Zh. Tekh. Fiz. **60**, No 1, 208 (1990).
[11] E. D. Belotskii, A. V. Koval'chuk, O. D. Lavrentovich, B. I. Lev, and V. V. Sergan, Ukr. Fiz. Zh. **35**, 888 (1990).
[12] J. Prost and P. S. Perhan, J. Appl. Phys. **47**, 2298 (1886).
[13] O. D. Lavrentovich and Yu. A. Nastishin, Europhys. Lett. **12**. Mo. 2, 135 (1990).
[14] O. D. Lavrentovich and V. M. Pergamenshchik, Mol. Cryst. Liq. Cryst. **179**, 125 (1990).
[15] U.-M. Wang, D. D. Snyder, and G. J. Nelson, Mol. Cryst. Liq. Cryst. **149**, 163 (1987).
[16] O. D. Lavrentovich, T. Ya. Marusii, Yu. A. Reznikov, and V. V. Sergan, Zh. Tekh. Fiz. **59**, No. 10, 199 (1969) [*sic*].
[17] M. Schopohl and T. J. Sluckin, Phys. Rev. Lett. **59**, 2582 (1987).

Translated by J. G. Adashko