Biaxial potential of surface-stabilized ferroelectric liquid crystals

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A biaxial surface potential Φ_s of smectic- C^* surface-stabilized ferroelectric liquid crystals (SSFLCs) is introduced in this paper to explain the experimentally observed electric-field dependence of polarization $\tilde{P}_{cell}(E)$, in particular the shape of the static hysteresis loops. Our potential consists of three independent parts. The first nonpolar part Φ_n describes the deviation of the prime director **n** (which is the most probable orientation of the long molecular axes) from the easy alignment axis \mathbf{R} , which is located in the boundary surface plane. It is introduced in the same manner as the uniaxial Rapini potential. The second part Φ_p of the potential is a polar term associated with the presence of the polar axis in a FLC. The third part Φ_m relates to the inherent FLC biaxiality, which has not been taken into consideration previously. The Φ_m part takes into account the deviations of the secondary director *m* (which is the most probable orientation of the short molecular axes) from the normal to the boundary surface. The overall surface potential Φ_s , which is a sum of Φ_n, Φ_p , and Φ_m , allows one to model the conditions when either one, two, or three minima of the SSFLC cell free energy are realized depending on the biaxiality extent. A monodomain or polydomain structure, as well as the bistability or monostability of SSFLC cells, depends on the number of free-energy minima, as confirmed experimentally. In this paper, we analyze the biaxiality impact on the FLC alignment. We also answer the question of whether the bistable or monostable structure can be formed in an SSFLC cell. Our approach is essentially based on a consideration of the biaxial surface potential, while the uniaxial surface potential cannot adequately describe the experimental observations in the FLC.

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I. INTRODUCTION

The best known electro-optical effect in ferroelectric liquid crystals is the surface-stabilized ferroelectric liquid-crystal (SSFLC) mode [1] (which is also known as the Clark-Lagerwall effect), which was assumed originally as inherently bistable switching of the FLC director from one uniform state to the other [1,2]. However, later it became clear that, depending on the chemical structure of the FLC and its parameters, the boundary conditions, and the control voltage, SSFLC cells can also be monostable [3] or multistable [4,5], or even hysteresis-free V-shaped switching can be arranged [6–10] using SSFLCs.

There is still no unambiguous definition of the bistability and its interrelations with the hysteresis loop parameters of SSFLC cells. There are two main approaches to the interpretation of bistability, which itself is really observed in SSFLCs [1,3,4,11-14] and can be used for display and photonic devices.

On the one hand, there is a very common representation of the bistability as an effect of memorizing two light transmission states of SSFLC cells within a very limited time interval (up to several hundred milliseconds) [1,2,11-13]. This effect actually exists irrespective of the chemical structure of the FLC and the boundary conditions (except as described in [3,6-10]), and it is easily explained by the viscous torque in ferroelectric liquid crystals [15]. However, one can hardly consider this purely dynamic effect to be the inherent origin of bistability (in the thermodynamic sense) of SSFLC display cells.

On the other hand, the thermodynamically steady bi- and multistability of SSFLCs exists, with memorizing any light transmission state during the practically unlimited time (hundreds or even thousands of hours) after the driving pulse is switched off [3-5,16]. Obviously, this indicates the presence of two thermodynamically stable minima of the free energy of the SSFLC cells. We emphasize that we define the bistability precisely as the presence of two stable minima of the free energy of the SSFLC cell, while the presence of one minimum signifies monostability.

The above-mentioned thermodynamically steady effects are sorted with the shape of the static hysteresis loop of the SSFLC cell [3,4,16], or in other words, with a surface potential that includes parameters of the anchoring energy of the FLC with the bounding surface. Note that the static hysteresis loop indicates the independence of the voltage coercivity and the loop shape on the applied triangular voltage frequency f. The static loop can be obtained in experiments at f < 0.01 Hz [3,4,16] because at higher frequency the viscous torque dominates over torques caused by the surface [17].

Despite the fact that the FLC is the inherently biaxial structure, which is characterized by the biaxial order parameter [18,19], a specific form of the classical uniaxial Rapini potential [20] was postulated [17,21–25] to calculate the FLC anchoring energy potential Φ with the bounding surface.

The phenomenological expression for the uniaxial Φ potential of the SSFLC structure has been written similar to the Rapini potential in the classical manner as a linear combination of the invariants constructed from the bounding surface normal, the FLC director, and the unit polarization vector. It was done, for instance, by Eq. (7) in Ref. [17] or by Eq. (6) in Ref. [26]. However, in correspondence with the proposed Φ potential, the FLC director is not oriented along the easy alignment axis (rubbing direction) due to the nonpolar dispersive interactions, as is required by the Rapini potential physical essence. Only polar interactions caused by the high value of the spontaneous polarization P_s provide this kind of director orientation [17,21–28].

Indeed, as was proven experimentally in Ref. [29], at a small P_s value $(3-4 \text{ nC/cm}^2)$ the FLC director n is not aligned along the rubbing direction just after the cell filling, before application of an electric field. One reason for this phenomenon could be a balance between polar and nonpolar dispersive interactions.

In the framework of the approach under consideration, a steady bistability can exist only at a small spontaneous polarization, while at P_s around 100 nC/cm² or larger the SSFLC structure is driven into the monostable state [27]. This basic statement of the theoretical approach has never been confirmed experimentally. On the contrary, the SSFLC display, which provides the long- (several days) time memorization of images after the power supply is switched off, was made with the FLC spontaneous polarization around 150 nC/cm² [30].

The Φ potential suggested in Refs. [21–28] does not describe properly the shape of the static hysteresis loop measured in experiments, as was proven in Ref. [24]. As a consequence, the conditions of thermodynamically steady bistability [3–5,16], as defined above, are still not confirmed experimentally in terms of the surface potential.

Thus, there are three differences listed above between the experiments and the classical model of the uniaxial surface potential [21–23], which is applied to the inherently biaxial SS-FLC structure. At the same time, no models of biaxial surface potential have been proposed for SSFLC cells. Perhaps, the biaxiality of the surface potential, which is, strictly speaking, quite necessary because of the biaxial order parameter of the C^* phase [18,19], seemed to researchers to be unimportant in practice, since the biaxiality value (approximately 0.001–0.005) measured in optical experiments does not exceed 2% of the birefringence value [31–33]. However, the optical biaxiality and biaxiality of the surface potential are far from the same.

Nevertheless, a model of the biaxial surface potential has been proposed for bent-core smectic liquid crystals [34]. In addition, within the framework of this model, the boundary surfaces were considered to be isotropic, which means, in fact, that there is no easy alignment axis for the FLC director. The model of the surface potential for SSFLC cells, where the direction of an easy alignment axis evidently exists, should obviously be revised for SSFLCs.

The main objective of this work is to elaborate on the biaxial surface potential that could be applied efficiently for the SSFLC cells, while the validity criterion of our approach will be a quantitative comparison of the experimental and theoretical static hysteresis loops. The concept of the FLC molecular structure biaxiality extent will be introduced. It will enable one to describe quantitatively the experimentally observed dependence of the static hysteresis loop shape on the FLC molecular properties. The conditions for the emergence of SSFLC monodomain or polydomain structures depending on the biaxiality extent are also among the priorities of this work. Because of the aspects mentioned above, this paper



FIG. 1. Notations for the SSFLC smectic- C^* layer structure. MCP is the designation of a flat molecular core plane; unit vectors N, K, and R are coplanar and belong to the ZY plane; and angles β and δ also belong to the same ZY plane.

opens up additional opportunities for improvement of the FLC alignment quality.

The layout of the paper is as follows. In Sec. II we introduce our notations, describe our theoretical approach, present our biaxial surface potential, and evaluate the shape of the static hysteresis loops. Experimental details are given in Sec. III, where we describe the samples and the setup used in our measurements. In Sec. IV we analyze the correspondence between theory and experiment, summarize, and discuss the results. In Sec. V we conclude our approach.

II. THEORY AND CALCULATIONS

A. Model and notations

The proposed form of the biaxial surface potential is not strictly justified, but it is a hypothesis based on a modification of the uniaxial Rapini potential [20]. The potential involves the well-known Φ_n and a supplementary term Φ_m that accounts for a contribution of molecular biaxiality to the surface potential of the FLC. As a first step, we introduce our notations to write properly Φ_n for the SSFLC.

We denote the XY plane in Fig. 1 as the plane of the boundary surface, which is uniaxially anisotropic because of the ordinary rubbing or photoalignment techniques [30,35,36]. The easy alignment axis known as the R vector is in the XY plane. The uniaxially anisotropic boundary surface causes the FLC director n preferably along the R vector. Let us introduce angle ψ between n and Rvectors.

The position of the *n* director in a smectic- C^* layer is given by the angles θ and φ [2]. Note that the normal *N* to the smectic layers, generally speaking, has an inclination by some δ angle $(0 \le \delta \le \theta)$ with respect to the boundary surface [29] and also with respect to the *R* vector; see Fig. 1. This statement has been proven by x-ray studies [29] and is always confirmed in our electro-optical experiments. At the same time, in our model we do not take into account the so-called "chevron" structure of tilted layers [29] since chevron-free FLCs [37–39] are quite promising for applications and therefore are always used in our experiments [3,4,16,17].

The key feature of our model is an explicit consideration of the SSFLC structure biaxiality, which was never done before now in terms of the surface interactions. The biaxiality is introduced in our model in a simple form, through a concept of planar, and not cylindrically symmetric, molecular cores that are located in the molecular cores plane (MCP), which is shown in Fig. 1. Vectors n and N, in correspondence with our model, belong to the MCP. The concept of flat molecular cores came initially from the large amount of experimental evidence that the thermodynamically steady bistability of SSFLC cells, explained above, can be obtained only using FLCs composed mainly of molecules with flat molecular cores. Molecular structures of this type are given in Sec. III.

We consider a biaxial orientation of molecular cores, which can be characterized by a nonpolar vector m, which is the director of the ordered short molecular axes. This vector is perpendicular to the MCP; see Fig. 1.

The unit vector K in Fig. 1 shows a direction of easy alignment of vector m. Direction of the K vector can be, in general, inclined by angle β with respect to the normal to the solid boundary surface.

The nonzero value of angle β can originate, for example, from the surface roughness and also because of the small deviations of the real molecular cores shape from an ideal MCP. Thus, the energetically favorable orientation of the MCP (Fig. 1) tends to the plane of a solid surface. Deviation of the *m* vector from the easy alignment direction *K* is denoted as χ , while ψ is the angle between *n* and *R*.

B. The biaxial surface potential

The uniaxial approximation Φ_n of the Rapini potential [20] of the SSFLC structure in terms of the above notations can be written in the following form:

$$\Phi_n = \frac{W_n}{2} [1 - (\vec{n}, \vec{R})^2] = \frac{W_n}{2} \sin^2 \psi, \qquad (1)$$

where W_n is the anchoring energy coefficient for the *n* director with respect to the easy alignment axis **R**. Expressing $\sin^2 \psi$ in terms of trigonometric functions of angles δ , φ , and θ , we obtain

$$\Phi_n = -\frac{W_n}{2} (\sin \delta \, \cos \varphi \, \sin \theta + \cos \delta \, \cos \theta)^2.$$
 (2)

The biaxial Φ_m term is an even function of m, because the m vector is nonpolar. Therefore, it can also be written in the form of the Rapini potential:

$$\Phi_m = \frac{W_m}{2} [1 - (\vec{m}, \vec{K})^2] = \frac{W_m}{2} \sin^2 \chi, \qquad (3)$$

where W_m is the anchoring energy coefficient of the *m* director with respect to the easy alignment axis *K*, and χ is the angle between *m* and *K* (Fig. 1). Expressing $\sin^2 \chi$ via trigonometric functions of the angles δ , φ , and θ , we obtain

$$\Phi_m = -\frac{W_m}{2} \sin^2 \phi \cos^2(\delta - \beta). \tag{4}$$

When writing expressions (2) and (4), we have neglected the constant terms.

Vectors \boldsymbol{n} and \boldsymbol{m} are the eigenvectors of the tensor order parameter of a biaxial medium that is under consideration. In FLCs a polar direction exists, which is given by the unit vector \boldsymbol{p}_1 [2] that is collinear to vector \boldsymbol{m} . Therefore, in the surface potential it is necessary to take into account the polar term Φ_p that is linear in \boldsymbol{p}_1 , which can be written in the following simplest form:

$$\Phi_p = \frac{W_p}{2}(\vec{p}_1, \vec{K}) = \frac{W_p}{2}\cos\chi = \frac{W_p}{2}\sin\varphi \,\cos(\delta - \beta), \quad (5)$$

where W_p is the polar part of the binding energy of vector p_1 with the direction of easy orientation K. The value of W_p is proportional to the order parameter S_p , which characterizes the ordering of the molecular dipole moments along the polar direction p_1 . Taking into account Eqs. (3)–(5), one obtains the final expression for the surface potential:

$$\Phi_{s} = -\frac{W_{n}}{2} [(\sin \delta \, \cos \varphi \, \sin \theta + \cos \delta \, \cos \theta)^{2} + \tilde{W}_{1} \sin^{2} \varphi \cos^{2} (\delta - \beta) \pm \tilde{W}_{2} \sin \varphi \cos (\delta - \beta)], \quad (6)$$

where $\tilde{W}_1 = W_m/W_n$ and $\tilde{W}_2 = W_p/W_n$ are the dimensionless anchoring energies, and the signs + or - correspond to the upper and lower boundaries of the SSFLC cell, respectively.

The surface potential (6) is a function of two independent variables δ and φ , varying in intervals $0 \leq \delta \leq \theta$ and $-\pi/2 \leq \varphi \leq \varpi/2$, respectively. Its dimensionless profile depends on the four dimensionless parameters: \tilde{W}_1 , \tilde{W}_2 , β , and θ . Angle θ depends only on the FLC chemical structure and temperature. It can be evaluated experimentally. Three other dimensionless parameters are associated with the interaction of the liquid crystal with the boundary surface.

Potential (6) has either one, two, or three minima depending on the values of \tilde{W}_1 , \tilde{W}_2 , and β . In particular, at $\tilde{W}_2 = 0$ and $\beta = 0$ the number of minima is determined by the value of \tilde{W}_1 only; see Fig. 2. The equipotential lines in Fig. 2 represent the results of calculations for the Φ_s potential (6) at a particular value of $\theta = 30^\circ$ at $\beta = 0$, \tilde{W}_2 is equal to zero, and there are different nonzero values of \tilde{W}_1 indicated in the caption of Fig. 2.

The positions of the surface potential (6) minima correspond to points 1, 2, and 3 in Fig. 3. The corresponding orientations of director **n** and the smectic-layer structures of the SSFLC are also shown in Fig. 3. In particular, point 1 in Fig. 3 corresponds to the case of the tilted smectic layers: $\delta = \theta, \varphi = 0$, which is illustrated by the image in the center. Points 2 and 3 correspond to orthogonal smectic layers and satisfy the conditions $\delta = 0, \varphi = +\pi/2$ (top image) and $\delta = 0, \varphi = -\pi/2$ (bottom image).

We emphasize the crucial role of the biaxial $\tilde{W}_1 = W_m/W_n$ parameter that makes possible the existence of two or three minima of the Φ_s potential (6), depending on the \tilde{W}_1 magnitude. One can see further that only nonzero W_m/W_n values can explain the steady bistability and numerical calculation of the static hysteresis loop having a realistic shape.

C. Calculation of the static hysteresis loops

To calculate the static hysteresis loop, which describes the dependence of the polarization p_{cell} of the SSFLC cell on the electric-field strength E, we consider a flat layer of a FLC of thickness d, located between two surfaces. Initially, before

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FIG. 2. Lines of the surface potential level and the positions of its minima at $\theta = 30^{\circ}$ and $\beta = 0$, $\tilde{W}_2 = 0$, and $\tilde{W}_1 = 0.1$ (a), 0.24 (b), and 0.35 (c). The dark areas correspond to the minima of the surface potential.

δ, rad (c)

application of the electric field, the smectic layers are inclined by the angle $\delta = \theta$, and the director *n* is oriented along the easy alignment axis *K*, as is shown in Fig. 1 and in the inset on the right in Fig. 3. The electric field is applied in the direction of the **Z** axis. The expression for the contribution F_e to the energy bulk density that relates to the interaction of the spontaneous polarization P_s and electric field *E* can be written as follows:

$$F_e = -P_s(\vec{m}, \vec{E}) = -P_s E_z \sin\varphi \cos\delta.$$
(7)

The elastic term in the expression for the free-energy density is not taken into account in the present paper because the intralayer elasticity constants of FLCs are from three to four orders of magnitude larger than those for nematic liquid crystals [28]. Therefore, the distortion of the director field inside the smectic layers is practically not observed at small thickness liquid crystal SSFLC cells, especially if the cells are zigzag-defect-free [37–39]. Hence, the motion of the director, caused by the electric field, occurs uniformly across the bulk of the SSFLC cell.

Taking into account all the restrictions and assumptions mentioned above, the free energy Φ per unit area of the liquid-crystal layer located between the two flat surfaces can be written in the following form:

$$\Phi = 2\Phi_s + F_e d. \tag{8}$$

Here the multiplier 2 takes into account the presence of two symmetric surfaces, while Φ_s and F_e are given by Eqs. (6) and (7), respectively. Under symmetric boundary conditions, the polar term with the $\tilde{W}_2 = W_p/W_n$ coefficient in Φ_s vanishes. For further calculations, it is convenient to write Eq. (8) in



FIG. 3. Positions of the surface potential minima (points 1, 2, and 3) in $\delta - \varphi$ coordinates at $\beta = 0$ and $\tilde{W}_2 = 0$. The insets show the positions of director **n** within the smectic cone, the molecular core plane, and the normal **N** to the smectic layers of the SSFLC cell, corresponding to the minima of the surface potential (6).

dimensionless form:

$$\Phi = \Phi/W_n$$

= -[(\sin \delta \cos \varphi \sin \theta + \cos \delta \cos \theta)^2
+ \tilde{W}_1 \sin^2 \varphi \cos^2 (\delta - \beta) + \tilde{E} \cos \delta \sin \varphi], (9)

where $\tilde{E} = E_z/E_c$ is the normalized electric field, and $E_c = W_n/P_s d$ is a characteristic field of the task.

The next task is to calculate the static hysteresis loop of the SSFLC cell. The gradient method was used as a first step to calculate the position of the $\tilde{\Phi}$ potential minima. The positions of its minima are characterized by the calculated values of δ^* and φ^* , which, on the other hand, depend on the normalized electric field \tilde{E} .

As a second step, a normalized polarization $\tilde{P}_{cell} = P_{cell}/P_s$ of the SSFLC cell was calculated:

$$\tilde{P}_{\text{cell}} = -\sin\varphi^*\cos\delta^*. \tag{10}$$

The gradient method starts from point 1 in Fig. 3, where the values φ^* and δ^* correspond to 0 and θ , respectively. Variation of the electric field leads to a change in the position of the minimum, and thus to a change in the cell polarization (10).

The $\tilde{P}_{cell}(\tilde{E})$ dependencies calculated in correspondence with (10) are shown in Fig. 4. Note that if the \tilde{W}_1 value is small enough, then the hysteresis does not exist at all [Fig. 4(a)] because of the absence of the potential minima at points 2 and 3 (Fig. 3).

Figure 4(b) shows the dependence $\tilde{P}_{cell}(\tilde{E})$ at $\tilde{W}_1 = 0.22$. Here we observe a double hysteresis $\tilde{P}_{cell}(\tilde{E})$ loop, which is typical for the antiferroelectric liquid crystals [40], although



FIG. 4. The calculated $\tilde{P}_{cell}(\tilde{E})$ dependencies at $\theta = 30^\circ$, $\beta = 15^\circ$, $\tilde{W}_2 = 0$, and \tilde{W}_1 values are 0.1 (a), 0.22 (b), 0.245 (c), and 0.25 (d).

the shape change of the loop as compared with Fig. 4(a) is related only to the change of \tilde{W}_1 . There are no minima of the surface potential at points 2 and 3 (Fig. 3) for a given \tilde{W}_1 value, and therefore thermodynamically stable (static) bistability does not exist in the framework of this approach. However, at a particular value of electric field \tilde{E}_1 , a new minimum of the free energy corresponding to point 1' in Fig. 4(b) arises, which is separated from the minimum at point 1 by a potential barrier. An increase in the electric field from value \tilde{E}_1 to value \tilde{E}_2 leads to an increase of \tilde{P}_{cell} along the $1 \rightarrow 2$ line. At a value \tilde{E}_2 of the electric field, the minimum of the free energy corresponding to point 2 disappears and the system jumps to state 2' [Fig. 4(b)]. A reduction in the electric field from value \tilde{E}_2 to value \tilde{E}_1 leads to a decrease of \tilde{P}_{cell} along the $2' \rightarrow 1'$ line. At \tilde{E}_1 , the free-energy minimum corresponding to point 1' disappears, and the system jumps to state 1 abruptly. The above considerations are a possible physical interpretation of the existence of a hysteresis loop of an antiferroelectric type (without any antiferroelectric packing of molecules, just because of the biaxiality effect) of SSFLC cells within the framework of the proposed approach.

At a further increase of biaxiality, for example at $\tilde{W}_1 = 0.245$, a hysteresis loop of the ferroelectric type emerges. In this case, a thermodynamically stable bistability arises in the sense that after switching off the electric field, depending on

the prehistory, one of the two states, either $+\tilde{P}_{cell}$ or $-\tilde{P}_{cell}$, is memorized in the absence of an electric field [Fig. 4(c)].

At $\tilde{W}_1 = 0.25$, the calculated hysteresis loop looks like the classical one for solid ferroelectrics, and it is characterized particularly by the presence of an internal branch [Fig. 4(d)]. The internal branch of the loop corresponds to the transition from point 1 to point 2 or 3 (Fig. 3) depending on the applied electric-field sign. This branch disappears when $\tilde{P}_{cell}(\tilde{E})$ reaches saturation.

It follows from calculations that the internal branch corresponds to the reorientation of smectic layers from the oblique $(\delta = \theta)$ to the "bookshelf" structure [1,2] when the smectic layers became perpendicular to the boundary surfaces ($\delta = 0$). This behavior was also confirmed in our experiments.

In Fig. 4 it is also shown that at certain values of the electricfield strength, the polarization values exhibit a discontinuity. That is due to the loss of stability of a certain equilibrium state and the transition of the SSFLC to a new equilibrium state.

On the contrary, the experiments in Refs. [3,4,16,24] show that $P_{cell}(E)$ dependencies are always continuous. This could be due to the fact that within each minimum some dispersions of values β and \tilde{W}_1 could be observed. One of the evident reasons for these dispersions is a spatial nonuniformity of the boundary surfaces or thermal fluctuations. Therefore, the more realistic $P_{cell}(E)$ dependencies are obtained when calculating the hysteresis loop if one considers an average over W_1 and β .

Let us approximate these dispersions by the following Gaussian functions:

$$f(\beta) = \frac{1}{\sqrt{2\pi}\sigma_{\beta}} \exp\left(-\frac{(\beta - \beta_0)^2}{2\sigma_{\beta}^2}\right),\tag{11}$$

$$f(\tilde{W}) = \frac{1}{\sqrt{2\pi}\sigma_{\tilde{W}}} \exp\left(-\frac{\left(\tilde{W} - \tilde{W}_0\right)^2}{2\sigma_{\tilde{W}}^2}\right).$$
 (12)

In Eqs. (11) and (12), the parameters σ_{β} and $\sigma_{\tilde{W}}$ are the dispersions, while parameters β_0 and \tilde{W}_0 are the mean values of β and \tilde{W} , respectively. The calculation of the meanvalue of cell polarization $\langle \tilde{P}_{cell}(E) \rangle$ was carried out numerically using the standard weight average:

$$\langle \tilde{P}_{\text{cell}}(E) \rangle = \int_{-\infty}^{\infty} f(\beta) f(\tilde{W}) \cdot \tilde{P}_{\text{cell}}(E;\beta,\tilde{W}) d\beta \ d\tilde{W}.$$
(13)

This approach gives a continuous $\langle \tilde{P}_{cell}(E) \rangle$ dependence that provides a rather good approximation of the experimental results, as is demonstrated below in the experimental part of the work.

III. EXPERIMENT

A. FLC mixtures

The SSFLC mode arises if the FLC layer is confined between the two substrates, and its thickness is less than the critical thickness d_c of the helix unwinding. In our experiments, we used the cells with FLC layer with a thickness varying from 1 to 20 μ m. We have developed the FLC mixtures with a large helix pitch p_0 (greater than 5 μ m) to ensure the conditions required for the existence of SSFLCs at any thickness d of the liquid-crystal layer, taking into account that $d_c \propto p_0^2$ [41]. The nonhelicoidal FLC obtained by mixing two different smectic-C^{*} compounds with opposite signs and different magnitudes of the spontaneous polarization as well as with opposite-handed helices [42] is a starting point of this approach.

Enhancing the above-mentioned approach for the development of FLC mixtures with high spontaneous polarization (not less than 100 nC/cm²) and large helix pitch, we used the two different chiral compounds with opposite-handed helices (opposite signs of the helix wave numbers $q_o = 2\pi/p_0$) but with the same sign of the spontaneous polarization. This unique pair of chiral compounds, which are chiral derivatives of terphenyl dicarboxylic acid (CDTA), were described earlier in Refs. [43,44], and they are shown here in Table I.

When creating the FLC mixtures, we have also taken into account the assumption that the concentration of CDTA in a multicomponent FLC mixture can strongly influence the biaxial parameter \tilde{W}_1 . The point is that the CDTA molecular cores (see the chemical structures in Table I) can be considered as rather flat and rigid because *p*-terphenyl belongs either to the C_{2h} or the C_2 group of symmetry, or to an average planar D_{2h} molecular symmetry, which is determined by additional conditions [45,46]. Moreover, these flat and rigid cores at a sufficiently high concentration in mixtures provide the formation of a zigzag-defect-free alignment in the same manner as chiral naphthalene derivatives [38]. Two types of TABLE I. Chemical structure of chiral derivatives of terphenyldicarboxylic acid (CDTA), their signs of P_s , and the helix wave number q_o .



FLC mixtures containing different concentrations of CDTA were used in the experiments.

The FLC mixture of the first type is a ferroelectric smectic- C^* liquid crystal obtained by mixing a nematic liquid crystal 2–(4'-octyloxyphenyl)–5-hexyl-pyrimidine:

$$C_6H_{13} \rightarrow OC_8H_{17}$$

with the chiral nonmesogenic diester of 2-heptanol and terphenyl dicarboxylic acid, which is shown in Table I as chiral compound no. 1 or CDTA-1. This FLC mixture, denoted as NFLC-1 and described in detail in Ref. [47], contains 19 mol% of the chiral compound CDTA-1 (Table I) and 81 mol% of the nematic liquid crystal mentioned above. Note that the smectic *C*^{*} obtained in this way can flow, allowing one to restore the ferroelectric liquid-crystal layer structure in the electro-optical cells after an action of mechanical stress, as happens with the cells filled with pure nematics. The phasetransition sequence of NFLC-1 is $Cr \rightarrow 15^{\circ}C \rightarrow C^* \rightarrow 38^{\circ}C \rightarrow$ $A^* \rightarrow 5^{9.5^{\circ}C} \rightarrow Is$, while $P_s \cong 20 \text{ nC/cm}^2$, $\theta \cong 14^{\circ}$, and $p_0 \cong$ $5 \,\mu\text{m}$ at $T = 23^{\circ}\text{C}$.

The FLC mixture of the second type is a ferroelectric smectic- C^* liquid crystal obtained by mixing the two chiral derivatives of terphenyl dicarboxylic acid shown in Table I with an achiral smectic-C host matrix. The host matrix is composed of two achiral smectic-C compounds, which are shown in Table II and are marked with the numbers 1 and 2.

The nonhelicoidal FLC was created in two stages. First, a three-component eutectic mixture was developed, which is an intermediate FLC mixture (IMFLC) that includes the host matrix and the chiral nonmesogenic dopant CDTA-1, as can be seen from Table II. The phase-transition sequence of the mixture is $Cr \rightarrow {}^{21}{}^{\circ}C \rightarrow C^* \rightarrow {}^{60}{}^{\circ}C \rightarrow A^* \rightarrow {}^{74}{}^{\circ}C \rightarrow Is$, while $P_s = 75 \text{ nC/cm}^2$ and $p_0 = 350 \text{ nm at } T = 23 {}^{\circ}C$.

Further, the CDTA-2 compound (Table I) was added to the IMFLC mixture (Table II) in order to compensate for the helix pitch and to enlarge the spontaneous polarization. The result is shown in Fig. 5.

It is clear from Fig. 5 that $q_o \rightarrow 0$ when the concentration of CDTA-2 is between 12 and 16 mol%. Therefore, one of the mixtures corresponding to the vertical dashed line

TABLE II. Chemical structure of an intermediate eutectic FLC mixture (IMFLC).

Ұ	Chiral compound	Molar percent
1	$C_8H_{17} \xrightarrow{N} OC_{10}H_{21}$	50
2	C ₈ H ₁₇ O-	25
3	$\begin{array}{c} CH_{3} \\ (S) \\ (S) \\ C_{6}H_{13} \end{array} \xrightarrow{O} \begin{array}{c} CH_{3} \\ O \\ C_{6}H_{13} \end{array} \xrightarrow{O} \begin{array}{c} CH_{3} \\ O \\ C_{6}H_{13} \end{array}$	25
	(CDTA-1)	

in Fig. 5 was selected for experiments as the FLC mixture of the second type, which means a nonhelicoidal FLC with high spontaneous polarization $P_s = 120 \text{ nC/cm}^2$ and $\theta \cong 30^\circ$ at 23 °C. Ferroelectric liquid crystals with high spontaneous polarization (around 100 nC/cm²) form the states with uniform polarization, and some explanations are presented in Refs. [26,34,48]. Any splayed states and zigzag defects do not exist in this case. This basic statement is also valid for the developed FLC mixture.

The phase-transition sequence of this FLC (denoted as FLC-224) during heating from the solid crystalline phase is $Cr \rightarrow {}^{18.5}{}^{\circ}C \rightarrow C^* \rightarrow {}^{63.3}{}^{\circ}C \rightarrow A^* \rightarrow {}^{79}{}^{\circ}C \rightarrow Is$, while the crystallization at cooling from the smectic- C^* phase occurs at around 5–10 °C. The FLC-224 mixture contains around 34 mol% of CDTA compounds with flat and rigid cores, and NFLC-1 contains 19 mol% of the CDTA-1. The molecular origin of the biaxial anchoring \tilde{W}_1 could be due to the flat and rigid CDTA molecular cores. The value of \tilde{W}_1 depends on the CDTA concentration in mixtures. Hence, a comparative



FIG. 5. The helix wave number $q_o = 2\pi/p_0$ (open circles) and the spontaneous polarization (balls) of IMFLC mixtures with CDTA-2 vs CDTA-2 concentration. Measurements were carried out at 23 °C. The helix pitch p_0 was measured in 20 μ m vertically aligned cells.

experimental analysis of the SSFLC cells filled with FLC-224 and NFLC-1 should be done.

B. Techniques used for the preparation and investigations of FLC cells

The SSFLC cells with homogeneous planar alignment were fabricated using the rubbed polyimide PMDA-ODA [36,49] as the alignment layers that cover indium tin oxide (ITO) layers deposited onto the glass substrates. An island structure of alignment layers was observed using the AFM technique in the same manner as was done in [3,16,49,50] to avoid bistability suppression due to a depolarization field [51,52] that becomes crucial if the alignment layers are continuous dielectrics, while the FLC spontaneous polarization is high enough. Such discontinuous alignment layers provide conductivity of the electronic kind prohibiting charge accumulation between the liquid crystal and polyimide, but, at the same time, a good alignment quality remains [3,16,49,50]. The islandlike structure is a result of a polyimide solution flow at spin-coating of an ITO rough surface: the polyimide areas with a thickness of about 10 nm fill the valleys between the ITO peaks, as was proven with AFM Solver NT-MDT and electronic microscope Quanta 600-F [3,49].

The homeotropic or vertical alignment of the FLC helix axis in cells with 20 μ m FLC layer thickness was obtained by spin-coating a thin layer of chromo stearoyl chloride onto the glass substrates, as was described earlier in Ref. [53]. This kind of cell was used only for measurements of the helix pitch using the methods described previously in detail in Refs. [54,55].

The 1, 5, 16, and 20 μ m cell gaps of the assembled experimental cells were created using glassy spacers and then were fixed by epoxy glue. The cells were filled with the FLCs in isotropic phase by capillary action.

The phase-transition temperatures were determined using differential scanning calorimetry (DSC) and polarization microscopy (POM). The POM allows one also to take microphotos of SSFLC textures. For this purpose, a polarizing microscope POLAM-P-312 was used.

Static hysteresis loops of SSFLC cells were measured at the applied triangular voltage with frequency $10^{-4}-10^{-3}$ Hz to exclude the influence of viscous torque on the voltage coercivity and on the loop shape [14,15,17]. In fact, the polarization reversal current $I_R(V_{cell})$ of the cells was measured as a function of the applied voltage V_{cell} . A low-frequency current registration circuit proposed in Ref. [49] is presented in Fig. 6.

The function generator supplies the voltage V_{FG} to a simple circuit formed by a calibrated standard resistance R_0^* in a number with SSFLC cells. Minimum noise was reached in the experiment at $R_0^* = 1 \text{ M}\Omega$. Since, at a very low frequency, the detection of one cycle implies hours of measurements, the cell is placed inside the box, controlling the thermal variation for a long time with an accuracy of about 1 °C. Agilent multimeters, driven by a computer via LABVIEW software, have been applied to this method.

We should note that generally the total current I_{Σ} passing through the SSFLC cell consists of three components:

$$I_{\Sigma} = I_D + I_{\Omega} + I_R = C \frac{dV_{\text{cell}}}{dt} + \frac{V_{\text{cell}}}{R} + S \frac{dP}{dt}, \quad (14)$$



FIG. 6. Scheme of the low-frequency detection circuit: the function generator supplies a triangular-in-time voltage at low frequency, from 0.1 to 1 mHz. The multimeters measure the standard resistance potential ($V_0 = R_0^* I_R$) and also the function generator voltage (V_{FG}). The computer drives the detection runs by means of the LABVIEW program and automatically collects the multimeters data using the GPIB interface. Then V_{cell} is determined as $V_{FG} - R_0^* I_R$.

where I_D is the capacitive current related to the high-frequency part χ_{∞} of the dielectric susceptibility, I_{Ω} is the Ohmic current of the free charge carriers, I_R is the polarization reversal current related to reorientation of the spontaneous polarization P_s vector after the sign reversal of the V_{cell} voltage, R is the cell resistance, C is the cell capacitance, P is a projection of the P_s vector on the electric-field E direction, and S is the cell area. In our experiments, the following condition was fulfilled (in the same manner as in Refs. [28,49]) due to the high degree of purification of all components of the mixtures:

$$I_R \gg I_D, I_\Omega. \tag{15}$$

Hence, the whole current measured in our experiments relates to reorientation of the P_s vector, which is restricted only by the surface interactions if the smectic layers are not deformed in an electric field. The last assumption is valid in a weak electric field and much higher than the temperature of the transition from the paraelectric to the ferroelectric C^* phase [28].

The relation between the polarization reversal current I_R and the dielectric susceptibility χ_{φ} of the SSFLC cell collective mode follows from their definitions:

$$I_R = S \frac{dP}{dt} = S \frac{dP}{dE} \frac{dE}{dt} = S \chi_{\varphi} \frac{dE}{dt}.$$
 (16)

At an applied triangular voltage, Eq. (16) transforms into the following equation [28,49]:

$$\chi_{\varphi}(E) = \frac{1}{4} \frac{I_R(E)}{SfE_0},$$
(17)

where f and E_0 are the frequency and amplitude of the field variation.

Further, from the measured $I_R(E)$ dependence, we find the P(E) dependence for the entire repolarization cycle, using the



FIG. 7. Dependence of the dimensionless polarization \tilde{P}_{cell} of a 1.5- μ m-thick SSFLC cell based on NFLC-1 on the magnitude of a dimensionless electric field \tilde{E} . Triangular pulses ($f = 5 \times 10^{-3}$ Hz) of the electric field are applied in our experiment. Red semiopen symbols correspond to the voltage change from negative to positive, and blue solid symbols correspond to the voltage change from positive to negative. The result of a theoretical calculation of the hysteresis loop at $\theta = 14^{\circ}, \beta = 5^{\circ}, \tilde{W}_1 = 0.0546$, and $\tilde{W}_2 = 0$ is shown with a solid line. The island structure of PMDA-ODA alignment layers was used to provide the planar alignment of the SSFLC cell.

following equation:

$$P(E) = \int \chi_{\varphi}(E) dE.$$
 (18)

In the end, we obtain the static hysteresis loop experimentally.

IV. RESULTS AND DISCUSSION

Figure 7 shows the measured $\tilde{P}_{cell}(\tilde{E})$ dependence in a 1.5 μ m SSFLC cell based on NFLC-1 as well as the calculated hysteresis loop. The measurements were carried out at a frequency of 5×10^{-3} Hz of the applied triangular voltage, which satisfies the condition of static measurement [3,16,49]. The experimental dependence shows a tendency toward the appearance of a hysteresis loop of the antiferroelectric kind, which was predicted theoretically in Sec. IIC [see Fig. 4(b)]. At the same time, it was reliably shown by x-ray diffraction analysis [47] that no anticlinic molecular packing in NFLC-1 is observed. Hence, we have experimental evidence that the hysteresis loop $P_{cell}(E)$ of the antiferroelectric kind can be observed in SSFLC cells simply at particular parameters of the boundary surface, as is predicted theoretically in the framework of our biaxial surface potential concept. Obviously, bistability is not observed in this case, but clearly pronounced monostability is registered in electro-optical experiments, which is clear also from the shape of the hysteresis loop in Fig. 7.

The result of theoretical calculation of the $\tilde{P}_{cell}(\tilde{E})$ hysteresis loop is shown in Fig. 7 with a solid thick line. The calculations were done in correspondence with the procedure described in Sec. IIC at $\theta = 14^{\circ}, \beta = 5^{\circ}$, $\tilde{W}_1 = 0.0546$, and $\tilde{W}_2 = 0$. The value $E_c = 1.24 \times 10^8$ V/m was calculated as the characteristic field $E_c = W_n/P_s d$ of the task. This choice



FIG. 8. Dependence of the dimensionless polarization \tilde{P}_{cell} of a 16- μ m-thick SSFLC cell based on FLC-224 on the magnitude of a dimensionless electric field \tilde{E} . Triangular pulses ($f = 5 \times 10^{-3}$ Hz) of an electric field are applied in our experiment. Open circles correspond to the voltage change from zero to positive, then from positive to negative, etc. The result of a theoretical calculation of the hysteresis loop at $\theta = 30^{\circ}$, $\beta_0 = 13.5^{\circ}$, $\sigma_{\beta} = 0.01$, $\tilde{W}_1 = 0.25$, $\sigma_{\tilde{W}} = 0.002$, and $\tilde{W}_2 = 0$ is shown with solid red balls. The island structure of PMDA-ODA alignment layers was used to provide the planar alignment of the SSFLC cell.

of approximation parameters provides the correspondence of experimental and theoretical results.

Using the values of the characteristic field E_c , spontaneous polarization, and the thickness d of a liquid-crystal layer, one can estimate the prime anchoring energy coefficient: $W_n = E_c P_s d = 38 \times 10^{-3} \text{J/m}^2$. Furthermore, using the value $\tilde{W}_1 = W_m/W_n = 0.0546$, we evaluate the biaxial anchoring energy coefficient $W_m = 2.2 \times 10^{-3} \text{ J/m}^2$.

Figure 8 shows the results of measuring the $\tilde{P}_{cell}(\tilde{E})$ dependence of a 16- μ m-thick SSFLC cell based on FLC-224 and the calculated hysteresis loop. The characteristic field at temperature 23 °C was estimated as $E_c = W_n/P_s d = 3.8 \times 10^6 \text{ V/m}$, while the measured spontaneous polarization is $P_s = 1.2 \times 10^{-3} \text{ C/m}^2$.

At an appropriate choice of approximation parameters (see the caption of Fig. 8), there is fairly good agreement in general between experimental and theoretical data.

Referring to the characteristic field E_c , the spontaneous polarization, and the thickness $d = 16 \,\mu\text{m}$ of the FLC-224 layer, one can estimate the prime anchoring energy coefficient W_n of FLC-224 with the boundary surfaces: $W_n = E_c P_s d =$ $73 \times 10^{-3} \text{ J/m}^2$. Then taking into account $\tilde{W}_1 = W_m/W_n =$ 0.25, we evaluate the biaxial anchoring energy coefficient $W_m = 18 \times 10^{-3} \text{ J/m}^2$ of FLC-224 with the boundary surfaces.

The experimental and calculated curves for the internal branch of the hysteresis loops are not identical, perhaps due to the fact that calculations assume a homogeneous orientation of vectors n and m in the initial state in the whole SSFLC cell. In the photograph presented in Fig. 9(b), one can, however, see the polydomain structure of the initial SSFLC layer texture.



FIG. 9. Textures of SSFLC cells obtained at PMDA-ODA alignment layers having the island structure. Microphotos were obtained using a POLAM-P-312 microscope, with an image size of 200 μ m × 250 μ m: (a) 1.1- μ m-thick NFLC-1 layer, $\tilde{W}_1 = 0.0546$; (b) 5.0- μ m-thick FLC-224 layer, $\tilde{W}_1 = 0.25$.

We emphasize in particular that the identical island structure of alignment layers [3,16,35,47] was achieved to provide the planar alignment of SSFLC cells based both on NFLC-1 and FLC-224, and at the same time to avoid the bistability suppression due to the depolarization field [51,52]. However, the evaluated anchoring coefficients W_n and W_m of NFLC-1 and FLC-224 with the identical boundary surfaces are completely different from each other; see Table III. Most likely, this could be explained by the differences in the molecular structure of NFLC-1 and FLC-224. The main difference between the molecular structures of NFLC-1 and FLC-224, as shown in Sec. III A, is the concentration of CDTA in these mixtures: 19 and 34 mol%, respectively (see Table III). The concentration of CDTA molecules with flat and rigid cores is considerably larger in FLC-224 compared to that in NFLC-1.

As a result, the value of biaxial anchoring coefficient W_m for FLC-224 is about one order of magnitude larger compared to the same parameter for NFLC-1, while the value of the prime anchoring coefficient W_n is different only by a factor of 2 in these two mixtures; see Table III. The shape of the hysteresis loops, as can be seen from a comparison of Figs. 7 and 8, also reflects the crucial role of biaxial anchoring.

The static hysteresis loop of FLC-224-based SSFLC cells exhibits a very pronounced voltage coercivity, which is sufficiently large for memorization of "+" and "–" $\tilde{P}_{cell}(\tilde{E})$ states after the applied voltage is switched off (Fig. 8). Note that both memorizable states correspond to the orientation of the flat molecular cores planes (MCP, Fig. 1) parallel to the plane of a solid surface (see also Fig. 3), when the angle between *n* and *R*

TABLE III. Prime W_n and biaxial W_m anchoring energy coefficients for NFLC-1 and FLC-224 with identical boundary surfaces are shown together with concentrations of chiral derivatives of terphenyl-dicarboxylic acid (CDTA) in the corresponding mixtures.

	$W_n(J/m^2)$	$W_m(J/m^2)$	Concentration of CDTA (mol%)
NFLC-1	38×10^{-3}	2.2×10^{-3}	19
FLC-224	73×10^{-3}	18×10^{-3}	34

vectors is close to θ , and the orientation of the *n*-director does not coincide with the easy alignment axis *R* in this case. Such a situation is actually observed in the experiment, but only when the values of W_m and \tilde{W}_1 are sufficiently large, as follows from the theoretical model of the biaxial surface potential proposed in Sec. II.

When the magnitudes of W_m and \tilde{W}_1 are small enough, as was considered above, the static voltage coercivity either does not exist at all [Fig. 4(a)] or is negligibly small [Figs. 4(b) and 7]. This case corresponds to the approach of a uniaxial surface potential, that is, the classical Rapini potential [20]. In this case, thermodynamic bistability with unlimited memory time does not exist, and only a dynamic hysteresis loop is observed, whose voltage coercivity $V_c(\omega, V)$ depends on the frequency ω and amplitude V of the applied voltage according to the expression given in Ref. [15]:

$$V_c = \sqrt{-2\frac{\gamma_{\varphi}dV\omega}{P_s}\log\left(tg\frac{\varphi_0}{2}\right) - \lg^2\left(tg\frac{\varphi_0}{2}\right)\frac{\gamma_{\varphi}^2d^2\omega^2}{P_s^2}}.$$
(19)

In Eq. (19), γ_{φ} is the rotational viscosity of the smectic- C^* phase [56], φ_0 is the initial magnitude of the φ angle, and d is the thickness of the SSFLC layer.

Obviously, the dynamic voltage coercivity (19), which is independent of the parameters of FLC and boundary surfaces, can provide some memory effects within a limited time frame under the action of periodically applied voltage [17]. We mention this effect here only to avoid confusion between the concepts of static and dynamic hysteresis loops, the first of which is determined only by the surface interactions, while the second is determined only by the viscous torque.

The proposed concept of the biaxial surface potential allows us to analyze not only the conditions for the existence of biand monostability of SSFLC cells but also the quality of their planar alignment, which also depends on the W_1 magnitude. In particular, if only one minimum of the surface potential exists at a small value $\tilde{W}_1 \leq 0.1$, in correspondence with the diagram in Fig. 2(a), then a perfect monodomain texture of the SSFLC cell is observed; see Fig. 9(a). On the contrary, if the three minima of the surface potential exist at $\tilde{W}_1 \cong 0.25$, in correspondence with the diagram in Fig. 2(b), then one can see a three-domain SSFLC cell texture; see Fig. 9(b). Each of the three types of domains corresponds to one of the three observed birefringence colors [57, 58], which is determined by the orientation of the optical axis of the corresponding domains. The 5- μ m-thick FLC-224 layer was chosen in this experiment in order to distinguish different domains by different birefringence colors, and these colors are seen clearly at $2 \le d \le 12 \,\mu\text{m}$ for this FLC.

After application of the electric field corresponding to saturation of the hysteresis loop of the FLC-224-based cell (Fig. 8), the polydomain structure becomes much less pronounced and it is stored for a very long time after the field is switched off (Fig. 10).

From a comparison of Figs. 9 and 10, it becomes clear that, even after application of the electric field, the optical quality of the SSFC cell corresponding to parameter $\tilde{W}_1 = 0.25$ is much worse than that of the cell corresponding to parameter $\tilde{W}_1 = 0.0546$.



FIG. 10. Textures of the 5- μ m-thick SSFLC cell based on FLC-224 obtained and stored for 3 h after application and switching off of the positive (a) and negative (b) electric pulses. Microphotos were obtained using a POLAM-P-312 polarization microscope, with an image size of 200 μ m × 250 μ m. The cell was fabricated at the PMDA-ODA alignment layers having island structure.

It is worth paying attention to the fact that in the photographs of the textures (Figs. 9 and 10) there are no zigzag defects, in contrast to the structures presented, for example, in Ref. [29]. Thus, we have no reason to discuss the so-called chevron structures of SSFLC cells. The absence of zigzag defects is also typical for de Vries smectics [59–61]. It can be assumed that the FLCs we are considering here also belong to the type of de Vries smectic, since the small parameter of smecticlayer shrinkage is proven for NFLC-1 by x-ray diffraction analysis [47]. On the other hand, the absence of zigzag defects is an experimental fact, explained within the framework of numerous approaches [37–39,61], and is not the subject of this work, while in our concept the biaxial anchoring \tilde{W}_1 parameter regulates optical quality of the SSFLC cells.

V. CONCLUSION

In spite of the fact that the proposed form of the potential is a mathematical model, it provides a description of the static hysteresis loop shapes, which we also measured experimentally. The potential makes it possible to understand the physical origin of the existence of mono- and polydomain textures, and hence to manage the alignment quality (optical quality) of the of SSFLC cells using materials-science methods. Our experimental results can be regarded as experimental confirmation of the proposed model of the surface potential.

Within the framework of the biaxial surface potential under consideration, the monostable and bistable states, as well as the quality of the alignment (or optical quality) of the SSFLC cell, depend on the value of biaxial anchoring \tilde{W}_1 . The \tilde{W}_1 value depends on the concentration of molecules with flat and rigid cores in an FLC material. The compounds with flat and rigid cores could be, for example, the chiral derivatives of terphenyl dicarboxylic acid.

When $\tilde{W}_1 \leq 0.1$, the surface potential can be considered as practically uniaxial and having almost the same properties as the well-known Rapini potential, in correspondence with the diagram in Fig. 2(a). Only the monostable state of SSFLC cells appears in this case, while their alignment quality is perfect.

If $\tilde{W}_1 \ge 0.25$, then the biaxiality of the surface potential is large. In this case, the free energy is characterized by the

presence of the three minima, as follows from the diagram in Fig. 2(b). Therefore, the three kinds of domains can exist in the SSFLC texture; see Fig. 9(b). The static hysteresis loop with the internal branch appears (Fig. 8), as in solid ferroelectrics, hence the bistability with long-time memory becomes an inherent property of the SSFLC cell.

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