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Multistability in the mixtures of smectic-C^* materials with compensated twisting power

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The effect of multistability in the mixtures of smectic-C^* materials with compensated twisting power, i.e., the existence of a large number of almost equiprobable states in the same mixture under the same conditions, is analyzed in the framework of elastic continuum theory. A simple molecular model is also considered. It is shown that multistability can follow from the bulk properties of the smectic-C^* materials with compensated twisting power, but with high spontaneous polarization. Multistability leads to the formation of ferroelectric domains, in which the director oscillates in space. The length and amplitude of this oscillation is tunable smoothly by an electric field. Theoretical results for the domain length agree completely with the experimental data. A suggestion is made as to why each domain structure is remembered without the energy consumption, when the electric field is abruptly switched off. The structural dependence on material parameters, such as the spontaneous polarization, the elastic constant, and the equilibrium wave number, is predicted.

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I. INTRODUCTION

The development of electronics capable of memorizing continuous gray-scale images is one of the main focuses in the science and technology of display devices [1–5]. For the past decade, there has been a high demand for the development of simple rewritable devices (such as electronic papers, banners, labels, etc.). There has been significant progress in these technological developments, particularly in understanding memory effects based on smectic-C^* [6–12], nematic [13–23], and cholesteric [24] materials. The fundamental problem to which the present paper is devoted is the description of the mechanisms responsible for the realization of *multiple memory* states *in the bulk* tunable by the electric field.

Memorization of multiple states switchable by the electric field was realized in ferroelectric smectic materials [25], which were obtained in [26,27] by mixing smectic-C^* compounds with opposite-handed helices, but with the same sign of spontaneous polarization. Later [5], a display prototype memorizing gray-scale images for an infinite time without energy consumption was created using these materials. However, the origin of memorization and of the multistability phenomenon has not been clearly understood to date.

In the present paper, we will consider a theoretical model describing the origin and basic properties of multistability in mixtures of smectic-C^* materials with compensated twisting power. This model, however, is only one possibility for the explanation of the experiments, and further experimentation can help to test our theory.

The paper is organized as follows. In Sec. II, the experiment revealing the multistability in smectic materials will be presented. In Sec. III, the molecular model will be considered. In Sec. IV, the elastic free energy will be generalized for the case of a mixture of smectic-C^* materials with compensated twisting power, and the structures of the multistable smectic materials will be found. In Sec. V, the results will be discussed and compared with the experimental data. Finally, conclusions will be made in Sec. VI.

II. EXPERIMENT

In [25], the two ferroelectric smectic-C^* materials, FLC-346 and FLC-497, developed in the Lebedev Physical Institute of RAS, were investigated. In both materials, the smectic-C^* compounds with opposite-handed helices, but with the same sign of spontaneous polarization, were mixed in some proportion, below and above which the conventional helical pitch diverges, while at this particular proportion (and some range of proportion around it) some ferroelectric domains (different from the helical pitches) tunable by the electric field arise. The existence of these domains in the absence of an electric field was reported earlier in [28]. Memorization of multiple states corresponding to various domain structures obtained in the electric field was reported in [25]. The alignment material was a photosensitive azodye SD-1. Using this dye, one can easily control the alignment layer thickness from 2 to 20 nm [1,2,4]. A photoalignment layer formed from SD-1 of about 10 nm thickness has been spin-coated onto ITO substrates and irradiated by linearly polarized uv light, as described in [1].

The textures of the ferroelectric domains arising in the 6-μm-thick cell of FLC-346 material placed between crossed polarizers are presented in Fig. 1 before application of an electric field (a) and after the application and removal of the field (b). The material possesses the spontaneous polarization $P_s = 1.05 \times 10^{-3}$ C/m^2 at $T = 23\,^\circ$C. The image area in both Figs. 1(a) and 1(b) is 45×45 μm^2. The micrograph presented in Fig. 1(b) was obtained 10 min after switching off the voltage across the cell. After the electric-field removal, the domain structure is remembered. The applied voltage was 250-μs alternating sign driving pulses with duty cycle 20% and amplitude ±15 V.

The textures of the ferroelectric domains arising in the 4.85-μm-thick cell of FLC-497 material after the application and removal of the electric field are presented in Fig. 2. The material possesses the spontaneous polarization

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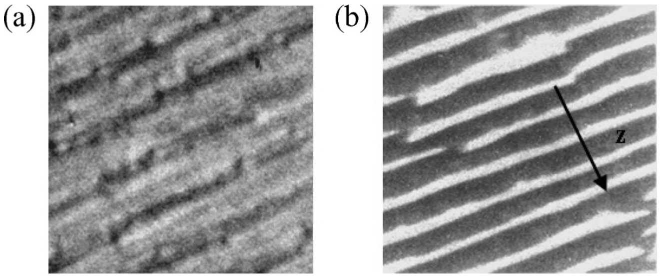

FIG. 1. Photograph of the planar cell with multistable smectic material FLC-346 placed between crossed polarizers: (a) before application of an electric field; (b) after the application and removal of the field. The image area in the photographs is 45 × 45 μm^2. Reproduced with permission from Ref. [25]. Copyright Pleiades Publishing, Inc., 2006.

$P_s = 0.95 \times 10^{-3}$ C/m^2 at $T = 23\,^\circ$C. The image area is 350 × 400 μm^2. The applied voltage was 250-μs alternating sign driving pulses with duty cycle 20% and amplitude ±3.3 V (a), ±4.7 V (b), ±6.5 V (c), and ±15 V (d).

The structures of the stripe domains in both Figs. 1 and 2 do not have a uniform director orientation. Each stripe has a gradient between bright and dark. The bright areas correspond to the maximum birefringence (when the local tilt plane is parallel to the glass substrate), while the dark regions correspond to the deviation of the local tilt plane from the orientation mentioned above. The amplitude of variation of the local tilt plane in space from the orientation parallel to the glass substrate, however, depends on the value of the electric field. The larger the value of the electric field, the smaller is the amplitude of variation. At the same time, each structure does not transform when the electric field is removed. It can only be changed by the new electric-field pulses of different voltage.

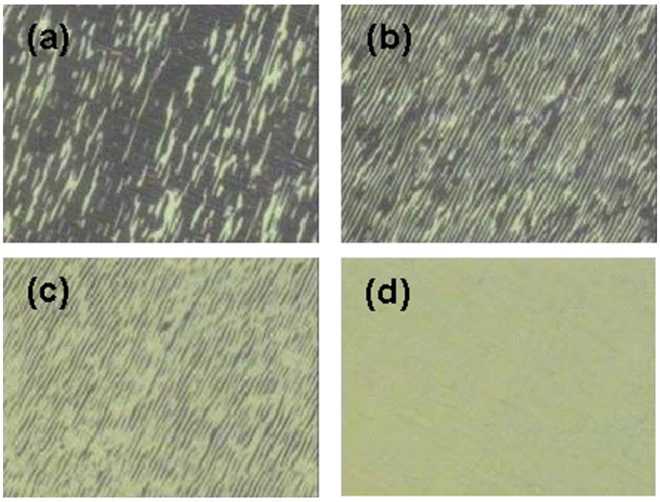

FIG. 2. Textures of multistable smectic material FLC-497 placed between crossed polarizers after application and switching off of the driving pulses with amplitudes (a) ±3.3 V, (b) ±4.7 V, (c) ±6.5 V, and (d) ±15 V. The image area in the photographs is 350 × 400 μm^2. Reproduced with permission from Ref. [25]. Copyright Pleiades Publishing, Inc., 2006.

III. MOLECULAR MODEL

In the present section, we will consider a simple molecular model showing how the compounds of the mixture used in the multistable smectic-C^* material can have opposite-handed helices but the same sign of spontaneous polarization. This model, however, could be only one of many possible models. The spontaneous polarization in each smectic layer i is known to consist of piezoelectric and flexoelectric contributions (see, for example, [29,30]):

$$\mathbf{P}_i = \hat{\boldsymbol{\chi}}\{c_p(\mathbf{n}_i\cdot\mathbf{k})[\mathbf{n}_i\times\mathbf{k}] + c_f[\mathbf{n}_i\times(\Delta\mathbf{n}_{i\pm1}\times\mathbf{n}_i)]\}, \tag{1}$$

represented by the first and second terms in Eq. (1), respectively, where $\hat{\boldsymbol{\chi}}$ is the residual dielectric susceptibility of each smectic layer (taking into account dielectric effects from the same and neighboring layers), $\mathbf{k}$ is the smectic layer normal, $\mathbf{n}_i$ is the nematic director in layer i, $\Delta\mathbf{n}_{i\pm1} \equiv \mathbf{n}_{i+1} - \mathbf{n}_{i-1}$ is the difference between director orientations in the neighboring layers, c_p is the piezoelectric constant, whose sign depends on the handedness of the chiral molecules, and finally c_f is the flexoelectric constant. The flexoelectric effect generally follows from the electrostatic interaction between molecules located in neighboring smectic layers, and in [29,30] it was shown that the lowest multipolar interaction contributing to the flexoelectric effect is the dipole-quadrupole interaction. In Fig. 3 it is schematically shown how the sign inversion of the molecular quadrupole can lead to the helix inversion in the sample.

Flexoelectric polarization in Sm-C^* arises due to the bend deformation, and it is always perpendicular to the tilt plane in each smectic layer. The handedness of the helical rotation is opposite in Figs. 3(a) and 3(b), so that the bend deformation is also in the opposite directions. However, since the charge distribution in Fig. 3(b) is inverse to that in Fig. 3(a), the flexoelectric polarization arises in the same direction in both Figs. 3(a) and 3(b). One notes that the inversion of charges presented in Figs. 3(a) and 3(b) does not reflect the principal handedness of the chiral molecules, which can be the same

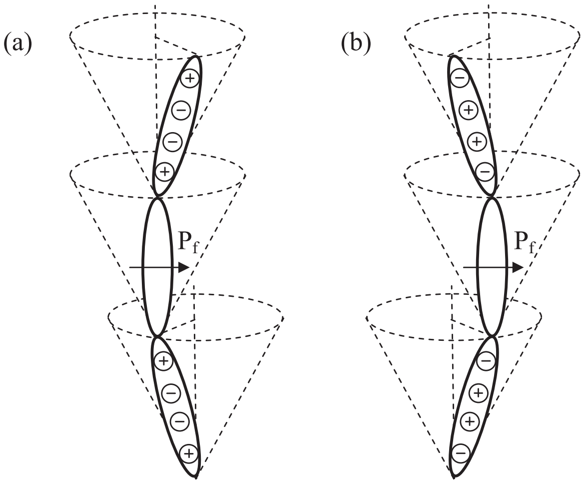

FIG. 3. Flexoelectric polarization causes opposite-handedness of the helical rotation in (a) and (b) because of the opposite signs of flexoelectric constants. Molecules are supposed to be chiral (not shown); their handedness can be the same in (a) and (b).

in both Figs. 3(a) and 3(b). Thus, both piezoelectric and flexoelectric polarizations can have the same signs in both Figs. 3(a) and 3(b), while the helical rotation should have opposite handedness.

It was shown in [31,32] and later in [29,30] that the contribution to the helical twisting power related to polarization originates only from the coupling of flexoelectric and piezoelectric polarizations (proportional to the multiple $c_f\, c_p$). Thus, it is enough to change the sign of the flexoelectric constant to obtain the opposite handedness of the helical rotation, which can be done without a change of the molecule's handedness, as demonstrated above. Here it is also important to mention that there is a contribution to the helical twisting power that does not originate from polarization effects, and which has the same origin as in chiral nematics [33,34].

IV. FREE ENERGY OF THE MULTISTABLE SMECTIC MATERIAL

Mixing the Sm-C^* compounds with opposite signs of flexoelectric constants, but the same sign of piezoelectric constants, as discussed in the preceding section, one should obviously observe the helical pitch divergence at a particular proportion, since the helix handedness is opposite in pure compounds. At this proportion, the spontaneous polarization, however, should not disappear, because it has the same sign in pure compounds. The mixture in [25] consisted of more than two compounds, and there existed (perhaps due to the surface) some range of proportion, within which the conventional helical pitch was suppressed completely, and the domains presented in Figs. 1 and 2 were observed instead. To investigate these domains, we should generalize the elastic continuum free energy for the higher gradient terms. In the general case, the elastic continuum free energy can be written in the form of expansion in Taylor series with respect to the small derivative $\varphi_z \equiv \partial\varphi/\partial z$, where φ is the azimuthal orientation of the tilt plane normal with respect to the electric-field direction $\mathbf{E}$ at point z along the smectic layer normal:

$$F = \sum_{k=0}^{\infty} a_k \varphi_z^k - P_s E \cos\varphi, \tag{2}$$

where $a_k \equiv (k!)^{-1}\partial^k F/\partial\varphi_z^k$ at $\varphi_z = 0$, and P_s is the absolute value of spontaneous polarization. In the case of cholesteric [36] or helical smectic [37–41] phases, the expansion in Eq. (2) is usually done up to the second term. Indeed, substituting $a_2 = \tilde{K}/2$, $a_1 = -\tilde{K}\tilde{q}_0$, completing the square, neglecting the constant remaining after the square is completed, and all the terms with $k > 2$, one obtains $\tilde{K}(\varphi_z - \tilde{q}_0)^2/2$ instead of the sum in Eq. (2). Here we use the tilde sign over the elastic constant $\tilde{K}$ and over the helical wave number $\tilde{q}_0$ to distinguish them from their analogs introduced for the mixtures of smectic-C^* materials with compensated twisting power below. The higher terms in expansion (2) only modulate the helix slightly.

However, because of the mixing of materials with opposite-handed helices, $\tilde{q}_0$ becomes equal to zero at a particular proportion, and, on the same grounds, all the coefficients a_k in expansion (2) with odd indices k become equal to zero. We also expect that the other remaining coefficients (with the even indices k) can vary strongly upon mixing. In the case of a mixture of smectic-C^* materials with compensated twisting power, what is modulated by the higher terms in expansion (2) is the uniform structure. The modulated uniform structure (in other words, the domain structure) should be observed in this case. Substituting $a_4 = K/2, a_2 = -Kq_0^2$, completing the square, neglecting the constant remaining after the square is completed, and all the terms with $k > 4$, one obtains instead of Eq. (2):

$$F \approx \tfrac{1}{2}K\left(\varphi_z^2 - q_0^2\right)^2 - P_s E \cos\varphi, \tag{3}$$

where we assumed that one of the coefficients, say a_2, became negative at this proportion between the components, while the next coefficient, a_4, remained positive. Indeed, if all the coefficients with even k were positive, $\varphi_z = 0$ would minimize the free energy at $E = 0$, and there would be no variation of the synclinic structure in the bulk (there would be no domains). On the contrary, if all the coefficients were negative, the corresponding smectic phase would be antiferroelectric (also without domains). At the same time, at $a_2 < 0$ and $a_4 > 0$, the new phase should arise where the optimal helical rotation angle $\Delta\varphi$ per smectic layer should be different from both zero and π, but the sign of this rotation should be undetermined. In particular, the optimal $|\Delta\varphi|$ value can be very small, and in this case the coefficient K in Eq. (3) should play the role of an elastic constant, while either $+q_0$ or $-q_0$ (without preference) should play the role of an equilibrium wave number (different from that in a conventional helical smectic) in the absence of an electric field.

What can be the origin of the director rotation without the sign preference? One notes that the coupling of spontaneous polarizations in the neighboring smectic layers is not optimal in the synclinic phase, and it is optimal in the anticlinic phase. However, at large positive coefficient a_4 (due to some other reasons, particularly dispersion interaction), the anticlinic phase cannot be realized in many materials, but the molecular transverse dipoles μ_{ef} contributing to the spontaneous polarization P_s can still be very large. In particular, in [30,35] it was shown that the largest electrostatic contribution $\mu_{\text{ef}}^4 \cos\Delta\varphi$ to the free energy follows from the second virial expansion of the dipole-dipole interaction, while expansion of $\cos\Delta\varphi$ in Taylor series contributes mostly to the second power of φ_z (with a negative sign, as it is required for the coefficient a_2 to be negative). In this case, $a_4 = K/2$ should depend weakly on μ_{ef} (and thus on P_s), and therefore from $a_2 = -Kq_0^2$ it follows that q_0 should be proportional to μ_{ef}^2 (and thus to P_s^2), which is in complete agreement with experimental data [28] for the wave number of the domain structure in the absence of an electric field. Thus, the uniform rotation of the director in an arbitrary direction matches the experimental observation at $E = 0$.

If generally the two equiprobable rotation directions exist in each smectic layer, then in total there should be 2^N almost equiprobable states in the whole sample, where N is the total number of smectic layers. However, the reversals (the smectic layers where $+\Delta\varphi$ changes to $-\Delta\varphi$ or vice versa) should not be favorable, because the reversal in layer i means the absence of rotation between layers $i-1$ and $i+1$, while the interaction between transverse dipoles in layers $i-1$ and $i+1$

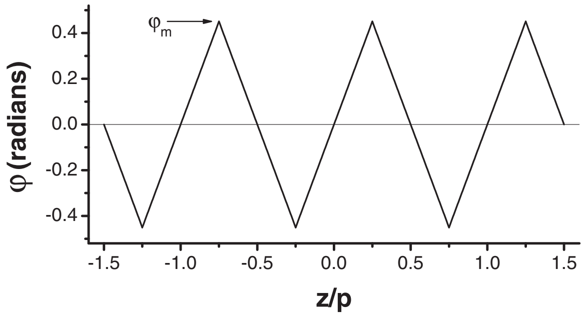

FIG. 4. Azimuthal orientation of the tilt plane normal with respect to the electric-field direction as a function of coordinate z along the smectic layer normal in the oscillating regime.

favors the director rotation between these layers in the same way as between neighboring layers. The interaction between layers $i-1$ and $i+1$ is obviously much smaller than that between the neighboring layers, but in the absence of an electric field it appears to be the only factor determining the presence or absence of reversals (except thermal fluctuations, which are also small), and therefore the rotation of the director indeed should be mostly uniform (almost without reversals) in the absence of an electric field.

After application of an electric field [compare Figs. 1(a) and 1(b)], the shape of the domains changes, and the domain length starts decreasing with increasing electric field (see Fig. 2), which is more specific to the oscillation of the tilt plane direction along the smectic layer normal than to the helical rotation. In this case, the angle between the tilt plane normal and the electric field direction varies along the smectic layer normal, as shown in Fig. 4, and one should consider a sequence of conjugated helical fragments of length $\ell = p/2$ (where p is the period of the structure) with opposite handedness (see the distribution of azimuthal angle φ along coordinate z in Fig. 4). When we define the small energy cost $\gamma \ll Kq_0^3$ of each reversal, then the free energy per each fragment can be written in the following form:

$$\mathcal{F} \equiv \gamma + \int_{-\ell/2}^{\ell/2} F(\varphi,\varphi_z^2)dz, \tag{4}$$

where $\varphi_z \equiv \partial\varphi/\partial z$, and the functional $F(\varphi,\varphi_z^2)$ is determined by Eq. (3).

V. RESULTS AND DISCUSSION

A. Equation of state

The state realized in the electric field is not uniform, as was mentioned above. The amplitude of variation of the orientation of the local tilt plane in space from parallel to the glass substrate, however, depends on the value of the electric field. Variation of functional (4), which can be done in a similar manner to, for example, that shown in Appendix A of [42], yields the following equation of state:

$$\frac{d}{dz}\left\{2\varphi_z^2\frac{\partial F}{\partial(\varphi_z^2)} - F\right\} = 0. \tag{5}$$

Applying constraint (5) to the particular functional (3), one obtains

$$\varphi_z = \pm\frac{1}{\sqrt{3}}q_0\left[1 + \frac{\tau}{k}\sqrt{1+4k^2\sin^2(\varphi/2)}\right]^{\frac{1}{2}}, \tag{6}$$

where $\tau \equiv \sqrt{3P_sE/(Kq_0^4)}$, and the parameter k must be obtained by minimization of the reduced free energy $f \equiv \mathcal{F}/\ell$. Substituting the solution (6) back into the expression (3) for the local free energy and integrating the latter within one regular fragment of length ℓ, one obtains

$$f = 2Kq_0^4\left\{\frac{G_{3/2} - G_{1/2} + \gamma^*}{G_{-1/2}} - \frac{1}{12}\frac{\tau^2}{k^2}\right\}, \tag{7}$$

where $\gamma^* \equiv \gamma/(2Kq_0^3)$ is the effective energy cost of the reversal, and the integrals G_i ($i = -1/2$, $1/2$, or $3/2$) are defined as follows:

$$G_i \equiv 3^{-i}\int_{-\varphi_m}^{\varphi_m}\left\{1 + \frac{\tau}{k}\sqrt{1+4k^2\sin^2(\varphi/2)}\right\}^i d\varphi, \tag{8}$$

where φ_m is the maximal deviation of the tilt plane orientation from the direction perpendicular to electric field. Here we assume that the structure is symmetrical with respect to this direction. In Eq. (7) we have also taken into account that the regular fragment length can be written as follows:

$$\ell = q_0^{-1}G_{-1/2}. \tag{9}$$

Minimizing the free energy f of one regular fragment [see Eq. (7)] with respect to parameters k and φ_m, one obtains the following set of equations:

$$G_{3/2} - G_{1/2} + \gamma^* = 0, \tag{10}$$

$$2 = \frac{\tau}{k}\sqrt{1+4k^2\sin^2\frac{\varphi_m}{2}}, \tag{11}$$

determining the equilibrium equation of state. The set of Eqs. (10) and (11) can be solved numerically for $k(\tau)$ and $\varphi_m(\tau)$. Consequently, the regular fragment length $\ell(\tau)$ can be calculated using Eq. (9).

B. Transition from monotonous rotation to oscillation in space

Substituting the ratio τ/k from Eq. (11) into Eq. (10) and expanding Eq. (10) in Taylor series with respect to k up to the square term, one obtains the following simple equation approximately determining $\varphi_m(\tau)$ at a small electric field:

$$\frac{3\gamma^*}{\sin\varphi_m - \varphi_m\cos\varphi_m} \approx \tau^2. \tag{12}$$

Equation (12) [and also the exact set of Eqs. (10) and (11)] has an infinite number of solutions $\varphi_m(\tau)$, and there exists some critical value $\tau_c = \sqrt{3\gamma^*}$ at which all these solutions, $\varphi_m(\tau_c) = \pi/2 + 2\pi n$, where n is an integer number from 1 to infinity, are separated from each other by 2π. From Eqs. (7), (10), and (11) it follows that the free energies of all solutions coincide at τ_c. One can check (see Fig. 5) that τ_c is a single point of intersection of $f(\tau)$ for all solutions. Above τ_c, the solution with the lowest free energy corresponds to $n = 1$, while below τ_c the free energies of solutions decrease with

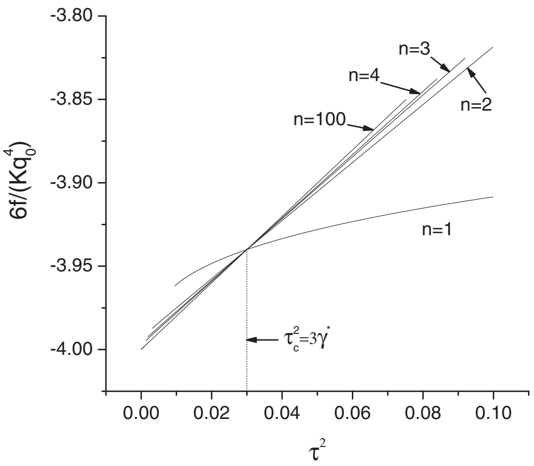

FIG. 5. Local minima free energies as functions of parameter $\tau^2 = 3\,P_sE/(Kq_0^4)$, in correspondence with Eq. (12). Here $\gamma^* = 0.01$. For each minimum n, the amplitude of oscillation φ_m belongs to the interval from $2\pi(n-1)$ to $2\pi n$.

n tending to infinity, which, in fact, means that φ_m tending to infinity satisfies the global free-energy minimum. Thus, below τ_c the monotonous rotation in an arbitrary direction corresponds to the absolute minimum free energy, while above τ_c the free-energy minimum corresponds to a conjugation of the deformed helical fragments with positive and negative derivatives φ_z (see Fig. 4). The first-order phase transition between these two states should happen at τ_c. The physics of this transition is in the competition between the reversal energy cost and the electric field energy. At large electric field, all the tilt planes tend to be perpendicular to the electric-field direction, and the number of reversals increases. On the contrary, at a small electric field the system tends to reduce the number of reversals. From Eq. (12) it follows that at $\tau > \tau_c$, the amplitude of oscillation $\varphi_m < \pi/2$ decreases and tends to zero when τ increases further. From the exact solution of Eqs. (10) and (11) corresponding to $n = 1$ it follows that either k is small (at small τ) or φ_m is small (at large τ), so that Eq. (12) approximates the solution of Eqs. (10) and (11) well even at a large electric field. In the same approximation, from Eqs. (6) and (9) it follows that

$$\varphi + \frac{1}{12}(\sin\varphi - \varphi\,\cos\varphi_m)\,\tau^2 \approx \pm q_0\,(z - z_i), \tag{13}$$

$$\ell \approx \frac{1}{q_0}\left[2\,\varphi_m + \frac{1}{6}(\sin\varphi_m - \varphi_m\,\cos\varphi_m)\,\tau^2\right], \tag{14}$$

where the values z_i correspond to the middle of each regular fragment. Equation (13) describes the azimuthal distribution of tilt planes along the smectic layer normal z, while Eq. (14) determines the length of each regular fragment.

C. Multistability and memory effects

In the present subsection, it will be shown why each state obtained in the electric field should not change when the electric field is removed. Unlike the pitch of a conventional helical smectic, at $\tau > \tau_c$ the regular fragment length in a multistable smectic material, where the director oscillates in space, decreases with increasing electric field, because $\varphi_m(\tau)$ is the decreasing function in this case [in correspondence with Eq. (12)]. From Eqs. (12) and (14), the period of oscillation of the director in space can be estimated as

$$p_{\text{osc}} = 2\ell \approx (4\,\varphi_m + \gamma^*)/q_0. \tag{15}$$

Since the period p_{osc} is approximately proportional to the amplitude φ_m, the variation of the tilt plane direction along the smectic layer normal is almost linear (the director rotation is almost uniform) within each regular fragment, as shown in Fig. 4. If the electric field is *abruptly* switched off, the reversals are not biased to some particular positions along the z axis anymore, and, in principle, they can appear in any smectic layer, as was discussed in Sec. IV. In the absence of an electric field, a huge set (2^N) of structures with uniform director rotation between each pair of neighboring reversals correspond to the local free-energy minima, and the ones with an equal total number of reversals have *exactly* the same free energies. Since the distribution of tilt planes $\varphi(z)$ within each regular fragment in the presence of an electric field is almost linear [in correspondence with Eq. (15)], it always fits some of the 2^N states corresponding to the local free-energy minima in the absence of an electric field. At the same time, different structures (with different numbers of reversals or even with the same number of reversals but different positions of the reversals) cost almost nothing in energy, but they require passing the barriers because they require the complete unwinding in some smectic layers and then rewinding in the opposite direction, and the barrier energy can be estimated as $Kq_0^4/2$ per each rewinding, in correspondence with Eq. (3). The spontaneous motion of the reversals along the z axis after the electric-field removal is not favorable, because the energy of the thermal fluctuation is obviously smaller than the barrier $Kq_0^4/2$. Therefore, the last structure obtained in the electric field is remembered after the electric-field removal. The difference between the structures obtained at different values of the electric field is in the distance between reversals, and thus in the amplitude of deviation of the tilt planes from the direction perpendicular to the electric field.

At the same time, at $\tau < \tau_c$ the regular fragment length is formally equal to infinity, and the pitch of monotonous rotation,

$$p_{\text{rot}} \approx 2\pi(1 + \tau^2/12)/q_0, \tag{16}$$

can be estimated by substituting $\varphi_m = \pi$ into Eq. (14). This regime, however, can only be realized by the application of a small electric field corresponding to $\tau < \tau_c$, but it cannot be realized by an abrupt removal of the electric field at $\tau > \tau_c$. The dependence of dimensionless period p/p_0 on the parameter $\tau^2 = 3\,P_sE/(Kq_0^4)$ in both cases ($\tau < \tau_c$ and $\tau > \tau_c$) is presented in Fig. 6, where the corresponding values of amplitude φ_m at $\tau > \tau_c$ are shown on the right scale. One notices a good correlation with experimental data. In particular, at $\tau > \tau_c$, the larger the value of the electric field, the smaller is the amplitude of variation of the tilt plane along the smectic layer normal. Each structure does not change when the electric field is removed. However, the electric field of different voltage can change the structure of the domains (the amplitude of variation of the local tilt plane and the domain length), but the

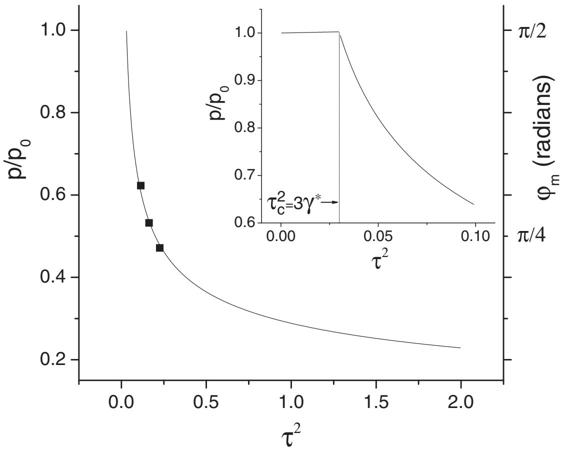

FIG. 6. Reduced period of multistable smectic structure as a function of parameter $\tau^2 = 3\,P_s E/(K q_0^4)$. Here $\gamma^* = 0.01$. The right scale represents the corresponding values of amplitude φ_m. An inset represents the smaller scale, at which the weakly first-order phase transition from the uniform rotation to the oscillation in space happens. The square bars reproduce the experimental variation of period calculated from Fig. 2 at substitution $p_0 = 28.3\ \mu$m and $K q_0^4/P_s = 17.8\ \mathrm{V}/\mu$m.

new structure will also be remembered when the electric field is switched off again.

VI. CONCLUSION

We developed a theoretical approach suggesting an explanation for the multistability observed in the mixtures of smectic-C^* materials with compensated twisting power. The elastic continuum theory was generalized for the case of a material in which the conventional helical pitch diverges, and domains arise that are different from the helical pitches. In this case, we predict that the director rotation between each neighboring smectic layer is in an arbitrary direction, where the absolute value of the local (per one smectic layer) rotation angle $|\Delta\varphi|$ is fixed in a particular material. Thus, formally, the equilibrium wave number $|q_0| = |\Delta\varphi|/\Delta z$ can be introduced. It was demonstrated that the mixtures of smectic-C^* materials with compensated twisting power, but with large spontaneous polarization, can be prepared by mixing the smectic-C^* compounds with the opposite signs of flexoelectric constants, but the same sign of piezoelectric constants, and the corresponding molecular model was suggested. A single parameter $\tau = \sqrt{3\,P_s E/(K q_0^4)}$ regulating the structure of domains was introduced, which is a combination of electric-field value E with three material parameters: spontaneous polarization P_s, elastic constant K, and equilibrium helical wave number $|q_0|$ in the absence of an electric field. We have found the distribution of tilt planes within each ferroelectric domain and the critical value τ_c, above which the ferroelectric domain length decreases with increasing electric field, exactly as in the experiment. Below τ_c, a different regime was found, in which the tilt planes rotate mostly in one (undetermined) direction. The existence of two different regimes also agrees with the experiment, as well as the quadratic dependence of the wave number on the spontaneous polarization in the absence of an electric field.

We suggest that our theoretical approach not only describes the origin and basic properties of multistability in the mixtures of smectic-C^* materials with compensated twisting power, but it also corresponds well with the experimental observations. In addition, it predicts how the electric-field dependence of the structure would change with variation of the material parameters (spontaneous polarization P_s, elastic constant K, and equilibrium wave number q_0), which can be measured or estimated from the molecular modeling.

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