

Solitons induced by alternating electric fields in surface-stabilized ferroelectric liquid crystals

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Propagation of solitary waves activated in thin ferroelectric liquid crystal cells under external, sinusoidally alternating electric fields is investigated using the electro-optic technique. It is shown that solitons give contributions only to the loss component of the response spectrum, within rather narrow ranges of frequencies and in sufficiently strong fields. The limit frequency, at which the amplitude of the velocity of the solitary waves is greatest, is found to be related to material constants of liquid crystals. Measuring this threshold frequency provides the capability to determine the elastic constant of surface stabilized liquid crystalline materials in the bookshelf or chevron layer geometries.

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Surface-stabilized ferroelectric liquid crystal (SSFLC) systems [1] have been applied in constructing both display and nondisplay electronic devices. The scale of commercialization of these systems is, however, relatively small, as a consequence of their tendency to form spontaneously structural defects [2]. In spite of the resulting technical troubles, SSFLCs are still considered as promising materials to develop innovative electro-optic switching technologies [3]. This mainly follows from the fact that field-induced switching processes between orientational bistable states occurring in SSFLCs are very fast, up to 1000 times faster than switching phenomena occurring in nematics [2]. Depending on the strength of an external electric field, the switching phenomena can proceed as a complete polarization reorientation [2,4] (when the field is strong enough), or as local reorientations, for example, as soliton excitations [5] (activated by weaker but sufficiently strong fields). Clearly, both kinds of these molecular reorientation processes are inherently nonlinear in nature. Furthermore, a threshold field amplitude, above which a periodically alternated field activates switching phenomena, depends on the field frequency.

Solitary waves in both nematic and smectic systems have usually been described taking into account an anisotropy contribution w_a to the energy density, given by $w_a = -\frac{1}{2}\varepsilon_0\Delta\varepsilon(\mathit{phn} \cdot \mathit{phE})^2$ with $\Delta\varepsilon = \varepsilon_{\parallel} - \varepsilon_{\perp}$ being the dielectric anisotropy, phn denoting the molecular director, and phE being an external electric field [6]. However, in smectic systems, especially in SSFLCs, the orientation of molecular electric dipole moments is not uniform giving rise to the appearance of the charge of density $\rho = -\nabla \cdot \mathit{phP}$, where phP is a polarization field [2]. This yields a depolarization contribution $w_d = \frac{1}{2\varepsilon_0}|\mathit{phP}|^2$ to the energy density [7]. As will be shown, orientational fluctuations of the local polarization play a more important role in the propagation of the solitary waves in SSFLCs under study here than their dielectric anisotropy. Obviously, the mere excitation of solitons is conditioned not only by the strength of applied fields, but also by a nonuniformity of molecular orientations, caused, to a large extent, by surface anchoring. Field-induced molecular reorientations can be investigated by determining the space dependence of the angle ϕ between phP and phE . In samples with the bookshelf

or chevron geometry, this dependence can approximately be determined using a one-dimensional dynamic field equation (chevron slabs can roughly be treated as dynamically independent) [2,8]. When the dielectric anisotropy is neglected and an electric field changing cosinusoidally with an angular frequency ω is applied perpendicular to sample plates, say in the x direction, this equation has the form [9]

$$K \frac{\partial^2 \phi}{\partial x^2} - \gamma \frac{\partial \phi}{\partial t} = P_S \sin \phi \left(E_0 \cos \omega t - \frac{1}{\varepsilon_0} P_S \cos \phi \right), \quad (1)$$

where K is the elastic constant describing deformations inside smectic layers, γ is the rotational viscosity, E_0 denotes the amplitude of the electric field, and P_S is the spontaneous polarization. Note that the material parameters K and γ occurring in the above motion equation both contain the factor $\sin^2 \theta$, where θ is the molecular tilt angle [2]. The second term in the right-hand side of Eq. (1) represents the torque acting on a molecule due to the existence of a depolarization field, induced by a nonuniformity of the polarization. In the case of the chevron geometry, an exact solitary wave solution to the above equation can be written as

$$\phi_{\pm}(x,t) = \arctan \frac{1}{\sinh \left(\alpha_{\pm} x + \frac{x}{\beta} - \frac{\mu}{\omega} \sin \omega t \right)}, \quad (2)$$

with α_{\pm} , β , and μ being constants (independent of space and time). The subscripts \pm refer to the lower ($-$) and upper ($+$) slabs of the chevron structure [8]. The constants α_{\pm} can be determined assuming the appropriate boundary conditions. The parameter β characterizes the kink width, while the parameter μ is related to the amplitude v_0 ($v_0 = \beta\mu$) of the velocity of kink propagation through a sample (in the direction perpendicular to cell plates). The relation of β and μ to the material constants can be derived directly from Eq. (1). Then, one obtains

$$\beta = \sqrt{\varepsilon_0 K / P_S}, \quad (3)$$

$$\mu = P_S E_0 / \gamma. \quad (4)$$

Thus, the parameters β and μ are equal, respectively, to the polar coherence length and the inverse of the time of polarization switching in ferroelectric liquid crystals [2]. According to Eq. (2), the rotational kink can move across the sample with the alternating velocity $v(t) = v_0 \cos \omega t$, provided that the field frequency is large enough (at a given

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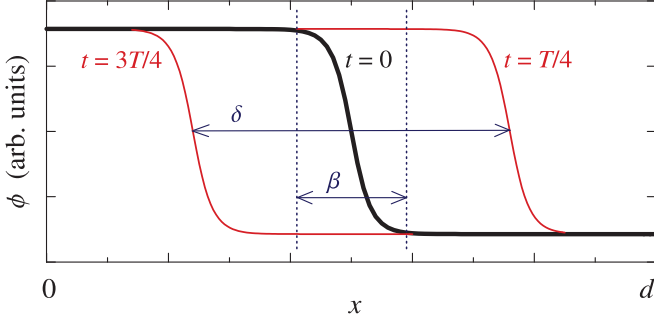


FIG. 1. (Color online) Qualitative shape of the soliton solution of Eq. (1). The thick, solid line represents $\phi(x,t)$ at $t = 0$, while thin lines refer to $\phi(x,t)$ at $t = T/4$ and $t = 3T/4$, where $T = 2\pi/\omega$ (i.e., when the kink reaches maximal and minimal positions, respectively). Additionally, the kink width β and the distance δ (over which the kink oscillates) are marked.

field amplitude). The kink motion through a smectic layer is illustrated in Fig. 1, where the distance $\delta = 2\beta\mu/\omega$, over which a kink can move during each period of the electric field, is also indicated. The minimal threshold frequency, $\omega = \omega_{\min}$ at which soliton waves appear, can be deduced from the condition that the maximal range of space oscillations of a kink is not greater than the sample thickness D or the thickness of one sample slab (in the cases of bookshelf or chevron arrangements of molecules, respectively) minus the kink width. One thus has $\delta \leq d - \beta$, where $d = D$ in the bookshelf geometry while $d = D/2$ in the chevron geometry, and hence

$$\omega_{\min} = 2\beta\mu/(d - \beta). \quad (5)$$

At a given, sufficiently large field amplitude, solitons cannot, however, be excited if field frequencies are higher than an upper threshold $\omega = \omega_{\max}$. For $\omega > \omega_{\max}$, the electric field oscillates too fast to provoke sufficiently strong noncollective molecular reorientations, necessary for the appearance of solitary kinks. Thus, solitary waves propagating perpendicular to the sample electrodes may appear at field frequencies belonging to the range $\omega_{\min} \leq \omega \leq \omega_{\max}$ with both the limit frequencies ω_{\min} and ω_{\max} being dependent on the field amplitude. In contrast to ω_{\min} , the threshold frequency ω_{\max} cannot simply be determined because that would require a knowledge of an explicit nonsolitary solution to Eq. (1).

The relation (5), together with Eqs. (3) and (4), gives the possibility to find one of the three material parameters K , γ , P_S , if two remaining constants are known. In particular, one can determine the parameter K , describing elastic properties of SSFLCs within smectic layers (the constants γ and P_S are relatively easy to measure). This is of great practical importance considering a continuing technological interest in SSFLC systems. It should be stressed that efforts to find elastic parameters have mainly been concentrated on measuring the elastic interlayer coupling in smectics with the helical structure [2,10]. The method based on the use of the relation (5) to determine the intralayer elastic coupling (much stronger than the interlayer one) will be shown to be very effective compared to the approach involving the measurement of the critical sample thickness for the helix [11]. Furthermore, the procedure under consideration here enables experimental determination

of the parameter K directly for a SSFLC sample of a particular thickness by measuring the threshold frequency ω_{\min} . This can be performed by registering response spectra $\varepsilon(\omega)$ of a liquid crystalline sample to an alternating external field and identifying a soliton contribution to these spectra. According to Eq. (2), solitons can yield the only contribution to the imaginary part of the response spectra. To prove that, consider the case of the chevron geometry [2]. Then, n th harmonic components of soliton increments of the real and imaginary parts of $\varepsilon(\omega)$ are, respectively [8], $\varepsilon'_n(\omega) \sim T^{-1} \int_0^T J(\omega, t) \cos(n\omega t) dt$ and $\varepsilon''_n(\omega) \sim T^{-1} \int_0^T J(\omega, t) \sin(n\omega t) dt$, where the voltage time period $T = 2\pi/\omega$, $J(\omega, t) = \int_{-d}^0 \Delta_-(x, t) dx + \int_0^d \Delta_+(x, t) dx$ is proportional to the total (containing all possible harmonic components) instantaneous response function, and $\Delta_{\pm}(x, t) = \cos \phi_{\pm}(x, t) - \cos \bar{\phi}_{\pm}(x, t)$ with $\bar{\phi}_{\pm}(x, t)$ being the functions $\phi_{\pm}(x, t)$ taken at $E_0 = 0$. Thus, using Eq. (2), one obtains $\varepsilon'_n(\omega) = 0$ for $n = 1, 2, \dots$, and $\varepsilon''_1(\omega) > 0$, but $\varepsilon''_n(\omega) = 0$ for $n = 2, 3, \dots$.

To illustrate the efficiency of the described procedure, measurements of the response $\varepsilon(\omega)$ of the mixture Felix 15–100 (Clariant) to a sinusoidally alternating external electric field have been performed by applying the polarizing microscope technique [12]. Note that the notation $\varepsilon(\omega)$ is used below for the resulting response spectra determined by means of the electro-optic method. Samples were prepared using typical thin glass cells (manufactured by Linkam and EHC) of thicknesses $D = 5, 12$, and $25.9 \mu\text{m}$, and with bounding plates coated with conducting semitransparent material (ITO). The plates were rubbed antiparallel to each other, providing the molecular orientation to be nearly parallel to electrodes. In consequence of strong surface interactions and relatively small cell thicknesses, the samples formed chevron smectic layer structure, which remained stable for temperatures $20 \leq T \leq 50 \text{ }^\circ\text{C}$ and for relatively high applied voltages, of amplitudes $U_0 = E_0 D \leq 40 \text{ V rms}$.

A typical Cole-Cole diagram revealing solitary waves is shown in Fig. 2 for the first harmonic components $\varepsilon'(\omega) \equiv \varepsilon'_1(\omega)$ and $\varepsilon''(\omega) \equiv \varepsilon''_1(\omega)$. The plot has been determined for a sample of thickness $D = 5 \mu\text{m}$, at $T = 27 \text{ }^\circ\text{C}$, and at ac applied voltages of the amplitude $U_0 = 20 \text{ V}$ and frequencies $f = \omega/2\pi$ ranging from 2 to 100 kHz. It is seen that $\varepsilon'(\omega)$ is constant (or nearly constant) in a frequency range. This is evidence of solitary waves traveling through both chevron slabs, although $\varepsilon'(\omega)$ is not zero for $\omega_{\min} \leq \omega \leq \omega_{\max}$. Generally, solitons can be excited only in some sample regions, while molecular motions in other sample regions can have a nonsolitary, although strongly nonlinear, character. Such a nonuniform dynamic behavior of liquid crystal samples is a consequence of an inhomogeneity of electric fields inside samples, a nonuniformity of cell plates (and thereby an inhomogeneity of surface anchoring potential), the occurrence of various types of defects in samples, and so on. Due to interlayer interactions, field-induced molecular reorientations in different system regions are not independent of each other, and mutual perturbations affect nonsolitary motions of molecules located even relatively far from borders of system regions in which solitons propagate [13]. As a result, solitary waves cause a damping of orientational motions of molecules located in regions remaining in the

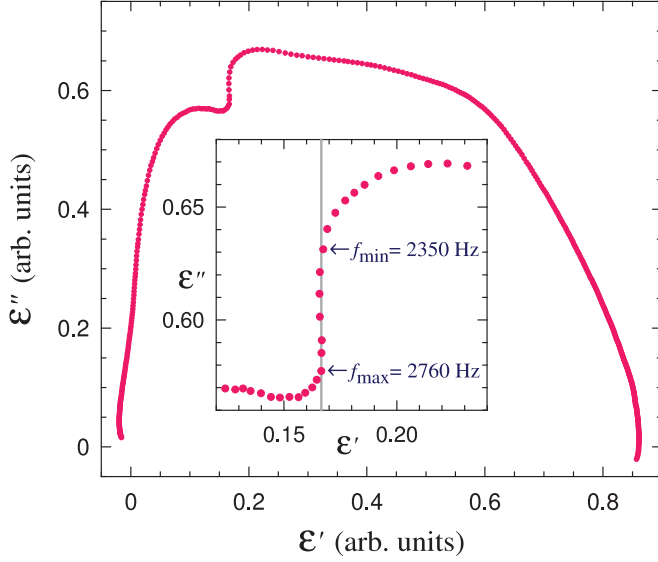


FIG. 2. (Color online) Cole-Cole diagram obtained for a sample of thickness $D = 5 \mu\text{m}$, at $T = 27^\circ\text{C}$ and $U_0 = 20 \text{V}$. Filled circles represent measurement points. The inset depicts the diagram region in which solitons manifest themselves [by a very weak frequency dependence of $\varepsilon'(\omega)$]. The limiting ordinary frequencies are given by $f_{\min} = \omega_{\min}/2\pi$ and $f_{\max} = \omega_{\max}/2\pi$.

nonsolitary dynamic regime. Accordingly, contributions to the electro-optic response coming from these sample regions are weakly dependent on the field frequency, in the range $\omega_{\min} \leq \omega \leq \omega_{\max}$. This explains the existence of a nonzero, nearly constant contribution to $\varepsilon'(\omega)$ for the frequency range in which solitary waves are excited.

The results of Fig. 2 have been obtained for the case when solitary waves simultaneously propagate through both parts (slabs) of chevroned smectic layers. However, one can expect that, in some cases, solitons can be excited only within one of the chevron slabs, as surface interactions (at sample plates) differ, in general [4,8], and the chevron interface makes the slabs nearly independent of each other [1,2]. In such cases, $\varepsilon'(\omega)$ should include a frequency-independent (solitary) contribution arising from one of the chevron slabs and a frequency-dependent (nonsolitary) contribution coming from the second slab. Consequently, $\varepsilon'(\omega)$ should then exhibit a slightly stronger frequency dependence for the range $\omega_{\min} \leq \omega \leq \omega_{\max}$ than in the case presented in Fig. 2. Indeed, such a not very weak dependence of $\varepsilon'(\omega)$ has been found for the same sample, at the temperature $T = 30^\circ\text{C}$ and at the voltage amplitude $U_0 = 30 \text{V}$. This is illustrated in Fig. 3. It is remarkable that the loss function $\varepsilon''(\omega)$ initially increases as the voltage frequency exceeds the threshold ω_{\max} . This indicates that the energy absorbed by the system during each voltage cycle suddenly increases as solitary waves disappear.

The Cole-Cole diagrams can be employed to determine the threshold frequency f_{\min} . Then, using the relation (5), one can calculate the elastic constant K . Values of this constant found at different T and U_0 for samples of different thicknesses are listed in Table I. The parameters P_S and γ , needed to calculate K , have been determined by applying the Diamant-Drenck-Pepinsky bridge technique [14] and the uniform switching method [15], respectively. It can easily be

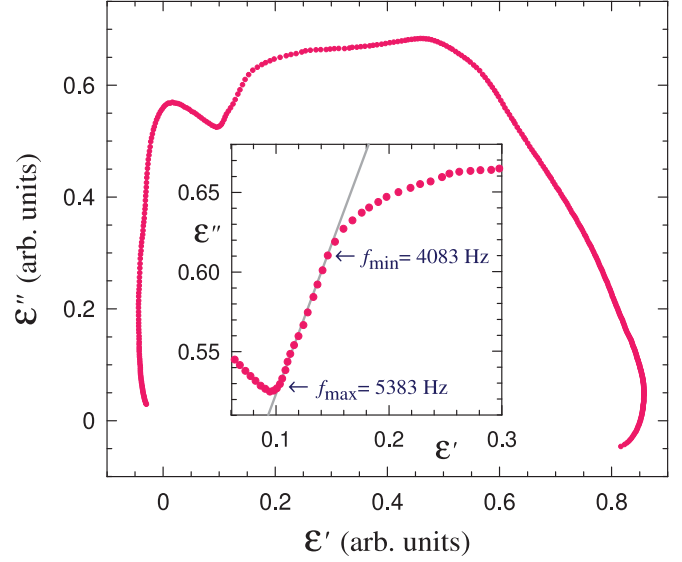


FIG. 3. (Color online) Cole-Cole diagram derived from experimental data (filled circles) for a sample of thickness $D = 5 \mu\text{m}$, at $T = 30^\circ\text{C}$ and $U_0 = 30 \text{V}$. The inset shows a fragment of the diagram revealing solitary molecular motions within only one of chevron slabs (for $f_{\min} \leq f \leq f_{\max}$).

verified that, for P_S and γ found for the studied systems and for the voltages utilized in experiments, the absolute value of the torque $m_e = -\partial w_e/\partial\phi$, associated with the density of the anisotropy energy w_a is much less than the absolute value of the torque $m_d = -\partial w_d/\partial\phi$, corresponding to the density of the depolarization energy w_d . The absolute ratio between these torques is given by $\zeta = |m_e/m_d| = \varepsilon_0^2 \Delta\varepsilon U_0^2 P_S^{-2} D^{-2}$. Using the data of Table I for $D = 5 \mu\text{m}$, $T = 30^\circ\text{C}$, and $U_0 = 20 \text{V}$, as well as the dielectric anisotropy value $\Delta\varepsilon = 0.6$ found for the studied liquid crystal material (at $T = 30^\circ\text{C}$), one obtains $\zeta \approx 0.01$. Although ζ increases as U_0 grows, it still remains very small for all voltages used in the experiments described in this report. Furthermore, Eqs. (3), (4), and (5) imply that ω_{\min} is proportional to E_0 . As seen in Table I, the experimental data obtained for f_{\min} approximately reflect this relation (for

TABLE I. Elastic constant K determined for different D , T , and U_0 . Appropriate values of f_{\min} , P_S , and γ are also given.

D (μm)	T ($^\circ\text{C}$)	U_0 (V)	f_{\min} (Hz)	P_S (Cm^{-2})	γ (Pa s)	K (N)
5	30	20	2760	2.9×10^{-4}	0.055	1.5×10^{-9}
		30	4080			2.0×10^{-9}
		40	7660			3.7×10^{-9}
	50	20	5900	2.0×10^{-4}	0.023	1.6×10^{-9}
		30	10990			2.3×10^{-9}
		40	15170			2.4×10^{-9}
12	30	20	1290	2.7×10^{-4}	0.055	1.4×10^{-8}
		40	2640			1.5×10^{-8}
	50	20	2190	1.8×10^{-4}	0.023	7.8×10^{-9}
		40	5900			1.2×10^{-8}
25.9	30	20	630	3.4×10^{-4}	0.055	7.8×10^{-8}
		40	1420			9.6×10^{-8}
	50	20	1130	2.5×10^{-4}	0.023	4.3×10^{-8}

a given temperature and voltage). However, as can easily be verified, the threshold frequency ω_{\min} theoretically derived by using the method presented above is independent of E_0 (contrary to the experimental results), when the dielectric anisotropy torque term is taken into account in Eq. (1), instead of the depolarization torque component. Therefore, the dielectric anisotropy contribution to the energy density has been neglected here.

A striking feature of the results obtained for the elastic constant K is its evident dependence on the electric field. A possible reason for this apparent dependence is the anisometric shape of molecules forming liquid crystals and field-induced changes in relative, time-averaged, orientations of neighboring molecules within each smectic layer. In fact, due to a large anisometry of liquid crystal molecules, their reorientations under an applied electric field can lead to a dependence of K on E_0 . Since the intralayer and interlayer elastic parameters differ, in general, by two or even three orders in magnitude [11], the

dependence of effective elasticity parameters on the relative orientation of molecules is then very strong, and thereby the observed dependence of K on E_0 can really be considered to be rather strong. Furthermore, the data of Table I show that K determined for samples of various cells not only is different, but also displays a different field dependence. This suggests that measured values of K are affected by surface interactions at cell plates, not included in the model.

In conclusion, the investigation of solitary waves propagating between boundary surfaces of SSFLCs by analyzing their high-field response spectra provides a useful method to determine elastic interactions between molecules within smectic layers. The only required material parameters are the spontaneous polarization and the rotational viscosity.

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- [1] N. A. Clark and S. T. Lagerwall, in *Ferroelectric Liquid Crystals, Principles, Properties and Applications*, edited by J. W. Goodbye, R. Blinc, N. A. Clark, S. T. Lagerwall, M. A. Osipov, S. A. Pikin, T. Sakurai, K. Yoshino, and B. Žekš, (Gordon and Breach, Philadelphia, 1991).
- [2] S. T. Lagerwall, *Ferroelectric and Antiferroelectric Liquid Crystals* (Wiley-VCH, Weinheim, 1999).
- [3] D. A. Allwood, G. Xiong, C. C. Faulkner, D. Atkinson, D. Petit, and R. P. Cowburn, *Science* **309**, 1688 (2005); B. B. Van Aken, J.-P. Rivera, H. Schmid, and M. Fiebig, *Nature (London)* **449**, 702 (2007); Y.-H. Shin, I. Grinberg, I.-W. Chen, and A. M. Rappe, *ibid.* **449**, 881 (2007); J. E. García, R. Pérez, D. A. Ochoa, A. Albareda, M. H. Lente, and J. A. Eiras, *J. Appl. Phys.* **103**, 054108 (2008).
- [4] M. A. Handschy, N. A. Clark, and S. T. Lagerwall, *Phys. Rev. Lett.* **51**, 471 (1983).
- [5] P. E. Cladis, H. R. Brand, and P. L. Finn, *Phys. Rev. A* **28**, 512 (1983); J. E. Maclennan, M. A. Handschy, and N. A. Clark, *ibid.* **34**, 3554 (1986); J. E. Maclennan, N. A. Clark, and M. A. Handschy, in *Solitons in Liquid Crystals*, edited by L. Lam and J. Prost, (Springer, New York, 1991).
- [6] P. Schiller, G. Pelzl, and D. Demus, *Liq. Cryst.* **2**, 21 (1987).
- [7] P. Rudquist *et al.*, *Mech. Mater.* **41**, 1125 (2009).
- [8] W. Jeżewski, W. Kuczyński, and J. Hoffmann, *Phys. Rev. E* **73**, 061702 (2006).
- [9] S. A. Pikin, in *Relaxation Phenomena. Liquid Crystals, Magnetic Systems, Polymers, High-Tc Superconductors, Metallic Glasses*, edited by W. Hasse and S. Wróbel, (Springer, Berlin, 2003).
- [10] W. Kuczyński, Ber. Bunsenges. Phys. Chem. **85**, 234 (1981); F. Gouda, K. Skarp, G. Andersson, H. Kresse, and S. T. Lagerwall, *Jap. J. Appl. Phys.* **28**, 1887 (1989); W. Kuczyński, *Mol. Cryst. Liq. Cryst.* **249**, 97 (1994); W. Kuczyński, K. Nowicka, D. Dardas, W. Jeżewski, and J. Hoffmann, *Opto-Electronics Rev.* **18**, 37 (2010).
- [11] J. A. M. M. van Haaren, *Physica B* **204**, 27 (1995).
- [12] W. Kuczyński, J. Hoffmann, and J. Małecki, *Ferroelectrics* **150**, 279 (1993); W. Jeżewski, W. Kuczyński, and J. Hoffmann, *Liq. Cryst.* **34**, 1299 (2007).
- [13] W. Jeżewski, W. Kuczyński, D. Dardas, K. Nowicka, and J. Hoffmann, *Soft Matter* **6**, 2786 (2010).
- [14] H. Diamant, K. Drenck, and R. Pepinsky, *Rev. Sci. Instrum.* **28**, 30 (1957).
- [15] C. Escher, T. Geelhaar, and E. Böhm, *Liq. Cryst.* **3**, 469 (1988).