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Electric-field-induced weakly chaotic transients in ferroelectric liquid crystals

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Nonlinear dynamics induced in surface stabilized ferroelectric liquid crystals by strong alternating external electric fields is studied both theoretically and experimentally. As has already been shown, molecular reorientations induced by sufficiently strong fields of high-enough frequencies can reveal a long transient behavior that has a weakly chaotic character. The resulting complex dynamics of ferroelectric liquid crystals can be considered not only as a consequence of irregular motions of particular molecules but also as a repercussion of a surface-enforced partial decorrelation of nonlinear molecular motions within smectic layers. To achieve more insight into the nature of this phenomenon and to show that the underlying complex field-induced behavior of smectic liquid crystals is not exceptional, ranges of system parameters for which the chaotic behavior occurs are determined. It is proved that there exists a large enough set of initial phase trajectory points, for which weakly chaotic long-time transitory phenomena occur, and, thereby, it is demonstrated that such a chaotic behavior can be regarded as being typical for strongly field-driven thin liquid crystal systems. Additionally, the influence of low-amplitude random noise on the duration of the transient processes is numerically studied. The strongly nonlinear contribution to the electro-optic response, experimentally determined for liquid crystal samples at frequencies lower than the actual field frequency, is also analyzed for long-time signal sequences. Using a statistical approach to distinguish numerically response signals of samples from noise generated by measuring devices, it is shown that the distribution of sample signals distinctly differs from the device noise. This evidently corroborates the occurrence of the nonlinear low-frequency effect, found earlier for different surface stabilized liquid crystal samples.

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I. INTRODUCTION

The discovery of ferroelectricity in liquid crystals [1] has stimulated not only efforts to explain mechanisms of this unexpected phenomenon but also has given rise to a new challenge for researching new ferroelectric liquid crystals (FLCs). Due to the possibility of direct driving molecular orientations by external electric fields, the FLCs have immediately been considered as materials important for technical applications. Variants of FLCs that have turned out to be very promising toward this aim are surface-stabilized thin-cell systems. The influence of cell surfaces on liquid crystal substances essentially reveals itself through promoting formation of appropriate smectic layer alignment of molecules but can also reveal itself through inducing ferroelectric ordering of liquid crystals that are not ferroelectrics in the bulk [2,3]. An interesting property of surface-stabilized ferroelectric liquid crystals (SSFLCs), in view of technical applications, is the existence of bistable orientational states of molecules (the switching between these states is the basis of working of various FLC-based devices) [3]. Since the threshold energy needed to reorient a molecule between the two states is rather large, the underlying field-induced switching is, by nature, a strongly nonlinear process. Strongly nonlinear electric field-induced reorientations of molecules in SSFLCs have mainly been studied in the context of the process of switching of molecules between their bistable orientational states, determined by surface anchoring interactions [2]. The use of the bistable switching phenomenon in modern display technologies has inspired investigations of the nonlinear dynamics of SSFLCs under strong impulses of the electric field [3]. In such cases, the motion of molecules within each of the smectic layers forming SSFLC systems is completely or almost completely driven by the electric field. Then, in spite of the presence of surface interactions, which tend to prevent molecules from reorienting, the resulting switching motion of molecules can be considered as a collective process with a single characteristic time. In the case of alternating (e.g., sinusoidally) applied fields, the amplitude of molecular reorientations diminishes, in general, as the field frequency grows. In consequence, even if the field amplitude is very large, the range of variations of the molecular azimuthal angle is far narrower at high-enough frequencies than the range between the azimuthal angles of molecules remaining in two stable orientational states. It should be pointed out that, although molecules are not allowed at strong fields of high frequencies to reorient as a whole with large amplitudes, soliton-like local molecular reorientations of amplitudes, comparable to those associated with switching processes, are also possible [4–6]. Such space localized molecular excitations can, however, be registered in rather narrow field frequency ranges.

Obviously, molecular reorientations induced by external fields of large amplitudes can also reveal strongly nonlinear character, even when they are not associated with localized (soliton-like) excitations. This is distinctly reflected in experimentally determined nonlinear response spectra of SSFLCs, especially at frequencies higher than the relaxation frequency [5,6]. It is remarkable that even fundamental (first harmonic) components of these response spectra exhibit a complex form that considerably differs from the simple Debye shape, typical for cases of weak fields. Nevertheless, according to conventional approaches to study nonlinear dynamic effects in FLCs [7–9], the nonlinearity should only be reflected in

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second- and higher-order harmonic response spectra but not in the fundamental harmonics. This may suggest that the expansion of the response function in terms of harmonics is not convergent for sufficiently strong applied fields. Furthermore, the spectra determined experimentally evolve in a very intricate way as the field amplitude grows, displaying irregular anomalies within wide field frequency ranges [10]. Such a complex nonlinear behavior of response function cannot be associated with the occurrence of structural and topological defects, impurities, or ionic currents. Clearly, the complex response of LCs on the action of strong fields can be interpreted as being a result of the appearance in SSFLCs many relaxation processes, each of which is characterized by a specific relaxation time. The origin of these processes can be expected to lie in a complicated interplay between the action of relatively strong alternating fields and a stabilizing influence of the surface interactions on molecules. Indeed, while the surface interactions are large only close to system boundaries, the applied electric field is nearly equal across the system. Additionally, the effect of the field on molecular reorientations is reduced in the high-frequency regime [11,12]. Accordingly, reorientations of molecules near boundary surfaces and in the middle of SSFLCs can be desynchronized [13–16] as the field amplitude grows, provided that field frequencies are high enough. Since the extent of such a desynchronization of molecular orientations can change with the distance of molecules from surfaces of the system, the field-induced molecular motion within smectic layers can then become partially or even entirely uncorrelated.

Since the response function belongs to the most basic quantities characterizing the dynamics of LC systems, a detailed explanation of the underlying mechanism of strongly nonlinear reorientations of molecules in SSFLCs is of great importance. To validate the presumption that the desynchronization of molecular motions in SSFLCs can indeed be considered as a direct consequence of a large complexity of strongly nonlinear response function, a simple model involving only interactions inside smectic layers has been introduced [10]. Then the azimuthal angle of molecules has been treated within the model as a scalar field depending on only one space variable. The resulting equation of the reorientational motion of molecules has numerically been solved for strong applied fields of high frequencies by taking an initial (in the absence of the external field) surface-induced nonuniform distribution of the azimuthal angle. As has been argued, the evolution of the molecular azimuthal angle exhibits very long chaotic transients [17–19], over which the spatial distribution of this angle remains nonuniform. Additionally, the complex nonlinear reorientational dynamics of SSFLCs has also been demonstrated to occur on short time scales, much less than transient periods but greater than the period of field oscillations. The resulting slow dynamic processes has been indicated to concern a modulation of the amplitude of variations of azimuthal angle oscillations with the field frequency. This modulation has been shown to proceed almost periodically with a quasifrequency which decreases in time (and which is less than the field frequency) [10].

It should be noted that the model used to describe strongly nonlinear effects in SSFLCs rather does not precisely reflect the complicated strongly field-driven behavior of real thin LC systems. In particular, this model relies on the assumption (not necessarily correct) that the smectic structure of SSFLCs is well defined, even in the presence of strong alternating fields. Nevertheless, the model yields a surprisingly consistent interpretation of both the origin of the complex anomalies of the response function and the appearance of a distinct response of the systems below the actual frequency of a strong external field. (In principle, when the applied is weak enough, the LC response registered at frequencies less than the field frequency is equal to zero.) Thus, the model can treated as a basis for investigating strongly nonlinear effects in SSFLCs. However, there remain some open questions that are essential not only to evaluate the applicability the model but also for better understanding chaotic dynamic processes in SSFLCs. The main questions refer to the nonuniqueness of initial conditions for chaotic trajectories describing the spatiotemporal evolution of the azimuthal orientation of molecules, the perturbation of chaotic trajectories due of noise generated by measuring devices, and a possibility of distinguishing experimentally registered LC response signals from device noises. In the present paper, these problems are analyzed in detail. Above al, it is indicated that there exists a set of initial phase trajectory points for which molecular reorientations display weakly chaotic character on long times. This initial parameter set (the so-called basin of weakly chaotic transients) is demonstrated to be quite large, and, thereby, the strongly field-induced behavior thin LC systems is argued to be rather typical for such systems. As concerns the effect of perturbations on the weakly chaotic trajectories, it is numerically shown that a weak random noise, imitating field frequency fluctuations in real systems, not only does not change the chaotic behavior of these trajectories but also leads to an increase of the duration of the weakly chaotic transients. Finally, an evidence is given for a distinct difference between long-time sequences of LC response signals, experimentally registered at frequencies lower that the current field frequency, and random signals (noises), produced by measuring devices.

The paper is organized as follows. In Sec. II, the theoretical description of field-induced nonlinear dynamics of ferroelectric SSFLC systems is recapitulated. This introductory section also contains a detailed analysis of chaotic molecular reorientation processes underlying nonlinear effects in these systems. In Sec. III, it is shown that the chaotic reorientation processes in SSFLCs have a generic character, and, thereby, that the occurrence of these effects in a wide class of real SSFLCs can indeed be explained on grounds of chaotic time evolution of field-induced molecular reorientations. Section IV concerns the effect of random noises on the theoretically determined time evolution of molecular orientations, while Sec. V discusses the influence of apparatus noises on experimentally registered strongly nonlinear field-induced responses of the considered systems. In particular, using a simple method to investigate long time sequences of complex response signals, it is demonstrated (Sec. V) that field-induced chaotic molecular reorientations, theoretically predicted to occur at frequencies less than the actual frequency of strong fields, can experimentally be detected in an unquestionable way. Section VI summarizes the obtained results.

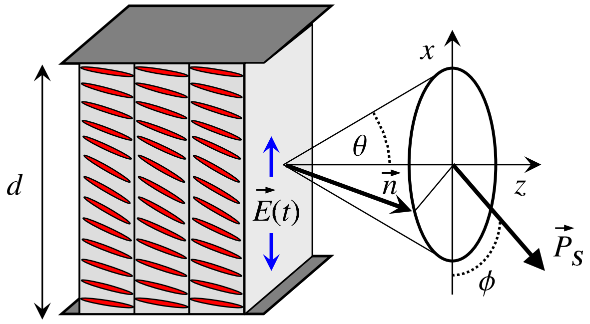

FIG. 1. Geometry of the studied liquid crystal system. Smectic layers are aligned perpendicular to plates of a cell of thickness d. The local polarization $\vec{P}_S$ is perpendicular to the molecular director $\vec{n}$ and lies parallel to the layer plane. The orientation of the director is determined by tilt and azimuthal angles, θ and ϕ, respectively. The external alternating electric field $E(t)$ is applied perpendicular to the boundary plates.

II. NONLINEAR DYNAMICS OF SSFLC SYSTEMS

A. Theoretical method

The investigation of nonlinear field-induced properties of SSFLCs is based here on the theoretical method described in Ref. [10]. In this section, essential steps of this method are briefly presented and discussed. The SSFLCs systems are known to form usually a folded (chevron) smectic layers structure. However, the angle of inclination (with respect to the direction normal to system surfaces) of the layers in the both chevron slabs is always very small. Furthermore, in the presence of strong fields, the smectic layers undergo straightening up towards the position perpendicular to boundary surfaces and the system transforms to a quasibookshelf structure. Therefore, the systems under consideration are assumed to form smectic layers aligned perpendicular to boundary surfaces. Then the vector of local polarization $\vec{P}_S = (-P_S \cos\phi\,,\ -P_S \sin\phi)$, where ϕ denotes the molecular azimuthal angle, is perpendicular to the molecular director $\vec{n}$ and lies parallel to the layer plane. The geometry of the studied systems is shown in Fig. 1.

Since couplings between smectic layers are relatively weak, molecular reorientations under strong fields can be investigated separately within each of the smectic layers. Thus, these reorientations can be described by determining the spatiotemporal dependence of the azimuthal angle ϕ between vectors of the local polarization and the external electric field $E(t) = E_0 \cos(\omega t)$, with E_0 and ω denoting its amplitude and angular frequency, respectively (see Fig. 1). The fluctuations of ϕ are investigated along the x direction, perpendicular to the sample plates. It should be pointed out that, in the case of thin SSFLCs, these fluctuations can be considered as being significant inside entire samples. Obviously, the space variation of ϕ within a smectic layer is initially (when the external field is absent) imposed by boundary conditions (at $x = 0$ and $x = d$, where d is thickness of the system). However, after applying a strong enough electric field, molecular motions are almost entirely field controlled and, consequently, the anchoring effects due to surface interactions can then be neglected. Nevertheless, the effect of action of these interactions should be taken into account by allowing ϕ to be spatially nonuniform before the field is applied. Because the molecular reorientations studied here do not concern local switching processes (soliton-like excitations), the depolarization effect, due to the appearance of polarization charges locally induced in a result of a strong local nonuniformity of the molecular orientation [5,6], is neglected. Then, the spatiotemporal dependence of $\phi = \phi(x,t)$, $0 \leqslant x \leqslant d$, due to applying an alternating electric field to sample plates, is described by the following motion equation [3]:

$$K\frac{\partial^2\phi}{\partial x^2} - \gamma\frac{\partial\phi}{\partial t} = P_S E_0 \sin\phi\, \cos\omega t, \tag{1}$$

where K is the elastic constant characterizing deformations inside smectic layers and γ denotes the rotational viscosity. Note that both K and γ contain the factor $\sin^2\theta$, where θ denotes the molecular tilt angle, i.e., $K = K'\sin^2\theta$ and $\gamma = \gamma'\sin^2\theta$ with K' and γ' being independent of θ. Using dimensionless variables $\tilde{x} = x/d$ and $\tilde{t} = t\omega$, Eq. (1) can be rewritten as

$$\frac{1}{2}c_1\frac{\partial^2\phi}{\partial\tilde{x}^2} - \frac{\partial\phi}{\partial\tilde{t}} = c_2 \sin\phi\, \cos\tilde{t}, \tag{2}$$

where $0 \leqslant \tilde{x} \leqslant 1$ and the parameters

$$c_1 = \frac{2K}{\gamma\omega d^2}, \tag{3}$$

$$c_2 = \frac{P_S U_0}{\gamma\omega d}, \tag{4}$$

with $U_0 = E_0 d$ being the voltage amplitude. It is to be noted that the electric field is treated here as being uniform inside liquid crystal cells, which is not necessarily true. However, various models based on such an approximation have been shown to describe dynamic properties of SSFLCs in a correct, at least qualitatively, manner [5,6,20]. A more general form of the motion equation for smectic (and nematic) liquid crystals involves three elasticity parameters [3,4]. Nevertheless, the simplified one-parameter equation of motion (like the one used here) has been shown to adequately describe molecular reorientations, in cases of both weak and relatively strong fields [7–9]. Indeed, the fundamental as well as higher-order harmonic (nonlinear) contributions to response spectra derived by using Eq. (1) are in agreement with respective components of spectra determined experimentally [7–9]. It should be noted that the parameters in Eq. (1) can be considered as effective temperature-dependent material parameters, accessible experimentally. In particular, since the measured polarization of FLCs usually includes both the dielectric and flexoelectric contributions, P_S occurring in Eq. (1) can be treated as an effective (containing these two contributions) local polarization.

The nonlinear motion equation describing azimuthal reorientations of molecules possesses solutions of different kinds, in accordance with different amplitudes and frequencies of the applied external electric field. For strong but not very strong fields, the solution to the motion equation of the type (1) has been postulated in the form of the harmonic expansion [7–9]. For such a solution, both the fundamental and higher-order harmonic components of the resulting response spectra involve a single relaxation time. However, at very strong fields, the response spectra of SSFLCs display a very complex form that can be considered a result of the appearance in these systems multiple relaxation processes with a continuous distribution of relaxation times rather than a single Debye relation process [3,4]. Thus, to describe adequately strongly nonlinear effects in SSFLCs, the solution of Eq. (2) can be postulated neither by assuming that it can be represented by the harmonic expansion nor by imposing any special functional form on the time dependence of the azimuthal angle. As concerns the space variation of ϕ, it has been argued that, for a wide class of cell systems, ϕ should possess a single extremum within untilted smectic layers of systems with the bookshelf geometry or within tilted layers (arms) of systems with the chevron geometry [3]. Furthermore, assuming that the $\tilde{x}$ dependence of $\phi(\tilde{x},\tilde{t})$ has a quadratic form, theoretically and experimentally obtained results for the dependence of the strength of the dielectric relaxation and the dependence the relaxation time on the cell thickness are in a very good agreement [20]. Thus, in such an indirect way, the quadratic form of the space dependence of ϕ has been demonstrated to be a reasonable approximation to a real profile of ϕ. Since, at strong high-frequency fields, the amplitude of variations of $\phi(\tilde{x},\tilde{t})$ cannot be large across whole smectic layers, the solution to Eq. (2) can thus be expressed in the following form:

$$\phi(\tilde{x},\tilde{t}) = a_0(\tilde{t}) + a_1(\tilde{t})\tilde{x} + a_2(\tilde{t})\tilde{x}^2, \tag{5}$$

where the coefficients $a_i(\tilde{t})$, $i = 0,1,2$, are space independent. Assuming that the external electric field is turned on at $\tilde{t} = 0$, the initial values $a_i(0)$ of these coefficients can essentially be found by specifying appropriate boundary conditions on the static equation for angular momentum balance [Eq. (2) with $c_2 = 0$]. In general, the boundary conditions are given by the relations

$$\phi(0,0) = \phi_0, \;\; \phi(1,0) = \phi_1$$

and

$$\frac{d}{d\tilde{x}}\phi(\tilde{x},0)\big|_{\tilde{x}=0} = \phi_0', \;\; \frac{d}{d\tilde{x}}\phi(\tilde{x},0)\big|_{\tilde{x}=1} = \phi_1',$$

with ϕ_0, ϕ_1, ϕ_0', and ϕ_1' being finite constants, determined by the surface anchoring interactions. Thus, at $\tilde{t} = 0$, one obtains the following relationships: $a_0 = \phi_0, a_1 + a_2 = \phi_1 - \phi_0, a_1 = \phi_0'$, and $a_2 = (\phi_1' - \phi_0')/2$. It should be noted that, in the case of FLCs, the boundary conditions are essentially nonsymmetric with respect to cell plates [2,21], i.e., $\phi_0 \neq \phi_1$ and $\phi_0' \neq -\phi_1'$. Obviously, in both symmetric and nonsymmetric cases of boundary conditions, the coefficients $a_i(0)$, $i = 1,2,3$, are, in general, nonzero. However, the boundary conditions are governed by surface anchoring interactions, which involve unknown, in general, parameters. It is worth noting that there is no microscopic model which would adequately describe electrochemical processes at interfaces between cell plates and liquid crystal molecules. In consequence, the surface anchoring potential have been postulated in different, pure phenomenological manners [2,21]. Instead, the solutions to Eq. (2) are found here for $a_i(0)$ chosen from some ranges, excluding the case of uniform initial distribution of the azimuthal angle, when $a_1(0)$ and $a_2(0)$ are simultaneously equal to zero. Since the interaction of molecules with a strong enough external electric field is dominant, the influence of surfaces on the molecular orientation can be neglected as soon as the strong field is turned on. Accordingly, the action of surface anchoring is here taken into account only through boundary conditions in the absence of the external electric field. Thus, the influence of cell surfaces on the orientation of liquid crystalline molecules is accounted for by taking nonzero values of $a_1(0)$ and $a_2(0)$ (then ϕ is nonuniform at $t = 0$). For simplicity, surface interactions are considered as independent of time. After applying a strong electric field, the system is driven by the field, but, as will be shown below, the initial nonuniformity of $\phi(\tilde{x},\tilde{t})$ can be of great consequence for its time evolution even over very long time periods. It should be pointed out that, although the coefficients a_i change according to electric field oscillations, they do not necessarily have identical functional form. Clearly, the cases when both the coefficients a_1 and a_2 are decreasing (increasing) functions of time correspond to a field-stimulated reduction (magnification) of the nonuniformity level of ϕ. Since $|\phi| < 2\pi$ for sufficiently high field frequencies, one can assume that $a_i(0) = O(1)$, $i = 1,2,3$, and that $a_i(t) = O(1)$, $i = 1,2,3$. Substituting (5) into (2), one obtains equations describing the time evolution of the coefficients a_i, $i = 0,1,2$,

$$\frac{d}{d\tilde{t}}a_0 = c_1 a_2 - c_2 \cos\tilde{t}\, \sin a_0, \tag{6}$$

$$\frac{d}{d\tilde{t}}a_1 = -c_2 a_1 \cos\tilde{t}\, \cos a_0, \tag{7}$$

$$\frac{d}{d\tilde{t}}a_2 = -c_2 \cos\tilde{t} \left(a_2 \cos a_0 - \frac{1}{2}a_1^2 \sin a_0 \right), \tag{8}$$

with the nonlinearity parameters c_1 and c_2 being given by Eqs. (3) and (4), respectively.

B. Numerical analysis

To solve Eqs. (6)–(8) numerically, the parameters c_1 and c_2 must be specified. These parameters have been estimated using values of material constants experimentally found at temperature $T = 30\,^\circ\mathrm{C}$ for the standard liquid crystalline mixture Felix 17-100, introduced into cells of thickness $d = 5\ \mu\mathrm{m}$ [5]. Note that most of the experimental results presented below have been obtained just for this mixture at the same temperature ($T = 30\,^\circ$C, using cells of thickness $d = 5\ \mu$m). The material parameter values used here are as follows: $P_S = 2.9 \times 10^{-4}\,\mathrm{Cm}^{-2}$, $\gamma = 0.055$ Pas, and $K = 1.5 \times 10^{-9}$ N [5,6]. Values of the amplitude and the frequency, $f = \omega/(2\pi)$, of the applied voltage have been taken to be $U_0 = 20,40,60$ V, and $f = 3,4,5$ kHz, respectively. Then the Runge-Kutta-Gill numerical integration method of fourth order [22] has been applied using discrete integer-valued time $t_n = t_0 + nh$, $(n = 0,1,2,\ldots)$, where t_0 is the time at which the electric field is switched on and $h > 0$ is the sampling interval (taken here to be $h = 0.01$). Obviously, the application of the numerical integration procedure yields discrete flow maps which can be iterated beginning with some initial values $a_0^{(0)}$, $a_1^{(0)}$, and $a_2^{(0)}$ (at $n = 0$) of the coefficients a_0, a_1, and a_2, respectively. All numerical results presented below have been obtained taking $t_0 = 0$. General properties of the resulting trajectories have been analyzed by representing graphically

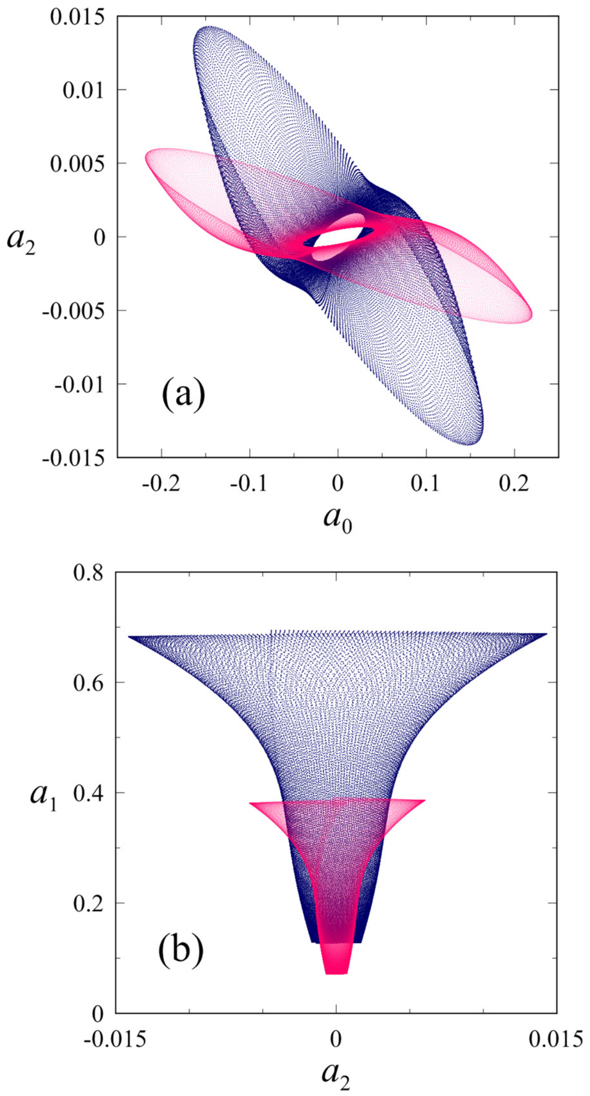

FIG. 2. Trajectory, determined by Eqs. (6)–(8) for $U = 20$ V, $f = 4$ kHz, and for the initial conditions $a_0^{(0)} = -0.01$, $a_1^{(0)} = 0.3$, and $a_2^{(0)} = -0.003$. Projections of the trajectory onto the (a_0,a_2) plane (a) and onto the (a_2,a_1) plane (b) are shown for 185 000 initial iteration steps (blue points) and for 6×10^6 iterations steps (red points) after 10^7 skipped trajectory points.

their projections onto two-dimensional subspaces of the space (a_0,a_1,a_2).

A typical trajectory exhibiting complex pattern is shown in Fig. 2. This trajectory has numerically been generated for $U_0 = 20$ V and $f = 4$ Hz, starting from the initial conditions $a_0^{(0)} = -0.01$, $a_1^{(0)} = 0.3$, and $a_2^{(0)} = -0.003$ (at $n = 0$). The projections of the trajectory onto the planes (a_0,a_2) and (a_2,a_1) are drawn in Figs. 2(a) and 2(b), respectively, for 1.85×10^5 initial iterations ($1 \leqslant n \leqslant 185\,000$) and for 6×10^6 trajectory points generated after $n = 10^7$ iteration steps (i.e., after a long time delay). The drawings of Fig. 2 show that the trajectory is neither periodic nor quasiperiodic, displaying a very slow evolution towards a more compact object. Indeed, the area occupied by the trajectory contracts in the a_1 and a_2 directions and simultaneously slowly broadens in the a_0 direction as n increases. Thus, this may indicate that the trajectory does not evolve towards an attractor [19]. However, due to computer accuracy limitations, one cannot definitely conclude if the trajectory falls onto a one-dimensional subspace of the phase space (a_0,a_1,a_2), determined by $a_1 = 0$ and $a_2 = 0$. Nevertheless, the very slow evolution of the trajectory is characteristic for the *chaotic transients* towards an attractor [18]. Clearly, the occurrence of an asymptotic tendency of a_1 and a_2 to vanish as n grows implies that the action of strong applied fields of appropriately high frequencies leads to a very slow suppression of inhomogeneity in the azimuthal orientation of molecules.

The transitory weak-chaotic behavior trajectories determined by Eqs. (6)–(8) has also been confirmed by calculating local Lyapunov exponents (LLEs) for long time sequences $n = 1,2,\ldots,N$ with $N \gg 1$ [10]. As has been argued, the largest LLE found for each of trajectories derived for different (large) voltage amplitudes is positive although very small and appears to tend to zero for sufficiently large number of trajectory points. More precisely, LLEs display rather irregular N dependence for initial trajectory points, i.e., when N is not very large. After exceeding a limiting number $N = N_1$, dependent on U_0, the LLEs (not only the largest LLE) monotonously tend to zero as N increases. However, due to numerical limitations, the tendency for λ_1 to vanish cannot be analyzed for arbitrary large N. Nevertheless, such a tendency has been confirmed up to the largest number of numerically generated trajectory points, i.e., for $N_2 = 2 \times 10^8$ [10]. As an example, $N_1 \approx 3 \times 10^6$, the largest LLE $\lambda_1 \approx 4 \times 10^{-3}$ at $N = N_1$ and $\lambda_1 \approx 10^{-3}$ at $N = 2 \times 10^8$ for $U_0 = 20$ V, while $N_1 \approx 3 \times 10^7$, $\lambda_1 \approx 6 \times 10^{-3}$ at $N = N_1$ and $\lambda_1 \approx 3 \times 10^{-3}$ at $N = 2 \times 10^8$ for $U_0 = 40$ V. Clearly, such a behavior of the LLEs is typical for systems exhibiting transitory weak chaos [19]. It must, however, be stressed that, although the above discussion concerns trajectories generated for different amplitudes of applied voltages, it is limited to some initial trajectory points, i.e., to some initial space distribution of the azimuthal angle (in real systems, this distribution is determined not only by properties of mere liquid crystal materials but also by surface interactions). Then, for a reliable interpretation of strongly nonlinear properties of real SSFLC systems, it is important to investigate the extent of subspace of the initial trajectory points for which chaotic transients occur.

III. BASIN OF CHAOTIC TRANSIENTS

A characteristic property of the chaotic transients described above is their long lifetime. In cases of systems exhibiting the occurrence of chaotic transients, the analysis of their asymptotic behavior is very difficult, as the transients mask underlying attractors [17,18]. In consequence, the determination of the set of initial trajectory points for which trajectories asymptotically fall on an attractor, i.e., the determination of the basin of attraction, is essentially not possible. Instead, one can then explore a set of initial points for which transient phenomena appear. Such a set of initial trajectory points can be qualified as a *basin of weakly chaotic transients*. Clearly, although the meaning of this term is not precise, as particular transients can be associated with different attractors, the concept of the basin of chaotic transients can be useful for studying transitory effects of long lifetimes [17,18,23–26].

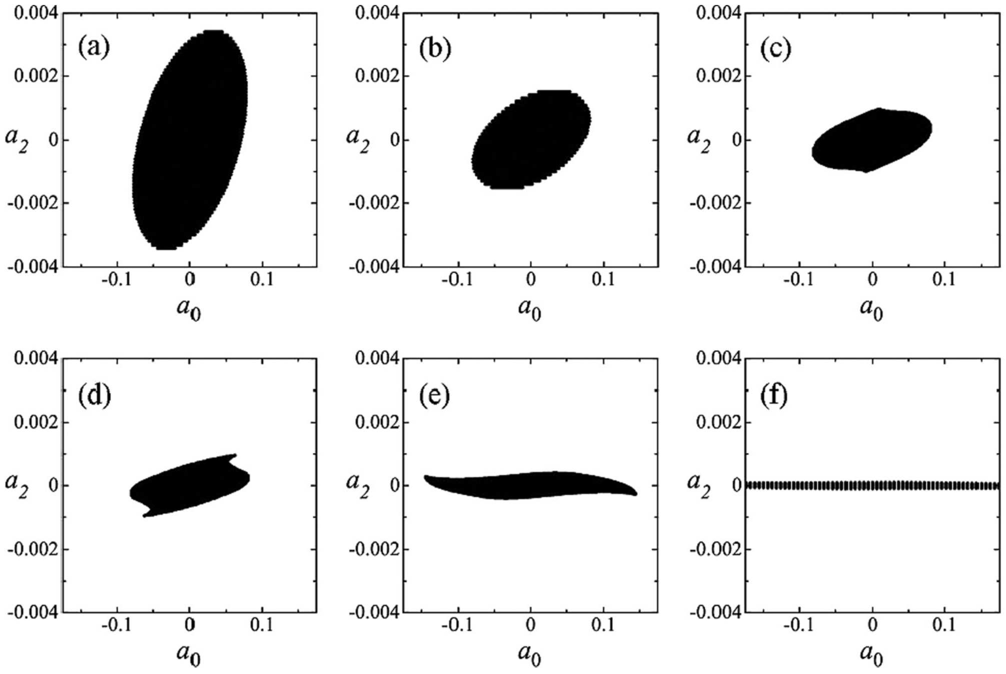

FIG. 3. Sets of initial values of coefficients a_0 and a_2 for which trajectories exhibit long weakly chaotic transients (like those illustrated in Figs. 2 and 3). Each set has been obtained for $U_0 = 20$ V and $f = 4$ kHz but for different initial values of $a_1^{(0)}$: $a_1^{(0)} = 0.3$ (a), $a_1^{(0)} = 0.2$ (b), $a_1^{(0)} = 0.15$ (c), $a_1^{(0)} = 0.12$ (d), $a_1^{(0)} = 0.1$ (e), and $a_1^{(0)} = 0.05$ (f). The sets (shown as black regions) represent respective sections of the basin of chaotic transients in the plane (a_0,a_2).

To illustrate the influence of the initial conditions on the evolution of the studied system and to show that the long-time transitory behavior of the trajectory investigated above is observed not only for special initial conditions, the basin of transients, i.e., the set of initial values $a_0^{(0)}$, $a_1^{(0)}$, and $a_2^{(0)}$, for which trajectories reveal the existence of chaotic transients and remain for a long time within a finite-volume subspace of the phase space, has been studied. Since the determination of the complete basin of transients would require very enormous computations, the investigation of this basin is confined here to an analysis of its cross sections in the planes (a_0,a_1), (a_0,a_2), and (a_1,a_2). More precisely, initial values of pairs of the parameters a_i, $i = 0,1,2$, have successively been chosen from an appropriate rectangle defined in one of the planes, taking the value of the third parameter to be fixed. Then, a given initial trajectory point from a chosen, sufficiently large rectangle has been counted in the basin of chaotic transients if, after $N_0 \gg 1$ initial iteration steps, the trajectory remains within the rectangle and behaves chaotically over further N_1 iteration steps. The results obtained for $U_0 = 20$ V, $f = 4$ kHz, $N_0 = 10^7$, and $N_1 = 6 \times 10^5$ are presented in Figs. 3, 4, and 5. The areas shown in Fig. 3 represent sections of the basin of chaotic transients in the plane (a_0,a_2) for different fixed initial values of $a_1^{(0)}$. To gain more complete insight into the shape of basin of chaotic transients, analogical sections of this basin but in the (a_1,a_2) plane (for fixed values of $a_0^{(0)}$) and in the (a_0,a_1) plane (for fixed values of $a_2^{(0)}$) are presented in Figs. 4 and 5, respectively. It is seen that the basin has a rather simple compact shape and comprises only relatively small (in absolute values) parameters $a_1^{(0)}$ and $a_2^{(0)}$, compared to possible parameter values, determined by the condition that $-\pi \leqslant \phi(\tilde{x}) \leqslant \pi$ for each $\tilde{x} \in [0,1]$. This does not mean, however, that the appearance of such transients in SSFLC systems is, in general, unlikely. Indeed, when surface anchoring interactions are not very strong, the inhomogeneity of the distribution of the azimuthal angle within smectic layers is not large [2,3]. Clearly, such a situation corresponds to small (in absolute values) starting parameters $a_1^{(0)}$ and $a_2^{(0)}$.

IV. SPATIOTEMPORAL DEPENDENCE OF THE AZIMUTHAL ANGLE

Having obtained time solutions of Eqs. (6)–(8), one immediately gets the spatiotemporal dependence of the azimuthal angle $\phi(\tilde{x},\tilde{t})$ [Eq. (5)]. This enables one to examine complex field-induced reorientations of molecules within smectic layers. As the temporal dependencies of a_i, $i = 1,2,3$, in general, differ, the time evolution of $\phi(\tilde{x},\tilde{t})$ differs for various molecular positions $\tilde{x}$. It proves that, in addition to fast oscillations of ϕ with frequency equal to the frequency of the applied voltage, there also occur relatively slow, almost periodic, temporal modulations of the amplitude of these oscillations. This is distinctly illustrated in Fig. 6(a), where an initial temporal evolution of ϕ, up to 2×10^4 iterations, is presented at different molecule positions, for an enormous voltage amplitude $U_0 = 60$ V (allowing a better visualization) and for the voltage frequency $f = 4$ kHz. A continuation of this plot, after 10^6 iteration steps, is shown in Fig. 6(b). As seen in Fig. 6, the period of the modulation grows with time while the depth of the modulation slowly decreases with time, reflecting a long transitory behavior of the trajectory determined by Eqs. (6)–(8) for appropriate initial conditions. It is also visible that both the depth A_m and the phase β_m of modulation vary with $\tilde{x}$, although the modulation

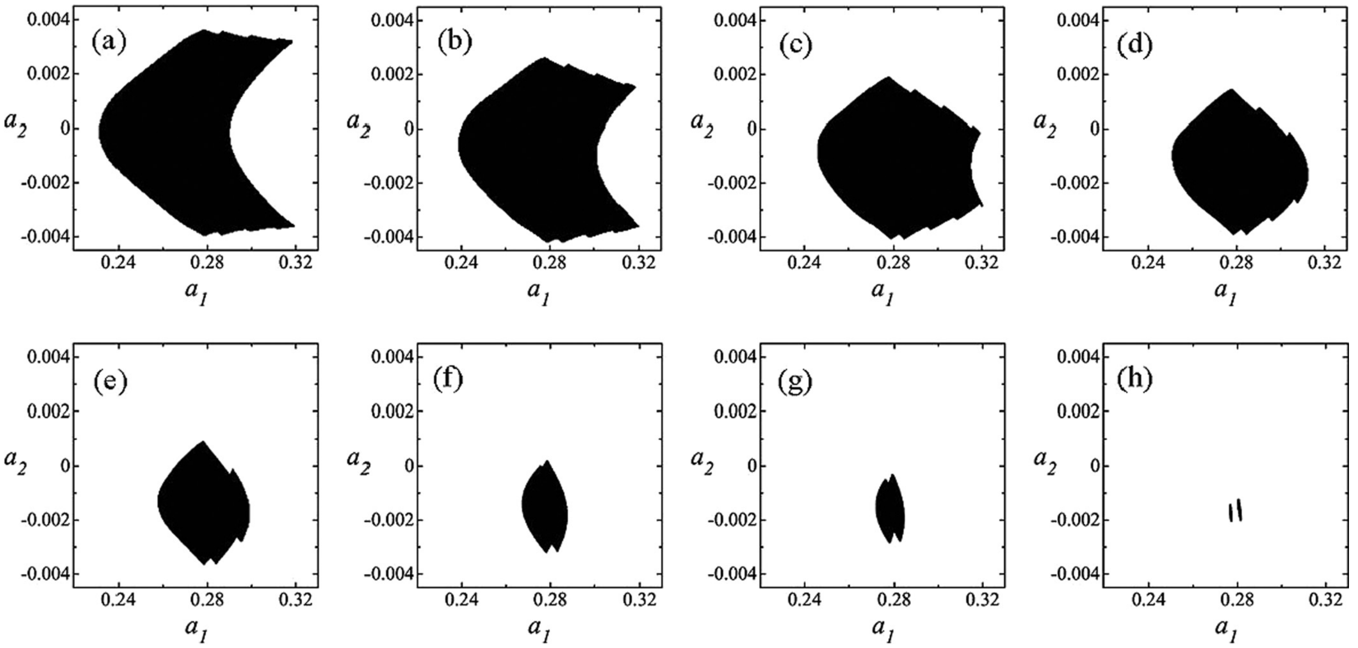

FIG. 4. Sets of initial values of coefficients a_1 and a_2 leading to weakly chaotic transients of the type shown in Figs. 2 and 3 for different values of the remaining parameter: $a_0^{(0)} = -0.01$ (a), $a_0^{(0)} = -0.05$ (b), $a_0^{(0)} = -0.07$ (c), $a_0^{(0)} = -0.08$ (d), $a_0^{(0)} = -0.09$ (e), $a_0^{(0)} = -0.1$ (f), $a_0^{(0)} = -1.05$ (g), and $a_0^{(0)} = -0.11$ (h). The resulting sections of basin of chaotic transients have been obtained at $U_0 = 20$ V and $f = 4$ kHz.

quasifrequency f_m is independent of $\tilde{x}$. Furthermore, A_m, β_m, and f_m all depend on the field frequency f, as illustrated in Fig. 6. This has vast implications for understanding the complexity of nonlinear response spectra $\varepsilon(\omega)$ of SSFLCs. Indeed, the dependence of A_m, β_m, and f_m on f as well as the space dependence of β_m must result in a non-Debye form of the fundamental harmonic component of response spectra at sufficiently high field frequencies, i.e., at frequencies for which the system manifests the chaotic behavior. To validate this in an explicit manner one would know, however, the functional form of the $\phi(\tilde{x},\tilde{t})$ dependence of contributions to $\varepsilon(\omega)$, determined at particular molecule positions $\tilde{x}$. Clearly, the mere dependence of the modulation phase β_m on $\tilde{x}$ is a sign of at least partial deorganization, or more precisely desynchronization, of reorientational motions of molecules within each smectic layer. More specifically, although the phase of fast oscillatory molecular reorientations (with the field frequency) is synchronized, the overall reorientations of molecules are not synchronized due to the space dependence of the phase of the modulation of fast reorientation oscillations. It should be pointed out that such complex field-induced motions of molecules can appear only if the distribution of the azimuthal angle within smectic layers is nonuniform in the absence of the external electric field. Then, after switching on the external electric field, this distribution varies, in general, but remains nonuniform for long time periods.

Since the quasifrequency of the modulation is less than the field frequency, one can expect the appearance of a response signal of SSFLCs below the running frequency of the applied field. Obviously, the modulation of fast field-induced oscillations of the azimuthal angle can occur only if the field puts the system into the chaotic state. As well known, however, chaotic systems are very sensitive to even very weak perturbations [27]. In real systems, field-induced molecular motions are perturbed by noise generated by experimental devices. Such a device noise can originate, e.g., in fluctuations of the frequency and/or the amplitude of the applied electric field. To investigate the influence of random perturbations

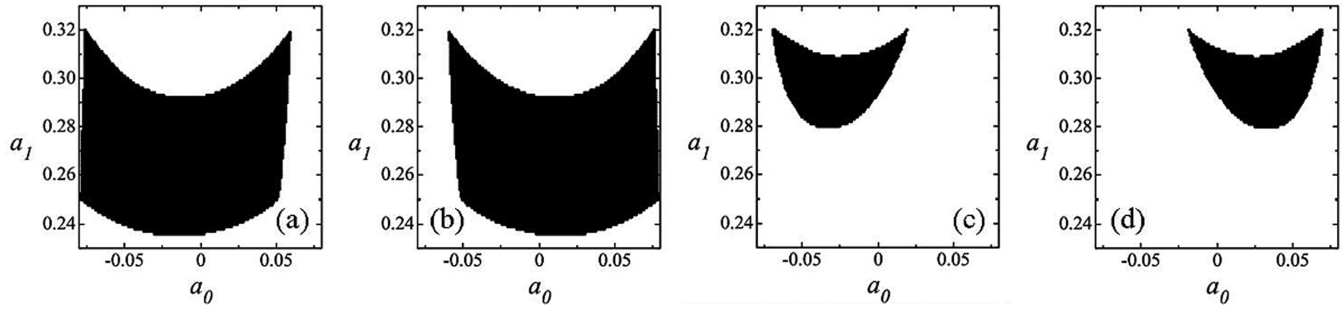

FIG. 5. Sets of initial values of coefficients a_0 and a_1 that yield weakly chaotic transients for different initial values of $a_2^{(0)}$: $a_2^{(0)} = -0.001$ (a), $a_2^{(0)} = 0.001$ (b), $a_2^{(0)} = -0.003$ (c), $a_2^{(0)} = 0.003$ (d), as determined at $U_0 = 20$ V and $f = 4$ kHz.

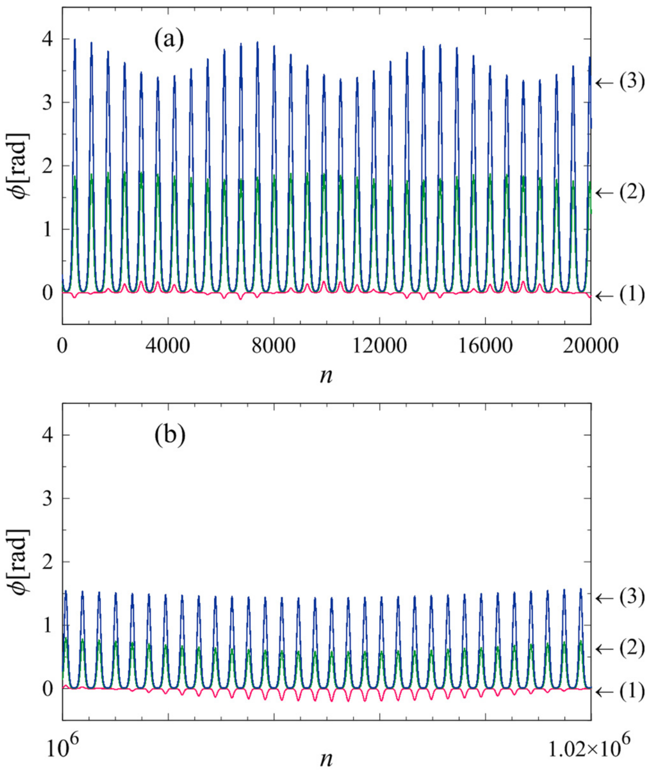

FIG. 6. Molecular azimuthal angle as a function of the number of points of trajectory determined for $U_0 = 60$ V, $f = 4$ kHz, and $\tilde{x} = 0.01$ (1), $\tilde{x} = 0.5$(2), $\tilde{x} = 1.0$ (3). Starting trajectory points were taken to be $a_0^{(0)} = -0.001$, $a_1^{(0)} = 0.3$, and $a_2^{(0)} = 0.01$. The plot (a) shows the azimuthal angle for $N = 2 \times 10^4$ trajectory points, while the diagram (b) displays continuation of the plot (a) for iteration steps $10^6 \leqslant n \leqslant 10^6 + 2 \times 10^4$.

on the field-induced molecular reorientations within the framework of the considered model, the motion equations (6)–(8) have also been solved for discrete-valued time series $t_n = t_0 + nh + nh\, r_\mu(n), n = 1,2, \ldots$, with $r_\mu(n) = \mu\, \mathrm{ran}(n)$, where μ denotes the perturbation (noise) level and the function ran(n) takes a random value from the range $[-0.5, +0.5]$ at each n. In some sense, the resulting perturbation of the trajectory, given by Eqs. (6)–(8), and thereby a modification of the time dependence of ϕ reflects the effect of fluctuations of the field frequency in real systems. The result of disturbing $\phi(\tilde{x},\tilde{t})$ under numerically simulated random noise is illustrated in Fig. 7 for the noise level $\mu = 0.2$. It is seen that, in the presence of noise, the modulation of oscillations of ϕ has a rather complex form. In particular, the noised modulation does not proceed with a single frequency (or with a single quasifrequency). Nevertheless, a simple comparison of plots of Figs. 6(a) and 7(a) shows that even if the level of random noise is relatively high, modulations of the oscillation amplitude of ϕ are not completely destroyed or masked. It is also remarkable that the amplitude of oscillations of ϕ determined for large n is much greater when the random noise is introduced into the system, although it remains smaller than the oscillation amplitude of ϕ derived for small n, in the absence of noise [as seen in Figs. 6(b) and 7(b)]. This implies that a weak random noise extends the duration of chaotic transient processes appearing in the studied model system (cf. Refs. [17] and [18]). It should, however, be noted that noises generated by real devices cannot necessarily be quite random. It is also obvious that if the level of the random noise would be sufficiently high, the noise could have a destructive influence on the resulting chaotic transients, leading to their rapid vanishing.

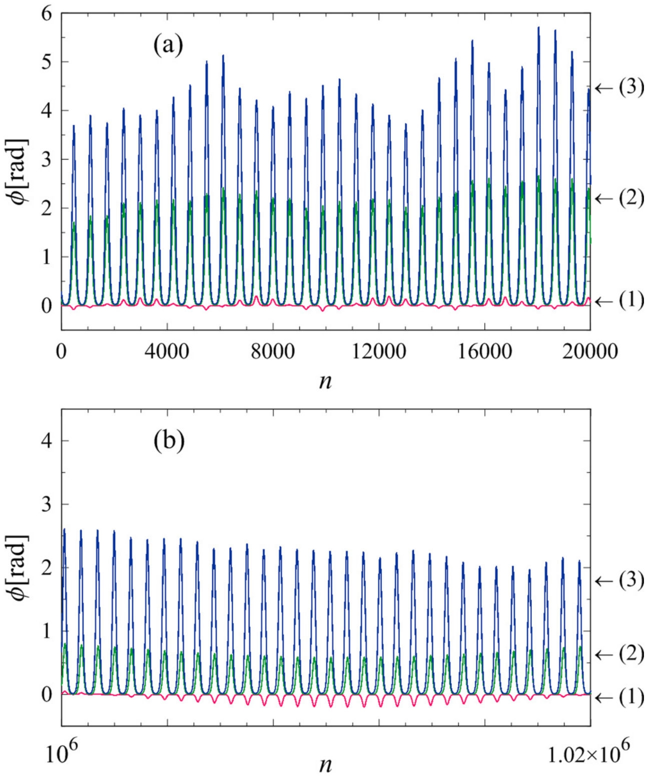

FIG. 7. Plots of the time dependence of the molecular azimuthal angle, analogous to drawings shown in Figs. 6(a) and 6(b), respectively, but obtained in the presence of random noise of the level $\mu = 0.2$ (see the text).

V. MODULATIONS OF ORIENTATIONAL OSCILLATIONS AND DEVICE NOISE

It is rather obvious that complex chaotic reorientational motions of molecules within smectic layers give rise to changes in optical transmission of the sample placed between crossed polarizers and, thereby, lead to the occurrence of the electro-optic effect. Thus, using the photodiode light detector, the resulting transmittance changes can be converted into appropriate variations in the output voltage U_{out}, which are easy to register [10]. As has already been shown, chaotic motions of molecules are reflected not only in the electro-optic response (determined as a function of the field frequency) but also in an electro-optic response at frequencies lower than the frequency of strong enough applied voltages [10].

According to theoretical results presented in Sec. IV, one would expect that the modulation of ϕ could result in an electro-optic signal at a single frequency, dependent on the amplitude and frequency of the external field. However, the modulation is not strictly periodic, as molecular motions undergo very slow chaotic transients. Furthermore, the modulation of molecular reorientations can strongly be affected by fluctuations of the amplitude and frequency of the external electric field. It should be pointed out that the modulations appear when the system remains chaotic and, hence, when the system is very sensitive on even small perturbations [17,18]. Consequently, real systems display the existence of a broad frequency band rather than a single frequency signal in the electro-optic response spectrum registered below the field frequency (provided that the field amplitude is large enough) [10]. Naturally, complex molecular modulated reorientations induced by high alternating voltages should also be visible in similar (modulated) time sequences of the electro-optic (dielectric) response ($U_{\rm out}$), measured at fixed frequencies, lower than the voltage frequency.

To detect experimentally the aperiodic modulation of fast field-driven oscillatory reorientations of molecules in a direct way, long time sequences of the intensity of light transmitted through the system has been recorded at frequencies less than the current field frequency. The sequences have been obtained by taking the time sampling interval $\Delta t = 5 \times 10^{-4}$ s. Experiments have been performed for the Felix 17-100 liquid crystal contained in a cell of thickness $d = 5\ \mu$m. All measurements have been carried out for fixed temperature $T = 30\,^\circ$C, i.e., 47 °C below the temperature of the transition of this SSFLC from ferroelectric to paraelectric phase, and for the applied voltage of different amplitudes but of the same frequency $f = 4$ kHz. At these thermal and field conditions, the layer structure of the studied samples have displayed a large stability, in spite of the occurrence of the transformation effect of the chevroned smectic layers to quasibookshelf layers with smectic layers oriented nearly perpendicular to bounding plates (electrodes) [3]. Results obtained for sufficiently high voltages indicate that the amplifier output voltage $U_{\rm out}$ exhibits irregular time dependence for various chosen frequencies $f_s < f$ of the selective amplifier. The dependence indeed has a form of aperiodically modulated fast periodic oscillations. For $f_s = 176$ Hz and $U_0 = 20$ V, this is illustrated in Fig. 8(a), where rather irregular, aperiodic modulation of fast periodic oscillations of $U_{\rm out}$ is noticeable. On the contrary, $U_{\rm out}$ does not display clear modulations, not only for $U_0 = 0$ but also for $U_0 = 2$ V, as shown in Figs. 8(c) and 8(b), respectively. Note that, at $U_0 = 2.0$ V, numerical solutions of Eqs. (6)–(8), found for different initial conditions, do not reveal chaotic behavior, too. This suggests that the voltage amplitude $U_0 = 2$ V is rather too weak in order to any distinctly chaotic molecular motions could be activated. It should be pointed out that signals recorded at $U_0 = 0$ come only from noises generated by registering devices of the experimental setup, such as the photodiode, selective amplifier, and voltmeter [10]. Clearly, these noises also occur for $U_0 = 2$ V and $U_0 = 20$ V. However, it is remarkable that the time variation of the azimuthal angle, theoretically derived in the presence of random noise for $U_0 = 20$ V (Fig. 7), is qualitatively consistent with the time dependence of the output voltage [Fig. 8(a)]. Then, Fig. 8 gives an explicit evidence for the existence of modulation of oscillatory molecular motions induced by alternating voltages of sufficiently large amplitudes.

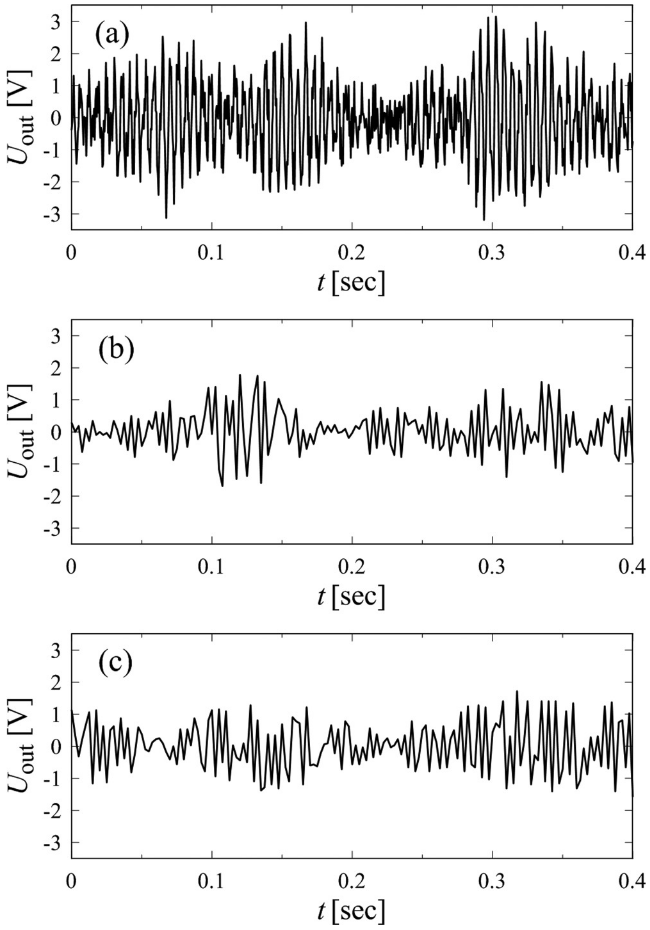

FIG. 8. Exemplary oscilloscope image of time variations of the output voltage of the selective voltmeter, obtained for a time step $\Delta t = 5 \times 10^{-4}$ s. The voltmeter was set to the frequency $f_s = 176$ Hz while the voltage frequency was fixed at $f = 4$ kHz. The voltage amplitude was $U_0 = 20$ V (a), $U_0 = 2$ V (b), and $U_0 = 0$ V (c).

To characterize the variability of $U_{\rm out}$ over long time periods in a more precise manner, a method to describe multifractal sets is here adapted [28]. Although this method has originally been introduced to explore normalized distributions appearing in the context of complex multifractal objects, it can also be used in more general cases, when considered sets are not necessarily fractals. Accordingly, time sequences of measurements of the output voltage $U_{\rm out}(t_i)$, $i = 1,2,\ldots,N$, where t_i denotes time at which the ith measurement of $U_{\rm out}$ is carried out, are analyzed below by using the discrete normalized variable

$$u_i = \frac{|U_{\rm out}(t_i)|}{\sum_{i=1}^{N} |U_{\rm out}(t_i)|} \quad i = 1,2,\ldots,N, \tag{9}$$

where $N \gg 1$ is the length of a sequence of measurements. The sampling time step (Δt) is assumed to be fixed for entire voltage sequences. Next, we define the generating function

$$\lambda_N(q) = \sum_{i=1}^{N} u_i^q \tag{10}$$

with $q \in (-\infty,\infty)$ denoting a continuous variable. Consequently, the time variability of $U_{\rm out}$ can be characterized by

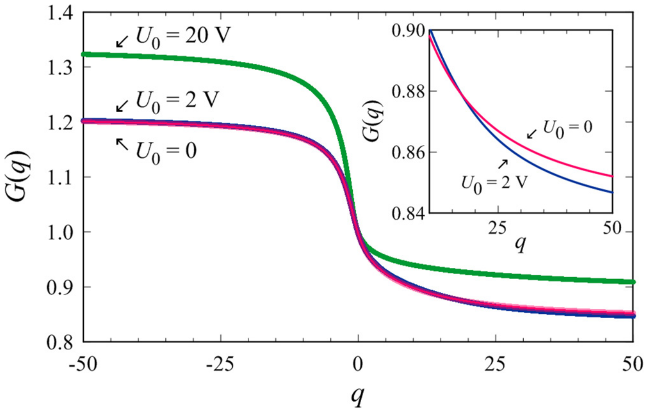

FIG. 9. The function $G(q)$ plotted for discrete time sequences of the output voltage as measured for the applied voltage of amplitudes $U_0 = 0, 2$ V, and 20 V. All experimental data were obtained for $\Delta t = 5 \times 10^{-4}$ s and $N = 5 \times 10^4$.

the function

$$G(q) = \lim_{N\to\infty}\left[\frac{1}{1-q}\frac{\ln \lambda_N(q)}{\ln N}\right]. \tag{11}$$

Due to normalization of $U_{\text{out}}(t_i)$ [see Eq. (9)], $G(q = 1)$ is always equal to 1. Note that, in a rather unrealistic case when $U_{\text{out}}(t_i)$, $i = 1,2,\ldots,N$, are all equal, i.e., when recorded signals exactly constitute white noise, $G(q) = 1$ for all q. However, when the output voltage fluctuates in time, one has $G > 1$ for $q < 0$ and $G < 1$ for $q > 0$ [28]. Thus, complex sequences of registered output voltage can be characterized by the function $G(q)$. This function is drawn in Fig. 9 using experimental data obtained for applied voltage amplitudes $U_0 = 0, 2$ V, 20 V, taking that $\Delta t = 5 \times 10^{-4}$ s and $N = 5 \times 10^4$. It is evidently seen that $G(q)$ determined for $U_0 = 20$ V distinctly deviates from $G(q)$ derived both for $U_0 = 0$ and $U_0 = 2$ V. However, the functions $G(q)$ obtained for $U_0 = 0$ and $U_0 = 2$ V differ from each other in a very slight degree, especially in regime of large q (see the inset in Fig. 9). This clearly indicates that, at sufficiently high alternating voltages, there appears a low frequency response (measured at frequencies lower than the voltage frequency), in addition to noise coming from experimental devices.

The appearance of quasimodulations of fast oscillations of the azimuthal reorientations of molecules and, consequently, the occurrence of a broad band in response spectra of real nonlinear SSFLC systems can be interpreted as a sign of the existence in the system different time scales. This usually indicates that the system simultaneously undergoes different dynamic processes, each of which can be characterized by an individual relaxation time [29,30]. In view of results of Secs. III and IV, strong applied fields of sufficiently high frequencies can drive SSFLCs into a chaotic state, associated with the quasiperiodic modulations of molecular reorientational oscillations with the field frequency. Both the amplitude and the phase of the modulations have been found to be space and field-frequency dependent. Consequently, in the chaotic state of the studied systems, molecular reorientations are at least partially desynchronized. Due to the distinct space dependence of the modulation depth, the partial desynchronization reflects the appearance within each of the smectic layers small regions of space, in each of which reorientations of molecules can approximately be described by a single relaxation time, dependent on the distance of the small space region from one of the boundary surfaces. Thus, the complex form of the high-frequency part of the nonlinear response spectra of real SSFLC systems can be considered a spatial desynchronization of molecular reorientations, arising when the systems is in chaotic state. It is worth noting that the emergence of chaos in the studied systems is possible only if the initial (in the absence of the applied field) space distribution of the azimuthal angle within smectic layers is nonuniform, although it has been shown that chaos can also appear in coupled identical oscillators without assuming any inhomogeneities of the resulting oscillatory network [31]. Obviously, in real SSFLCs, the initial space nonuniformity of molecular azimuthal orientations is caused by surface anchoring interactions. Additionally, this nonuniformity can also be influenced by various dynamic processes that are not taken into account within the considered model, such as ionic currents and motions of zig-zag defects, i.e., specific bookshelflike regions forming spontaneously in the regular chevron structure of SSFLC systems of rather small thicknesses [32–34].

VI. CONCLUSIONS

Molecular reorientation processes induced in thin SSFLCs by strong alternating external electric fields have been studied both theoretically and experimentally. The analysis of numerical solutions of the balance torque equation has indicated that strong enough fields of appropriately high frequencies can degrade synchronized molecular reorientations, preferred at weak fields, in such a way that reorientations of molecules forming smectic liquid crystals reveal a long transient behavior of weakly chaotic character. It has also been proven that there exists a set of initial trajectory points (the so-called basin of weakly chaotic transients), for which chaotic long-time transitory phenomena occur. The long-lasting transients have been argued to be connected with a relatively slow, quasiperiodic modulation of fast oscillations performed by molecules with the field frequency. This modulations have numerically been shown to slow down with time and to be sensitive on noise perturbations of the studied systems. Moreover, the modulations have been argued to be responsible for field-induced effects associated with the appearance of a complex frequency dependence of nonlinear response of SSFLC systems at high frequencies (above the relaxation frequency) and for the occurrence of a low-frequency band in the response spectra of these systems below the running field frequency. Both these effects have been verified experimentally by applying the electro-optic technique. Although the simple dynamic model considered here does not perfectly reflect the intermolecular interactions in real SSFLC systems, it allows one to explain unexpected nonlinear effects induced in these systems by strong electric fields. In particular, the analysis of reorientational molecular motions carried out within this model leads to the conclusion that measurable, strongly nonlinear effects in thin LC samples originate in complex, chaotic spatiotemporal processes within smectic layers. It should be stressed that the effects described in this paper may have important implication for technical applications of SSFLCs. As is well known, the understanding of the complex behavior of SSFLCs at strong applied fields plays a crucial role in constructing various FLC-based devices which exploit nonlinear molecular reorientations (examples of such devices are optical modulators, optical switches, and microdisplays). Therefore, the elaboration of methods to control chaotic molecular reorientations in SSFLC systems under strong fields has practical significance. In particular, the invention of methods to diminish the duration of chaotic transients or to suppress them could contribute to the improvement of SSFLC devices.

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