Theoretical model for layer rotations in smectic-A^* liquid crystals subject to asymmetric electric fields

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Recently, observations of the rotation of smectic layers under certain experimental conditions have been reported in the literature. In this work, the mechanism of such rotations in chiral smectic-A^* liquid crystals under the action of asymmetric, periodic electric fields is studied theoretically. The general conditions for layer rotations have been established using symmetry arguments, and a generalized dynamical theory of the chiral smectic-A^* phase, coupling layer rotations and the electroclinic effect, has been developed. The theory is applied in the specific case when an asymmetric sawtooth electric field is applied over the system, and the dependence of the average angular velocity of the smectic layers on the relevant material constants of the liquid crystal and experimental control parameters is calculated. By rewriting the final equations into dimensionless form, it is demonstrated that the system exhibits a universal behavior, reducing the number of independent material constants and control parameters considerably. [S1063-651X(99)09211-9]

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I. INTRODUCTION

During the past few years, the experimentally observed rotation of smectic liquid-crystal layers under the action of external electric fields has attracted a significant amount of attention [1–5]. This unusual effect is observed only in chiral smectic phases and is believed to be related to the electroclinic effect [6]. At present, however, there exists no theoretical explanation of such a layer rotation and it is the purpose of this paper to present a theoretical model for the simplest case of the chiral smectic-A^* (SmA^*) phase. From a general point of view, the phenomena of layer rotations can be explained by an analysis of different torques acting on the smectic layer normal. Thus the paper begins with a discussion of torques created by an external electric field applied over a smectic liquid-crystalline system. Following this discussion, some general conditions for layer rotations that follow from symmetry arguments are considered.

The ordering of smectic-C^* (SmC^*) liquid crystals can generally be described by specifying two unit vectors. These vectors represent the layer normal $\mathbf{a}$ and the c director $\mathbf{c}$, the latter denoting the tilt direction of the director $\mathbf{n}$ within the smectic layers. If an external electric or magnetic field is applied over the system, a torque $\mathbf{\Gamma}^{\mathrm{ext}}$ is exerted on the director. The consequence of this torque is most easily investigated by dividing $\mathbf{\Gamma}^{\mathrm{ext}}$ into one part, $\mathbf{\Gamma}_{\parallel}^{\mathrm{ext}}$, which is parallel to the layer normal, and another part, $\mathbf{\Gamma}_{\perp}^{\mathrm{ext}}$, which is confined within the smectic layers. It is easily seen that the torque $\mathbf{\Gamma}_{\parallel}^{\mathrm{ext}}$ acts to rotate the director (or the c director, which is equivalent) around the smectic cone at constant tilt, while $\mathbf{\Gamma}_{\perp}^{\mathrm{ext}}$ tends to rotate the director in such a way that the tilt changes [7]. If one studies a system for which the tilt is assumed to be fixed, the consequence of the torque $\mathbf{\Gamma}_{\perp}^{\mathrm{ext}}$ is instead to rotate the entire smectic layers.

Generally, when studying the dynamics of SmC^* liquid crystals from a theoretical point of view, only the torque $\mathbf{\Gamma}_{\parallel}^{\mathrm{ext}}$ is taken into account. This approach is equivalent with the assumption that the smectic layers remain fixed, irrespective of which torques are acting on the system. This implies that there must be some external stabilizing countertorque $\mathbf{\Gamma}^{c}$ that compensates the torque $\mathbf{\Gamma}_{\perp}^{\mathrm{ext}}$, which, as is easily derived from the form of the smectic stress tensor [7], inevitably must be nonzero in most situations. The origin of this countertorque is of course the substrates which normally surround a liquid-crystalline sample. If the countertorque is strong enough, the solution of the dynamical equations can be divided into two parts. The $\mathbf{\Gamma}_{\parallel}^{\mathrm{ext}}$ equation governs the rotation of the c director, while the $\mathbf{\Gamma}_{\perp}^{\mathrm{ext}}$ equation just gives the countertorque required to keep the smectic layers fixed. It is an experimental fact that in most cases the smectic layers are unaffected by external forces, and thus the approach of neglecting $\mathbf{\Gamma}_{\perp}^{\mathrm{ext}}$ when studying c-director dynamics is justified in these cases.

Recently, however, in both SmC^* and SmA^* liquid-crystalline systems confined between parallel glass plates in the bookshelf geometry, a macroscopic rotation of the layer normal has been observed [1–5], both when ac and dc electric fields have been applied across the cell. In these observations the rotation axis of the layer normal is parallel to the electric field, i.e., the smectic layers are not tilted with respect to the surrounding glass plates, but the system remains in the bookshelf geometry.

The basic conditions for the layer rotation in the SmA^* phase in external electric fields can be understood using some very general symmetry arguments. Consider the simplest possible case of a flat SmA^* layer with the electric field $\mathbf{E}$ applied parallel to the smectic plane, perpendicular to the glass plates surrounding the cell. One knows from experiments that in such systems the layers are rotating around the direction of the external field. This is not surprising as the electric field is the only vectorial physical quantity of the system. The rotation of smectic layers is characterized by the

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angular velocity $\boldsymbol{\omega}$ and for symmetry reasons this is expected to be parallel to the electric field $\mathbf{E}$. If one considers only a linear relation between $\boldsymbol{\omega}$ and $\mathbf{E}$, one arrives at

$$\boldsymbol{\omega}=\kappa\mathbf{E}, \tag{1}$$

where κ is determined by some material parameters of the SmA^* phase.

One notes that Eq. (1) is a relationship between the polar vector $\mathbf{E}$ and the pseudovector $\boldsymbol{\omega}$. This linear relationship can only be valid if the constant κ is a pseudoscalar and therefore the medium must be chiral. Thus the smectic-layer rotation can only take place in chiral smectic phases. Second, one notes that the vectors $\boldsymbol{\omega}$ and $\mathbf{E}$ have different transformation properties with respect to time reversal. The angular velocity changes signs under time reversal while the electric field is not necessarily changed. This contradiction can be resolved by substituting Eq. (1), which relates the angular velocity to the external electric field, by a similar relation between the angular velocity and the time derivative of the electric field $\dot{\mathbf{E}}$,

$$\boldsymbol{\omega}=\sigma\dot{\mathbf{E}}. \tag{2}$$

Now the two sides of Eq. (2) have the same symmetry provided the constant σ is a pseudoscalar.

From a general point of view, Eq. (2) indicates that the rotation of chiral smectic layers in a time-dependent electric field is allowed by symmetry. The corresponding angular velocity must change signs under time reversal of the external field, which indeed is observed experimentally. It should be noted that the rotation is actually induced by the time derivative of the electric field and thus it is not expected to occur in a constant field. The latter conclusion, however, appears to be in contradiction with experiments because layer rotation (in the SmC^* phase) has been observed in a dc electric field [5]. On the other hand, in this experiment the liquid crystal was doped by charged impurities which produced an electric current across the cell. One notes that the symmetry of electric current $\mathbf{I}$ is exactly the same as that of the time derivative of the electric field $\dot{\mathbf{E}}$. This means that one can write down the same type of linear relationship between the angular velocity and the electric current

$$\boldsymbol{\omega}=\tilde{\sigma}\mathbf{I}. \tag{3}$$

Thus the rotation of chiral smectic layers can also be induced by an electric current.

Equations (2) and (3) prove the general possibility of layer rotation in the SmA^* phase and describe some conditions required for this effect. At the same time, nothing is said about the particular mechanism of the rotation in chiral smectic phases. In this paper such a mechanism for the SmA^* phase is proposed. Before discussing the equation of motion in this case, one notes that in all experiments with a periodic electric field $\mathbf{E}(t)$, the time derivative $\dot{\mathbf{E}}(t)$ changes signs during the period. According to Eq. (2), this means that the layers are rotating in opposite directions for positive and negative values of $\dot{\mathbf{E}}$, respectively. In practical applications the period of the electric field is small, and thus one is only interested in the constant part of the angular velocity (i.e., the zero-frequency component of ω).

The average angular velocity ω_0 can be obtained by integration of Eq. (2) over time

$$\boldsymbol{\omega}_0=\frac{\sigma}{\tau}\int_0^{\tau}\dot{\mathbf{E}}\,dt, \tag{4}$$

where τ is the period of the field. For any periodical field $E(t)$ the integral in Eq. (4) vanishes, i.e.,

$$\int_0^{\tau}\dot{\mathbf{E}}(t)\,dt=0, \tag{5}$$

and thus, at average, there is no rotation. This problem can be resolved by taking into account that the smectic layers are stabilized by the countertorque $\boldsymbol{\Gamma}^c$, which has its origin from interactions with the surrounding glass plates. In this case the smectic layers are actually rotating if the driving torque exceeds some threshold value. In other words, the layers are only rotating during the part of the period when the time derivative $\dot{\mathbf{E}}$ is sufficiently large. Equation (2) is valid only during this part of the period as well. Now the integral in Eq. (4) does not vanish because one actually has to integrate only over these parts of the total period when the derivative $\dot{\mathbf{E}}(t)$ is large enough.

In this paper, a theoretical model for describing the layer rotation in chiral smectic systems subjected to ac electric fields across the sample is presented. Only dynamic effects are considered. This means that only layer rotations related to time-dependent electric fields are taken into account, and thus the layer rotation observed by Yoshino *et al.* [5] in a dc electric field, which is related to ionic impurities in the sample, is not considered. The study is also restricted to the SmA^* phase and we postpone the study of the SmC^* phase to future work.

The outline of the paper is as follows. In Sec. II the mechanisms causing layer rotations in the SmA^* phase are discussed. The coordinates employed in the calculations are defined and a general discussion of torques in smectic liquid crystals is performed. In Sec. III the electroclinic effect in the SmA^* phase, which is the driving force of the layer rotation in this phase, is discussed and the concept of electroclinic torque is introduced. The general equation governing the dynamics of the layer normal in the SmA^* phase is derived in Sec. IV and in Sec. V this equation is solved for the case when an asymmetric sawtooth electric field is applied to the system. Finally, in Sec. VI the outcome of the calculations is discussed, showing that the results describe existing experimental data correctly. Thus, to our knowledge, the present paper presents the first coherent model for describing layer rotations in the SmA^* phase.

II. FORMULATION OF THE PROBLEM—INTRODUCTION OF NOTATIONS AND DEFINITION OF COORDINATES

In this work we study the dynamics of a SmA^* liquid crystal in the bookshelf geometry. Thus the smectic layers are standing perpendicular to the surrounding glass plates, the layer normal always being parallel to these plates. By

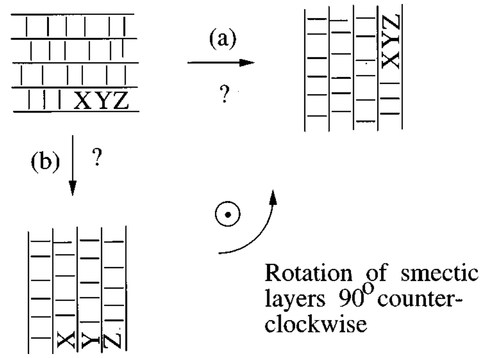

FIG. 1. The two extreme ways of rotating smectic layers. (a) Each molecule stays in its original layer implying that the rotation must be accompanied by a macroscopic mass flow. (b) The molecules only move on a microscopic scale, gradually jumping between the layers.

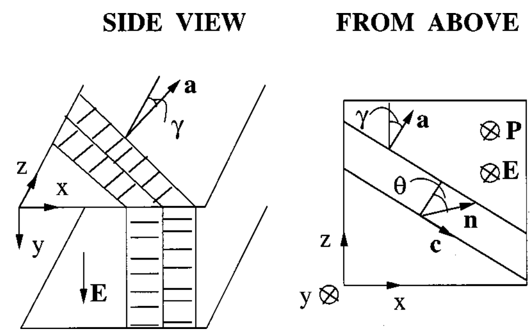

FIG. 2. Definition of coordinates in the present work. Assuming that the system remains in the bookshelf geometry, two coordinates are needed in order to describe it completely. These are the angle γ between the layer normal and the z axis, and the angle θ between the director and the layer normal (the tilt). Both these coordinates are introduced in such a way that they are positive for a rotation around the positive y axis.

applying an ac electric field perpendicular to the glass plates, the system is switching, the mechanism of the switching being the electroclinic effect [6]. However, in contrast to all previous treatments of the problem, we do not assume the smectic layers to be fixed in space. As concluded by symmetry arguments in the preceding section, in the SmA^* phase a time-dependent electric field is expected to exert a torque on the layers, causing these to rotate around an axis parallel to the field. Thus the system will remain in the bookshelf geometry during the switching, however the layer normal has now the possibility of rotating around this axis.

At this stage it is appropriate to discuss what should be meant by a rotating smectic layer. Mathematically, this is synonymous to a system for which the layer normal $\mathbf{a}$ is changing its direction in time. There are, however, several ways to interpret this situation from a microscopic point of view as depicted in Fig. 1. In this figure is shown a system for which the smectic layers are rotated 90° counterclockwise. To keep track on the mass flow associated with this rotation, imagine that three molecules are labeled, denoting them X, Y, and Z. By using the very naive picture that a smectic system consists of separate individual layers, which are piled up on top of each other, the rotation would be accompanied by a macroscopic mass flow, and the three molecules X, Y, and Z would move according to (a). This picture is probably highly unrealistic. If instead one interprets a rotation of a smectic layer as a combination of a change of the preferred direction in which the molecules are pointing and a motion of the molecules on a microscopic scale of the order of half a layer thickness or less (b), the layer normal can be allowed to rotate without being accompanied by a huge macroscopic mass flow in the system. On the other hand, now the molecules which once formed one smectic layer, may find themselves after a rotation belonging to different layers. This latter model for the layer rotation is probably the more correct one. However, we will not discuss the layer rotation from this microscopic point of view, but rather we describe the phenomenon from a macroscopic point of view by keeping track only on the mathematical quantity $\mathbf{a}(t)$.

In Fig. 2, the coordinates used in this work are defined. The surrounding glass plates are taken to be parallel to the xz plane, while the y axis is perpendicular to these plates. The electric field, which is parallel to the y axis, is taken to be positive along the positive y direction. In order to describe the layer normal $\mathbf{a}$, which for standing layers is always confined within the xz plane, an angle γ is introduced, counting γ positive for a rotation of $\mathbf{a}$ around the positive y axis. With the present assumptions, the director will always be confined within the xz plane. To describe the tilt of the director with respect to the layer normal, it is thus sufficient to introduce one coordinate θ. Also θ is introduced in such a way that θ is positive for a rotation of the director with respect to the layer normal around the positive y axis.

When, due to the electroclinic effect, the tilt is changing in time after an ac electric field has been applied across the system, a torque will be exerted on the layer normal. As discussed before by symmetry arguments, and as will be shown later by a straightforward mathematical argument, this torque is parallel to the electric field and will thus tend to rotate the smectic layers around this, still keeping the system in the bookshelf geometry. Once the layers start rotating, one should expect some kind of frictional torque to be exerted on the layers due to the interaction between the substrate and the liquid crystal. Thus we introduce a threshold torque Γ_0 into the model having the consequence that for a driving torque which is less than Γ_0, a balance between the driving torque and the frictional one is possible and no rotation of the smectic layers occurs. Only when the driving torque exceeds Γ_0 should a rotation of the smectic layers be expected to be observed.

III. THE ELECTROCLINIC EFFECT

When an electric field is applied parallel to the smectic layers across a SmA^* liquid crystal, the director becomes tilted with respect to the layer normal and the system exhibits a net polarization P due to the electroclinic effect [6]. As this effect is the driving force of the layer rotation studied in this work, a brief theoretical discussion of the electroclinic effect within the framework of Landau theory is given below.

As a starting point for the discussion, a Landau expansion of the free-energy density for the SmA^*-SmC^* phase transition is used. In the SmA^* phase, a homogeneous electric field will couple only to spatially uniform changes of the order parameters, which for our purpose can be chosen to be θ and P, and accordingly we set $d/dz \equiv 0$ in the expression for the free-energy density. To lowest order, in the SmA* phase one can now write [8]

$$g = \tfrac{1}{2} a\theta^2 + \frac{1}{2\varepsilon} P^2 - CP\theta - EP. \tag{6}$$

In this equation, a, ε, and C are the usual Landau coefficients, where only the coefficient a is assumed to be temperature dependent,

$$a = \alpha(T - T_0). \tag{7}$$

Introducing the SmA*-SmC* phase transition temperature T_c, one can show [8] that a can be rewritten as

$$a = \alpha(T - T_c) + \tilde{K} q_0^2 + \varepsilon C^2, \tag{8}$$

where $\tilde{K}$ is a renormalized elastic constant and q_0 is the wave vector of the pitch at T_c. The equilibrium values of the tilt and polarization in the presence of a static electric field E are denoted θ_{eq} and P_{eq}, respectively. These are calculated by minimizing the free-energy density given by Eq. (6),

$$\frac{\partial g}{\partial \theta} = a\theta - CP = 0, \tag{9}$$

$$\frac{\partial g}{\partial P} = \frac{1}{\varepsilon} P - C\theta - E = 0. \tag{10}$$

Equation (9) implies

$$P_{\mathrm{eq}} = \frac{a}{C} \theta_{\mathrm{eq}}, \tag{11}$$

which, when substituted into Eq. (10), gives

$$\theta_{\mathrm{eq}} = \frac{\varepsilon C}{a - \varepsilon C^2} E, \tag{12}$$

or, by using the relation given by Eq. (8),

$$\theta_{\mathrm{eq}} = \frac{\varepsilon C}{\alpha(T - T_c) + \tilde{K} q_0^2} E. \tag{13}$$

For convenience we introduce the shorthand notation

$$a_0 = \alpha(T - T_c) + \tilde{K} q_0^2, \tag{14}$$

allowing the equilibrium tilt in the presence of an electric field to be written as

$$\theta_{\mathrm{eq}} = \frac{\varepsilon C}{a_0} E. \tag{15}$$

For future use we also notice that Eqs. (8)–(10) enable us to rewrite $\partial g/\partial\theta$ according to

$$\frac{\partial g}{\partial \theta} = a_0 \theta - \varepsilon C E. \tag{16}$$

The Landau-Khalatnikov equations are now employed to write down the dynamic equations of the system as

$$\dot{\theta} = -\Gamma_\theta \frac{\partial g}{\partial \theta}, \tag{17}$$

$$\dot{P} = -\Gamma_P \frac{\partial g}{\partial P}. \tag{18}$$

The two constants Γ_θ and Γ_P are the kinetic coefficients related to the relaxation of tilt fluctuations and polarization fluctuations, respectively, and represent inverse viscosities, i.e., by introducing $\gamma_S = 1/\Gamma_\theta$ and $\gamma_P = 1/\Gamma_P$, one can instead write

$$\gamma_S \dot{\theta} = -\frac{\partial g}{\partial \theta}, \tag{19}$$

$$\gamma_P \dot{P} = -\frac{\partial g}{\partial P}. \tag{20}$$

Here γ_S is the soft-mode rotational viscosity [8] and γ_P is a generalized viscosity related to polarization fluctuations. From experiments one knows that there are two different relaxation times involved in the relaxation of θ and P, since these represent rotations of the molecules around their short and long axis, respectively [9]. It is therefore a good approximation in the problem studied here to assume that the relaxation of polarization fluctuations is infinitely faster than relaxation of tilt fluctuations. Accordingly one can set $\gamma_P = 0$ in Eq. (20). Physically this means that for any value of the tilt θ in the presence of an electric field E, the polarization P always adopts the equilibrium value corresponding to the given values of θ and E. Thus, from Eq. (20) one easily derives the corresponding equilibrium value of the polarization as $\partial g/\partial P = 0$, implying

$$P_{\mathrm{eq}} = \varepsilon C\theta + \varepsilon E. \tag{21}$$

The relaxation of the tilt towards its equilibrium value θ_{eq} given by Eq. (13) or Eq. (15) is governed by Eq. (19). Using Eq. (16), the equation governing the dynamic behavior of the electroclinic effect can thus be written as

$$\gamma_S \dot{\theta} = -a_0 \theta + \varepsilon C E. \tag{22}$$

Applying a static electric field E_0 over a SmA* liquid crystal at time $t = 0$, the solution to Eq. (22) is given by

$$\theta(t) = \frac{\varepsilon C E_0}{a_0} (1 - e^{-(a_0/\gamma_S)t}). \tag{23}$$

From Eq. (23) one deduces the electroclinic response time τ_r of the system as

$$\tau_r = \frac{\gamma_S}{a_0} = \frac{\gamma_S}{\alpha(T - T_c) + \tilde{K} q_0^2}. \tag{24}$$

Neglecting the renormalization $\tilde{K} q_0^2$ in the denominator of Eq. (24), one can estimate τ_r by inserting some typical values for α and γ_S. If these are chosen as $\alpha \sim 10^4\ \mathrm{N/m^2\,K}$

[10,11] and $\gamma_S \sim 1$ Pas [12], the estimated value of τ_r at $T - T_c = 2$ K is $\tau_r \sim 5\times10^{-5}$ s. The inverse of this time, $1/\tau_r \sim 10^4$ Hz, should correspond to the soft-mode relaxation frequency f_S [13] of the system. Indeed, this frequency has been experimentally determined [13,14] to be around 10–50 kHz, a couple of degrees above T_c. As the frequencies of the electric fields considered in this work are at most a couple of hundred Hz, the response time of the system is always much faster than the rate of change of the electric field. Thus, for each value of the electric field, we can always assume that the tilt adopts its corresponding equilibrium value θ_{eq} given by Eq. (15).

IV. DYNAMIC EQUATIONS OF THE SYSTEM WHEN THE SMECTIC LAYERS ARE NOT ASSUMED TO BE FIXED

The system we study is depicted in Fig. 2. It consists of a SmA* liquid crystal in the bookshelf geometry over which a time-dependent electric field has been applied, the field being parallel to the smectic planes. Due to the electroclinic effect, the director will not stay parallel to the layer normal, and the tilt θ is introduced as a dynamic variable of the system. In contrast to all previous theoretical treatments of the electroclinic effect, the smectic layers are not assumed to remain fixed, because from the symmetry considerations performed in Sec. I we expect that applying a time-dependent electric field over the system creates a torque tending to rotate the smectic layers around the field. This means that the system is expected to remain in the bookshelf geometry, however a second dynamical variable γ is needed in order to keep track of the orientation of the smectic layer normal as depicted in Fig. 2. To describe the system, we thus have to introduce two coordinates: γ, which is the angle between the layer normal and the y axis, and θ, which is the tilt of the director with respect to the layer normal. Both these coordinates are introduced in such a way that they are counted positive for a rotation around the positive y axis.

Today, no dynamical theory capable of describing the system studied in this work exists in the literature. The macroscopic dynamic theory for chiral and nonchiral SmA and SmC liquid crystals [7] shows clearly that there exists a coupling between layer rotations and rotations of the director around the smectic cone, and can be employed when studying layer rotations in the SmC* phase [16]. This theory, however, concerns only systems of constant tilt and is thus not applicable when the electroclinic effect is present in the switching as it must clearly be in the SmA* phase. The thermodynamic approach [8] (discussed in Sec. III) to the electroclinic effect, based on a Landau expansion, provides a good description of the electroclinic effect, but assumes the smectic layers to remain fixed. Thus neither of the two approaches is sufficient to model the system studied in this work.

One notes that the two dynamical variables in the system (i.e., the tilt angle θ and the layer rotation angle γ) are not equivalent from a thermodynamic point of view. The tilt angle θ can be called a thermodynamic variable, because the free energy of the SmC* phase depends upon it. The dynamics of θ is relaxational in nature, i.e., the tilt angle relaxes to its equilibrium value determined by the field. In the case of fixed smectic layers, this relaxation process is described by the Landau-Khalatnikov equations (17) or (22). In contrast, the angle γ does not enter the expression for the free-energy density of the ideal SmA* or SmC* phases. On the other hand, the change of γ is obviously associated by some energy dissipation. In this sense the variable γ is analogous to the director $\mathbf{n}$ in the homogeneous nematic phase. In the latter case the free-energy density also does not depend on the orientation of the director, but the change of the orientation is accompanied by a dissipation. In the SmA* phase, when the system is subject to an external electric field, the variables θ and γ are coupled, this coupling determining the rotation of the smectic layers. The dynamics of such a complex system can be described by the more general approach based on the Rayleigh dissipation function. The application of this approach to the study of the dynamics of nematic liquid crystals is discussed in detail by Vertogen and deJeu [17].

In this approach the state of the system (which is assumed to be close to thermodynamic equilibrium) is specified by a set of macroscopic variables X_i and their time derivatives $\dot{X}_i$. The properties of the system are described by using the free-energy density functional $g = g(X_i)$ and the dissipation function $D = D(\dot{X}_i)$. Then the dynamics of the system is described by a set of Euler-Lagrange equations,

$$\frac{\partial D}{\partial \dot{X}_i} = -\frac{\partial g}{\partial X_i}. \tag{25}$$

Assuming the relaxation of the spontaneous polarization to be infinitely faster than the relaxation of the tilt, the polarization will always follow the tilt according to Eq. (21). The system studied is then characterized by the two dynamical variables γ and θ, which are determined from the two equations

$$\frac{\partial D}{\partial \dot{\gamma}} = -\frac{\partial g}{\partial \gamma}, \tag{26a}$$

$$\frac{\partial D}{\partial \dot{\theta}} = -\frac{\partial g}{\partial \theta}. \tag{26b}$$

In the general case, the dissipation function D is written as a quadratic form of the time derivatives $\dot{\gamma}$ and $\dot{\theta}$,

$$D = \tfrac{1}{2}\beta_1 \dot{\gamma}^2 + \tfrac{1}{2}\beta_2 \dot{\theta}^2 + \beta_{12}\dot{\gamma}\dot{\theta}, \tag{27}$$

where the last term describes the dynamical coupling between the variables γ and θ. For the dissipation (27) to be positive definite one must demand β_1 and β_2 to be positive as well as the determinant $\beta_1\beta_2 - \beta_{12}^2$. Thus the following inequalities must be fulfilled by the dynamical coefficients β_i:

$$\beta_1 > 0, \quad \beta_2 > 0, \quad |\beta_{12}| < \sqrt{\beta_1\beta_2}. \tag{28}$$

The coefficient β_{12} can thus adopt both positive and negative values, however its magnitude is restricted according to the last of the inequalities (28).

The elastic energy g in Eqs. (26) is of course the Landau energy given by Eq. (6). As there is no γ dependence in this expression, $\partial g/\partial\gamma=0$, while $\partial g/\partial\theta$ is given by Eq. (16). From Eqs. (26) and (27) one obtains

$$\beta_1\dot{\gamma}+\beta_{12}\dot{\theta}=0, \tag{29}$$

$$\beta_2\dot{\theta}+\beta_{12}\dot{\gamma}=-a_0\theta+\varepsilon CE. \tag{30}$$

Comparing with a system for which the smectic layers are assumed to be fixed [Eq. (22)], one can conclude that β_2 represents the soft-mode rotational viscosity, i.e., $\beta_2\equiv\gamma_S$. Instead, assuming the tilt to be identically zero one notices that the coefficient β_1 corresponds to the rotational viscosity of the smectic layers [18]. The coefficient β_1 can thus be identified to correspond to one of the viscosity coefficients (λ_4) in the above-mentioned dynamic theory [7] for Sm*A* and Sm*C* liquid crystals, i.e., $\beta_1\equiv2\lambda_4$, where β_1 is the dynamic coefficient introduced by Eq. (27) and λ_4 is one of the viscosity coefficients defined by the stress tensor given in Ref. [7].

One notes that the Landau-Khalatnikov equation (17) appears to be a particular case of Eqs. (26b) and (30). Indeed, in the case of fixed smectic layers the dissipation function D can be written as $D=\frac{1}{2}\beta_2\dot{\theta}^2$ and then Eq. (26b) is reduced to Eq. (17). Thus Eqs. (26b) and (30) describe a relaxation of the tilt angle θ in a rotating layer. On the other hand, Eq. (26a) represents a counterbalance of the two generalized torques acting on the smectic layer normal.

At this stage one should notice that there are three different time scales involved in Eqs. (29) and (30). The response time of the electroclinic effect τ_r given by Eq. (24) falls within the submillisecond regime as discussed at the end of Sec. III. The period of the electric fields considered in this work is typically a few tens of a millisecond, while the rotation of the smectic layers has been found experimentally [1,2,15] to correspond to a time scale of several seconds. When solving Eqs. (29) and (30) one can thus proceed in two steps. The angle γ defining the smectic layer normal can be assumed to be constant in a time interval corresponding to one period of the electric field. Thus the term $\beta_{12}\dot{\gamma}$ in Eq. (30) can be neglected and this equation can be approximated as

$$\beta_2\dot{\theta}=-a_0\theta+\varepsilon CE. \tag{31}$$

This is the equation governing the dynamical behavior of the electroclinic effect (22), recalling that we have already identified the dynamical coefficient β_2 with the soft-mode rotational viscosity γ_S. Once Eq. (31) is solved for $\theta(t)$, this solution is substituted into Eq. (29) to obtain $\dot{\gamma}$,

$$\dot{\gamma}=-\frac{\beta_{12}}{\beta_1}\dot{\theta}. \tag{32}$$

It is an experimental fact [1,2,15] that $\dot{\gamma}$ and $\dot{\theta}$ have the same sign, implying the condition

$$\beta_{12}<0. \tag{33}$$

For convenience we introduce the constant κ according to

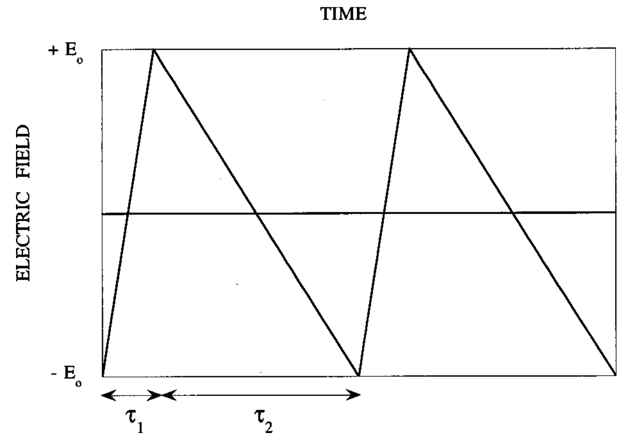

FIG. 3. Asymmetric sawtooth electric field. The field oscillates between $\pm E_0$ and the rise time τ_1 is assumed to be shorter than the decay time τ_2.

$$\kappa=-\frac{\beta_{12}}{\beta_1}>0, \tag{34}$$

allowing Eq. (32) to be rewritten as

$$\dot{\gamma}=\kappa\dot{\theta}. \tag{35}$$

This equation, together with Eq. (31), is the equation being used in the next section to study the rotation of the smectic layers induced by the electroclinic switching when an asymmetric sawtooth electric field is applied over the system.

V. ROTATION OF THE SMECTIC LAYERS DUE TO THE APPLICATION OF AN ASYMMETRIC ELECTRIC FIELD

In this section, the dynamical behavior of the smectic layers when the system is subject to an asymmetric sawtooth electric field is studied. The field under consideration is assumed to be oscillating between the values $\pm E_0$ and is depicted in Fig. 3. The rise time τ_1 is assumed to be shorter than the decay time τ_2, and an asymmetry ratio η of the field is introduced according to

$$\eta=\frac{\tau_2}{\tau_1}>1. \tag{36}$$

The frequency f of the field is related to τ_1 and τ_2 as

$$f=\frac{1}{\tau_1+\tau_2} \tag{37}$$

and is assumed to be much less than the soft-mode relaxation frequency of the system. This condition can be expressed as

$$\tau_1+\tau_2\gg\tau_r, \tag{38}$$

where τ_r is the electroclinic response time given by Eq. (24). This condition is well fulfilled as long as we consider fields with frequencies of 1 kHz or less. Studying one period of the electric field, the time dependence $E(t)$ of the electric field can be expressed as

$$E(t)=E_0\left(\frac{2t}{\tau_1}-1\right) \quad \text{when} \quad t\in[0,\tau_1], \tag{39}$$

$$E=E_0\left(\frac{2\tau_1}{\tau_2}+1-\frac{2t}{\tau_2}\right) \quad \text{when} \quad t\in[\tau_1,\tau_1+\tau_2], \tag{40}$$

while the rate of change of the electric field can be written as

$$\dot{E}=\frac{2E_0}{\tau_1} \quad \text{when} \quad t\in[0,\tau_1], \tag{41}$$

$$\dot{E}=-\frac{2E_0}{\tau_2} \quad \text{when} \quad t\in[\tau_1,\tau_1+\tau_2]. \tag{42}$$

Due to the fact that the rate of change of the electric field is much slower than the electroclinic response time, one can safely assume that during the switching the tilt will always adopt its equilibrium value (15) for the given electric field. The rate of change of the tilt is then simply given by

$$\dot{\theta}=\frac{\varepsilon C}{a_0}\dot{E}. \tag{43}$$

From Eqs. (35) and (43) the equation governing the dynamical behavior of the layers is now derived,

$$\dot{\gamma}=\frac{\kappa\varepsilon C}{a_0}\dot{E}, \tag{44}$$

which, by using Eqs. (41) and (42) can be written as

$$\dot{\gamma}=\frac{2\kappa\varepsilon CE_0}{\tau_1 a_0} \quad \text{when} \quad t\in[0,\tau_1], \tag{45}$$

$$\dot{\gamma}=-\frac{2\kappa\varepsilon CE_0}{\tau_2 a_0} \quad \text{when} \quad t\in[\tau_1,\tau_1+\tau_2]. \tag{46}$$

The net rotation, $\Delta\gamma$, of the smectic layers during one period of the electric field is now calculated as

$$\Delta\gamma=\int_0^{\tau_1+\tau_2}\dot{\gamma}\,dt. \tag{47}$$

The value of $\Delta\gamma$ calculated from Eqs. (45)–(47) is obviously zero. This is easily understood because the larger positive value of $\dot{\gamma}$ during the shorter time τ_1 is exactly compensated by the smaller negative value of $\dot{\gamma}$ during the longer time interval τ_2.

However, to achieve a system in the bookshelf geometry, some kind of surface treatment must be imposed on the glass plates surrounding the sample. Due to this there must exist a stabilizing torque Γ_s, which is responsible for keeping the preferred orientation of the layers. In order to rotate the layers, the driving torque must exceed some threshold, corresponding to the maximum possible value of the stabilizing torque, denoted by Γ_0. The stabilizing torque Γ_s has the nature of a friction torque and adopts the value needed to balance the driving torque as long as this is not large, implying that in this case $\dot{\gamma}=0$. However, if the driving torque exceeds a critical value, the stabilizing torque can no longer increase, but adopts its maximum value Γ_0, always opposing the rotation of the layers. Before adding this torque to the dynamical equations (45) and (46), some caution must be taken regarding the physical dimensions of the quantities studied. The coefficient β_1 represents a viscosity and has the unit Pas [7] while the unit of $\dot{\gamma}$ is s^{-1}. Rewriting Eq. (45) slightly as

$$\beta_1\dot{\gamma}=\frac{2|\beta_{12}|\varepsilon CE_0}{\tau_1 a_0}, \tag{48}$$

one notices that the unit of this equation is $Pa=N/m^2=N\,m/m^3$ and thus the dimension of this equation is torque per unit volume. As the stabilizing torque Γ_s only acts via the substrates, this quantity represents a torque per unit area, i.e., N/m. Writing down the dynamical equation for *one* smectic layer, the driving torque in Eq. (48), being a torque per unit volume, must be multiplied by μd, μ and d being the layer thickness and the sample thickness, respectively. The stabilizing friction torque Γ_s, representing a torque per unit area, must, however, be multiplied by 2μ, the factor 2 stemming from the fact that the sample is surrounded by two glass plates. By adding Γ_s with the proper sign to Eqs. (45) and (46), and multiplying each term with the relevant geometrical factor, one obtains the final dynamical equations of the layer normal,

$$t\in[0,\tau_1], \quad E_0 d>\frac{\Gamma_0\tau_1 a_0}{|\beta_{12}|\varepsilon C}\Rightarrow\dot{\gamma}=\frac{2\kappa\varepsilon CE_0}{\tau_1 a_0}-\frac{2\Gamma_0}{d\beta_1}, \tag{49a}$$

$$t\in[0,\tau_1], \quad E_0 d<\frac{\Gamma_0\tau_1 a_0}{|\beta_{12}|\varepsilon C}\Rightarrow\dot{\gamma}=0, \tag{49b}$$

$$t\in[\tau_1,\tau_1+\tau_2], \quad E_0 d>\frac{\Gamma_0\tau_2 a_0}{|\beta_{12}|\varepsilon C}\Rightarrow\dot{\gamma}=-\frac{2\kappa\varepsilon CE_0}{\tau_2 a_0}+\frac{2\Gamma_0}{d\beta_1}, \tag{50a}$$

$$t\in[\tau_1,\tau_1+\tau_2], \quad E_0 d<\frac{\Gamma_0\tau_2 a_0}{|\beta_{12}|\varepsilon C}\Rightarrow\dot{\gamma}=0. \tag{50b}$$

Substituting Eqs. (49) and (50) into Eq. (47), the net rotation $\Delta\gamma$ during one period of the electric field can be calculated. The average angular velocity $\omega=\langle\dot{\gamma}\rangle$ of the layers is then obtained as

$$\omega=f\Delta\gamma, \tag{51}$$

f being the frequency of the applied field. It is easily seen from Eqs. (49) and (50) that, for a sample of given thickness d, there exist two threshold fields, E_1 and E_2, for which the behavior of the system changes qualitatively. These thresholds are given by

$$E_1=\frac{\Gamma_0\tau_1 a_0}{d|\beta_{12}|\varepsilon C}, \tag{52a}$$

$$E_2=\frac{\Gamma_0\tau_2 a_0}{d|\beta_{12}|\varepsilon C}=\eta E_1, \tag{52b}$$

η being the asymmetry ratio of the field defined by Eq. (36). The assumption $\tau_2>\tau_1$ implies $E_1<E_2$ and the general be-

havior of the system is as follows. If $E_0 < E_1$, the stabilizing torque is large enough to overcome the driving torque and the layers remain fixed during the switching. If $E_1 < E_0 < E_2$, the layers rotate in a positive sense in the time interval $t \in [0,\tau_1]$, but remain fixed in the interval $t \in [\tau_1, \tau_1+\tau_2]$. If $E_0 > E_2$, the layers rotate both in a positive and a negative sense, however there is a net positive rotation during one period of the electric field. Employing Eqs. (47), (49), (50), and (51), the average angular velocity of the smectic layers can be calculated,

$$E_0 < E_1 \Rightarrow \omega = 0, \tag{53a}$$

$$\begin{aligned} E_1 < E_0 < E_2 \Rightarrow \omega &= \frac{2f|\beta_{12}|\varepsilon C E_0}{\beta_1 a_0} - \frac{2f\Gamma_0\tau_1}{d\beta_1} \\ &= \frac{2f\Gamma_0\tau_1}{d\beta_1}\left(\frac{E_0}{E_1}-1\right), \end{aligned} \tag{53b}$$

$$E_0 > E_2 \Rightarrow \omega = \frac{2f\Gamma_0}{d\beta_1}(\tau_2-\tau_1). \tag{53c}$$

In the next section is shown how the solutions (53) can be made more tractable by rewriting them into dimensionless form.

VI. DIMENSIONLESS FORM OF THE EQUATIONS

The key results of the previous sections are the dynamic equations (29) and (30), giving a quantitative description of the coupling between the electroclinic effect $(\dot{\theta}\neq 0)$ and the rotation of the smectic layers $(\dot{\gamma}\neq 0)$. Solving these equations in the specific case when as asymmetric sawtooth electric field is applied over the system, the result is summarized in Eqs. (52) and (53). The result of these equations is governed by six control parameters: the temperature T, the electric field strength E_0, the frequency f of the electric field, the rise and decay times τ_1 and τ_2 of the electric field, and the sample thickness d. It should, however, be observed that by the relation (37) only two of the three parameters f, τ_1, and τ_2 are independent. Furthermore, six material parameters enter the calculations. These are the three viscosities β_1, β_2, and β_{12} and the Landau coefficients α, ε, and C. Also the constant Γ_0, which is a measure of the interaction between the substrates and the liquid-crystalline layers, enters the calculations. Thus, before any numerical results can be produced from Eqs. (53), one needs to assign values to five independent control parameters and seven material constants. However, by rewriting the equations into dimensionless form it can be shown that the system exhibits a universal behavior, only depending on three independent control parameters and one combination of the material constants.

We now introduce three constants t^*, d^*, and E^* with the dimensions of time, length, and electric field, respectively,

$$t^* = \frac{\beta_2}{a_0}, \tag{54a}$$

$$d^* = \frac{\beta_2\Gamma_0}{a_0|\beta_{12}|}, \tag{54b}$$

$$E^* = \frac{a_0}{\varepsilon C}. \tag{54c}$$

These three constants represent a characteristic time, length, and electric-field strength, respectively, the physical significance of which is discussed below. We also introduce four dimensionless quantities for time, frequency, length, and electric-field strength according to

$$\tilde{t} = \frac{t}{t^*}, \tag{55a}$$

$$\tilde{f} = f t^*, \tag{55b}$$

$$\tilde{d} = \frac{d}{d^*}, \tag{55c}$$

$$\tilde{E}_0 = \frac{E}{E_0^*}, \tag{55d}$$

as well as the notations

$$\tilde{\theta} = \theta, \tag{56a}$$

$$\tilde{\gamma} = \gamma, \tag{56b}$$

$$\dot{\tilde{\gamma}} = d\gamma/d\tilde{t} = \frac{1}{t^*}d\gamma/dt, \tag{56c}$$

$$\dot{\tilde{\theta}} = d\theta/d\tilde{t} = \frac{1}{t^*}d\theta/dt. \tag{56d}$$

Substituting the *Ansätzes* (55) and (56) into the previously derived equations, these are transformed into dimensionless form. By this procedure, the thresholds given by Eqs. (52) are transformed into

$$\tilde{E}_1 = \tilde{\tau}_1/\tilde{d}, \tag{57a}$$

$$\tilde{E}_2 = \eta\tilde{E}_2 = \tilde{\tau}_2/\tilde{d}, \tag{57b}$$

where $\tilde{\tau}_1 = \tau_1/t^*$ and $\tilde{\tau}_2 = \tau_2/t^*$ are the dimensionless rise and decay times of the electric field, respectively, and $\eta = \tau_2/\tau_1$ is the previously defined asymmetry ratio of the electric field. In the same way, Eqs. (53) are rewritten as

$$\tilde{E}_0 < \tilde{E}_1 \Rightarrow \tilde{\omega} = 0, \tag{58a}$$

$$\tilde{E}_1 < \tilde{E}_0 < \tilde{E}_2 \Rightarrow \tilde{\omega} = 2\kappa\tilde{f}(\tilde{E}_0 - \tilde{\tau}_1/\tilde{d}), \tag{58b}$$

$$\tilde{E}_0 > \tilde{E}_2 \Rightarrow \tilde{\omega} = 2\kappa\tilde{f}(\tilde{\tau}_2 - \tilde{\tau}_1)/\tilde{d}. \tag{58c}$$

One notices that the results of Eqs. (57) and (58) depend on the three control parameters $\tilde{f}$, $\tilde{\tau}_1$, and $\tilde{\tau}_2$, which are not independent of each other. From Eqs. (36) and (37), the following relations can be derived:

$$\tilde{\tau}_1 = 1/[\tilde{f}(1+\eta)], \tag{59a}$$

$$\tilde{\tau}_2 = \eta/[\tilde{f}(1+\eta)]. \tag{59b}$$

Substituting Eq. (59) into Eqs. (57) and (58), we finally arrive at the expressions that will be employed for the numerical calculations below,

$$\tilde{E}_1\tilde{f} = 1/[\tilde{d}(1+\eta)], \tag{60a}$$

$$\tilde{E}_2\tilde{f} = \eta/[\tilde{d}(1+\eta)], \tag{60b}$$

$$\tilde{E}_0 < \tilde{E}_1 \Rightarrow \tilde{\omega} = 0, \tag{61a}$$

$$\tilde{E}_1 < \tilde{E}_0 < \tilde{E}_2 \Rightarrow \tilde{\omega} = 2\kappa[\tilde{f}\tilde{E}_0 - 1/\tilde{d}(1+\eta)], \tag{61b}$$

$$\tilde{E}_0 > \tilde{E}_2 \Rightarrow \tilde{\omega} = 2\kappa(\eta-1)/[\tilde{d}(\eta+1)]. \tag{61c}$$

From Eqs. (60) and (61) one observes that the number of independent control parameters has been reduced to three, i.e., the product of the frequency and the strength of the electric field, $\tilde{f}\tilde{E}_0$, the asymmetry ratio η, and the sample thickness $\tilde{d}$. It is also seen that only one combination of the material constants enters the calculations. This is the ratio κ, which is defined by Eq. (34).

Before proceeding, we shall identify the physical meaning of the scaling parameters, showing that these are measurable by straightforward experiments. Equation (15) gives the static electroclinic tilt angle as a function of the applied electric field. Comparing this equation with the definition of E^*, given by Eq. (54c), it is clear that by measuring the static electroclinic response θ_{eq} as a function of applied electric field E, the scaling parameter E^* is given as the inverse of the slope of the corresponding graph. From Eqs. (23) and (24) it is seen that the characteristic time scale defined by Eq. (54a) is related to the relaxation of the system back to equilibrium in the presence of an electric field. Indeed it can be shown [8] that t^* is related to the soft-mode relaxation frequency f_s, easily obtained in a dielectric experiment, by

$$t^* = \frac{1}{2\pi f_s}. \tag{62}$$

The characteristic length scale d^* can be determined by using Eq. (58b). From this equation one notices that for a fixed value of the electric-field strength there exists a threshold value $\tilde{d}_{\mathrm{th}}$ for the sample thickness,

$$\tilde{d}_{\mathrm{th}} = \tilde{\tau}_1/\tilde{E}_0, \tag{63}$$

below which the smectic layers cease rotating. Such a threshold has indeed been observed experimentally [3]. By measuring the threshold d_{th} for given values of τ_1 and E_0, the scaling parameter d^* is obtained as

$$d^* = d_{\mathrm{th}}\frac{t^*E_0}{E^*\tau_1}. \tag{64}$$

Thus d^* is easily obtained if t^* and E^* have already been determined.

In the dimensionless version of the theory presented here, it appears as if the temperature dependence of the behavior of the system has vanished. This is, however, not the case, because the temperature dependence is hidden in the choice of the temperature scaling parameter t^* [Eq. (54a)]. Thus, changing the temperature of the system, everything else being unchanged, the temperature dependence of the quantities calculated can implicitly be deduced by keeping track of the temperature dependence of $\tilde{f} = ft^* = f\beta_2/[\alpha(T-T_c) + \tilde{K}q_0^2]$.

Before plotting the results given by Eqs. (60) and (61), we must estimate between which values the control parameters $\tilde{f}\tilde{E}_0$ and $\tilde{d}$ are expected to vary. The assumption (38) guarantees that the tilt always follows the electric field. This assumption is fulfilled if the frequency of the applied field is much smaller than the relaxation frequency of the soft mode and can be formulated as

$$f \ll f_s = \frac{a_0}{2\pi\beta_2} = \frac{1}{2\pi t^*}, \tag{65}$$

where t^* is given by Eq. (54a) and the expression for the soft-mode relaxation frequency is given by [8]. Thus the present calculation is valid only for frequencies for which the relation

$$ft^* = \tilde{f} \ll \frac{1}{2\pi}. \tag{66}$$

In accordance with Eq. (66) we limit the choice of $\tilde{f}$ in the calculations to be $\tilde{f} \in [0,0.02]$. If the frequency is allowed to adopt larger values, the assumption (38) gradually ceases to be valid, and thus the basic assumption that the electroclinic response always follows the field is also violated. Keeping only the terms of lowest order in the Landau expansion (6) demands that the tilt is not too large. The dimensionless form of Eq. (13) can be written

$$\tilde{\theta} = \tilde{E}. \tag{67}$$

Limiting the study to systems for which $\theta < 30°$, the maximum value of $\tilde{E}$ is 0.5 and thus we limit the choice of $\tilde{E}$ to be $\tilde{E} \in [0,0.5]$.

For the sample thickness $\tilde{d}$ there exist a lower threshold, $\tilde{d}_{\mathrm{th}}$, below which the influence of the substrates dominates over the driving torque and no rotation of the smectic layers occurs. This threshold is calculated from Eq. (61b) as

$$\tilde{d}_{\mathrm{th}} = 1/[\tilde{f}\tilde{E}_0(1+\eta)], \tag{68}$$

and gives the lower limit of $\tilde{d}$ for given values of $\tilde{f}$, $\tilde{E}_0$, and η. In the next section are shown some numeric results obtained from the dimensionless model presented above.

VII. NUMERICAL RESULTS

We are now in the position to calculate numerical results from Eqs. (60) and (61). According to the discussion at the end of the preceding section, it is only meaningful to perform calculations for which the product $\tilde{f}\tilde{E}_0$ is of the order of 0.01 or less. In Fig. 4 is plotted the average angular velocity of the

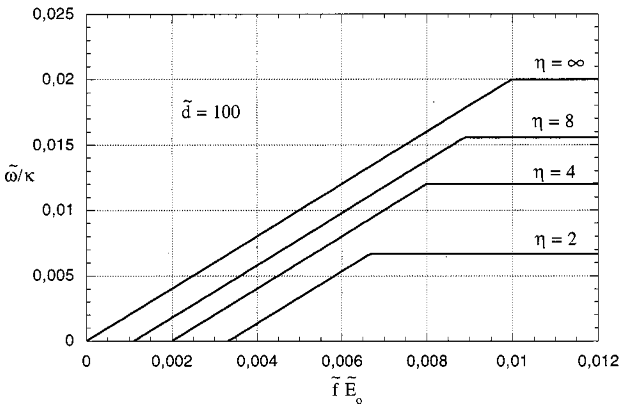

FIG. 4. Average angular velocity of the smectic layers divided by κ [Eq. (34)] as a function of the product between the frequency and the amplitude of the electric field. All quantities are expressed in dimensionless form according to Eqs. (54) and (55) and the calculations are performed for four different values of the asymmetry ratio $\eta = \tau_2/\tau_1$ of the field [Eq. (36)]. The value of the dimensionless sample thickness $\tilde{d}$ in the calculations is $\tilde{d} = 100$.

smectic layer normal divided by κ as a function of the product $\tilde{f}\tilde{E}_0$ for four different values of the asymmetry ratio η ($\eta = 2,4,8,\infty$). The reduced layer thickness in this calculation is chosen to be $\tilde{d} = 100$. One observes that there exists a lower value of $\tilde{f}\tilde{E}_0$ below which the layer normal ceases rotating, corresponding to the threshold given by Eq. (60a). It is clear from the figure that the threshold decreases with increasing η, approaching zero when η approaches infinity. It should be emphasized that the basic assumption that the electroclinic response always follows the electric field does not allow the limit $\eta \to \infty$ to be taken, because for a fixed frequency this would imply $\tau_1 \to 0$. However, already a value of $\eta = 50$, which is not too large to violate this assumption in most cases, corresponds to a saturated value of $\tilde{\omega}$ which differs by only 4% from the calculated value for $\eta = \infty$. Thus the graphs corresponding to $\eta = \infty$ can be used as a guideline for how the system behaves for large η. Increasing $\tilde{f}\tilde{E}_0$ above the threshold, $\tilde{\omega}$ increases linearly until the second threshold [Eq. (60b)] is reached and the average $\tilde{\omega}$ becomes saturated, independent of $\tilde{f}\tilde{E}_0$. For large values of η, this saturated value has an upper limit, $1/\tilde{d}$.

To investigate more thoroughly how the behavior of the system depends on the asymmetry ratio η, study Fig. 5. In the upper part of this figure is shown the two thresholds given by Eqs. (60), $\tilde{f}\tilde{E}_1\tilde{d}$ and $\tilde{f}\tilde{E}_2\tilde{d}$, plotted as functions of η. Without loss of generality it is assumed that $\tau_2 > \tau_1$, and thus the two thresholds are studied by varying η between unity and infinity. In the limit $\eta \to 1$, the asymmetry of the field vanishes and the two thresholds coalesce at the value $\tilde{f}\tilde{E}_0\tilde{d} = 0.5$. Increasing η, the lower threshold decreases towards zero while the upper one is saturated at the value $\tilde{f}\tilde{E}_2\tilde{d} = 1$. In the lower part of Fig. 5 is depicted the saturated value of the average angular velocity (denoted by $\tilde{\omega}_{\max}$) of the layer normal (i.e., the value when the second threshold is exceeded) divided by $\kappa/\tilde{d}$ as a function of η.

In Fig. 6 the thickness dependence of the rotation is investigated. The average angular velocity of the layers divided by k is plotted as a function of the sample thickness for the same values of η as were used in Fig. 4, choosing $\tilde{f}\tilde{E}_0 = 0.005$. One notices that for each value of η, there exists a threshold $\tilde{d}_{\text{tr}}$, below which the rotation vanishes. The threshold decreases towards zero when η increases towards infinity. The features of the graph can be interpreted in the fol-

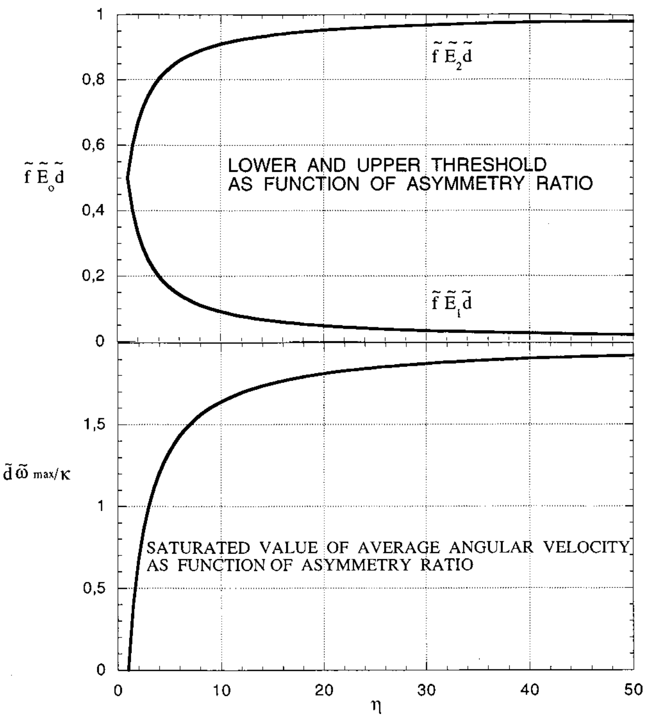

FIG. 5. In the upper part of the figure are depicted the two thresholds defined by Eqs. (60) as functions of the asymmetry ratio $\eta = \tau_2/\tau_1$ of the field [Eq. (36)]. The lower part of the figure displays the average angular velocity of the layer normal multiplied by the sample thickness and divided by κ [Eq. (34)] as a function of η when the second threshold is exceeded. All quantities are expressed in dimensionless form according to Eqs. (54) and (55).

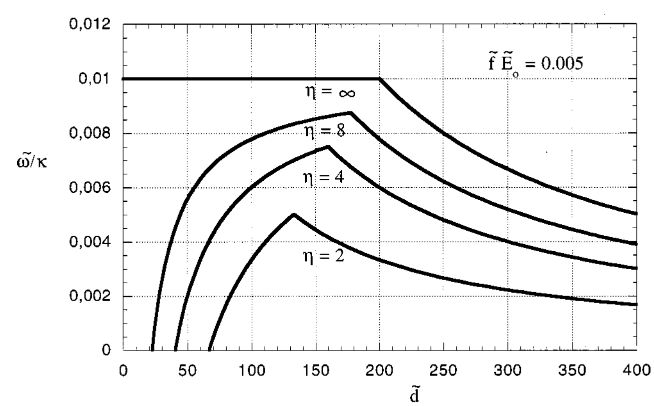

FIG. 6. Average angular velocity of the smectic layers divided by κ [Eq. (34)] as a function of sample thickness. All quantities are expressed in dimensionless form according to Eqs. (54) and (55) and the calculations are performed for four different values of the asymmetry ratio $\eta = \tau_2/\tau_1$ of the field [Eq. (36)]. The value of the product between the dimensionless frequency and the amplitude of the field in the calculations is $\tilde{f}\tilde{E}_0 = 0.005$.

lowing way. Once the threshold $\tilde{d}_{tr}$ is exceeded, the average angular velocity of the layer normal increases with increasing sample thickness. This is due to the fact that the driving torque is a bulk effect, thus increasing with increasing sample thickness. On the other hand, the countertorque Γ_0, acting only at the surfaces of the substrates, is independent of the sample thickness. As long as the second threshold [Eq. (60b)] is not exceeded, the layers rotate only during the fast changing part of the electric field (the τ_1 part). At a certain value of the sample thickness, however, the second threshold is exceeded and the layers rotate in opposite directions during the fast changing and the slow changing parts of the field. In this regime it is the difference between the driving torque and countertorque that is responsible for the net rotation of the layers. Increasing $\tilde{d}$, the relative difference between these torques during the fast and slow changing of the field decreases. Thus, for an infinitely thick sample the layers just oscillate back and forth, and the average rotation of the layers approaches zero.

VIII. DISCUSSION

According to the present model, the rotation of the layers in the SmA^* phase is driven by the electroclinic effect. If the frequency of the external electric field is sufficiently low, the tilt angle θ always follows the field and the time derivative of the tilt is proportional to that of the field. As it is an experimental fact, supported by the symmetry consideration performed in the Introduction, that the axis of rotation coincides with that of the electric field, we do not consider the possibility that the layers rotate in such a way that the bookshelf arrangement of the layers is destroyed. Thus the orientation of the layers needs to be described by one angle γ only, specifying the orientation of the smectic layer normal as depicted in Fig. 2. One important result of the present theory is Eq. (29), describing a balance of generalized torques acting on the smectic layers. According to this equation, $\dot{\gamma}\sim\dot{\theta}$, and as a result the rate of change of γ is proportional to the rate of change of the electric field as is shown by Eq. (44). This explains, in principle, why a time-dependent external electric field can rotate the smectic layers. One notes that the coefficient of proportionality in Eq. (44) contains the pseudoscalar C, which is nonzero only if the smectic material is chiral (cf. Sec. II, where the theory of the electroclinic effect is discussed). The coefficient C also determines the polarization induced by the tilt in the SmC^* phase. Thus the phenomenon of layer rotation is possible only in chiral smectic phases. This conclusion also follows from general symmetry arguments, discussed in the Introduction, and is supported by existing experimental data [4,15].

We note, however, that Eq. (44) alone cannot explain the net rotation of the smectic layers under the action of an asymmetric, periodic electric field, because the field rotates the layers in opposite directions during different parts of the period, these rotations exactly canceling each other. As discussed in Sec. III, also the interaction between the smectic layers and the substrates has to be taken into account. This interaction results in an additional frictional torque acting on the smectic layers, and due to this torque the net layer rotation becomes nonzero. The resulting dependence of the average angular velocity of the layers on the amplitude of an asymmetric sawtooth electric field (cf. Fig. 3) is depicted in Fig. 4. For $E<E_1$ there is no layer rotation because the rate of change of the electric field is too small to overcome the friction at the substrates. In the interval $E_1<E_0<E_2$, the layers rotate only during the rise of the electric field (provided $\tau_1<\tau_2$). Finally, for large fields ($E_0>E_2$) the layers rotates in opposite directions during the rise and decay of the field, respectively. The resulting average angular velocity of the layers in this case does not depend on the amplitude of the field and is proportional to the frictional torque Γ_0. In this regime the average angular velocity is also proportional to the difference $\tau_1-\tau_2$, which is obviously nonzero only for asymmetric fields. It might appear contradictory that the rotation of the layers is proportional to the friction torque Γ_0, but in accordance with the discussion at the end of Sec. VII it is the asymmetry in the regime above E_2 which is responsible for the rotation. This asymmetry is related to the parameter Γ_0/d and decreases when this parameter increases. It should also be observed from Eq. (52b) that by increasing this parameter for a fixed amplitude of the electric field the threshold E_2 increases, thus taking the system into the regime $E_1<E_0<E_2$, where Eq. (53b) clearly shows that ω decreases with increasing Γ_0/d, until ultimately the rotation completely vanishes for a large enough value of Γ_0/d.

The main results of the application of the theory developed in Sec. IV are contained in Eqs. (52) and (53). By rewriting these equations into dimensionless form, using the scaling parameters defined by Eqs. (54), the result can be reformulated according to Eqs. (60) and (61). Only one combination of the material constants, $\kappa=|\beta_{12}|/\beta_1$, enters these equations. From Eq. (61b) one notices that by plotting $\tilde{\omega}$ as a function of $\tilde{f}\tilde{E}_0$ in the regime $E_1<E_0<E_2$, the expected result is a straight line, the slope of which is 2κ, and thus κ can easily be determined from experiments. Concerning β_{12}, it is possible to conclude from the experimentally observed [1,2,15] sense of rotation of the smectic layers that β_{12} should be expected to be negative. In Figs. 4–6 the behavior of the system as a function of the various control parameters is demonstrated. These results can be used for an experimental check of the theory. Today, systematic investigations of the behavior of the system are very scarce in the literature. However, the features of the thresholds demonstrated in Figs. 4 and 6, as well as the general trends of the average angular velocity as a function of field strength, frequency, and sample thickness, seem to be confirmed by the few experiments being reported [1–4,15].

As mentioned above, the layer rotation has been systematically experimentally investigated mainly in the SmC^* phase [2–4,15] and there exists practically no quantitative experimental information about the characteristics of the rotation in the SmA^* phase. Experimental studies of the effect in the SmC^* phase are often performed using asymmetric square-well electric fields, which also rotate the layers in this phase. At the same time, the present theory indicates that there should be no rotation of the layers in the SmA^* phase under the action of an asymmetric square-well field, in contrast to a sawtooth one. This conclusion can easily be checked experimentally and, indeed, preliminary results confirm this prediction [15].

The present theoretical model explains the mechanism of layer rotations in the SmA^* phase. Most of the experimental studies of this effect, however, have been performed in the SmC^* phase. One notes that although the qualitative origin of the rotation in the SmC^* phase should, at least partly, be the same, the particular mechanism is expected here to be more complicated. In the SmC^* phase, the tilt angle is nonzero and as a result there exists one additional degree of freedom, corresponding to the rotation of the director around the smectic cone. Also the electroclinic effect is present in the SmC^* phase and as a result the dynamics of the SmC^* phase (including layer rotations) appears to be more complex. The generalization of the present theory to the SmC^* phase is currently in progress.

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