Solitonlike dynamics in submicron ferroelectric liquid-crystal cells

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Using a linear electro-optic response technique we have determined the phase mode dynamics of a chiral ferroelectric smectic-C^* liquid crystal confined in a wedge-type cell. Below the smectic-A–smectic-C^* phase transition we have observed two polar phase modes. The relaxation rate of the higher-frequency mode scales as the inverse square of the cell thickness and is identified as a transverse solitary wave in the splayed ferroelectric structure. The relaxation rate of the slow mode is on the other hand nearly thickness independent. By increasing measuring voltage, this mode shows a nonlinear crossover to the collective switching of the polarization of the cell as a whole. The observed dynamics is described within the Landau-Khalatnikov theory, where the polarization renormalized dynamics and the effects of confinement are taken into account. [S1063-651X(98)04806-5]

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I. INTRODUCTION

When a ferroelectric liquid crystal is confined between two polymer-coated surfaces, separated by a distance of the order of a micrometer, one usually observes a wide variety of textures [1–7]. These textures result from the interplay of the effects of confinement, which is dictated by the surface and the intrinsic behavior, which is imposed by the bulk liquid crystal. The nature and physical properties of these textures have been the subject of intensive research, which was primarily motivated by the potential use of ferroelectric smectic phases in fast switching electro-optic devices [8]. Here we show that a solitonlike dynamics, characteristic of incommensurate crystals, can be observed when a ferroelectric liquid crystal is confined in a bookshelf geometry between two parallel plates with equal and polar boundary conditions.

If the thickness of the cell is much smaller than the bulk helical period, the helix of the ferroelectric smectic-C^* structure in a thin cell is unwound by the surface forces [8,9] and the phase is homogeneously ordered in the z direction, i.e., normal to the smectic layers. Very often, ''splayed'' states of the director field are observed in real samples, where the electric dipole moments of liquid crystalline molecules have a preferential orientation at the surface, as shown in Fig. 1(a). The splayed texture is unwound and homogeneous along the layer normal, but is inhomogeneous in the transverse direction due to the polar surface coupling [2,6]. In fact, polar anchoring of liquid-crystalline molecules is quite widely observed [10–13]. The fundamental reason for the surface polarity is not necessarily related to the surface electrical properties, but is a general result of symmetry breaking by the wall. As a consequence, the surface interaction energy of a chiral molecule depends on the angle between the polarization and the surface normal, resulting in polar anchoring.

The dynamics of splayed states has been extensively studied in the past in the limit of strong external electric fields (for a review see Refs. [14] and [15]), where a collective switching of the cell as a whole takes place. However, no systematic analysis has been done in the very low-field limit, where the ''intrinsic'' dynamics of the system should be observed. Exceptions are studies of the dynamics in wedge-type cells in a range of thickness larger than several micrometers [16–18]. Here we report on the dynamics of splayed, ultrathin wedge-type cells using the linear electro-optic response technique. The cell thickness could be varied between several micrometers to less than half a micrometer. The electro-optic response of the cells was measured over a large range of measuring voltage. In this way we could follow the dynamics from the intrinsic, linear response regime at very small testing fields to the nonlinear regime in the limit of very large fields. In our experiments we have observed two modes and both have been identified as the phase modes, i.e., collective excitations of the molecular motion around the cone of the tilt angle. The relaxation rate of the faster mode is strongly thickness dependent and represents excitations of the phase profile in a direction perpendicular to the confining surfaces. The thickness dependent relaxation rate of the fast mode indicates the solitonlike nature of this mode, which is a result of confinement and polar boundary conditions. The relaxation rate of the slower mode is nearly thickness independent. By increasing the measuring voltage, this mode shows a crossover into a switching of the polarization of the cell as a whole.

II. THEORY

Let us consider a ferroelectric liquid crystal, confined in a bookshelf geometry between two parallel plates, separated by a distance d, as shown in Fig. 1(a). The boundary conditions are polar and fixed on both confining surfaces, which leads to the well-known splayed (twisted) states. The director field is homogeneous in the y-z plane, and is inhomogeneous in the x direction due to this polar anchoring. The splay distortion of the director field is accompanied by the splay distortion of the polarization field [2]. This results in an induced space charge $\rho_{\text{induced}}(\vec{r}) = -\text{div}\,\vec{P}$, which has substantial influence on both the dynamic and static properties of splayed states.
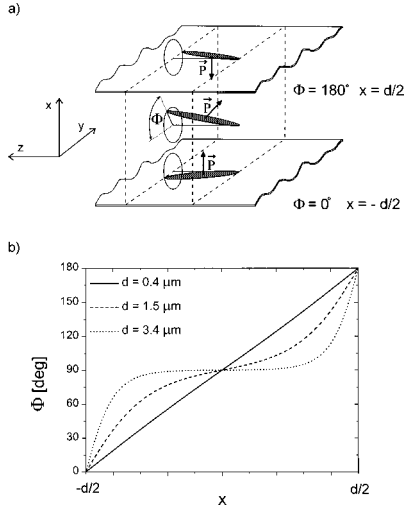
FIG. 1. (a) The ''splayed'' state of a ferroelectric liquid crystal in very thin homogeneous layers for polar surface anchoring. The dipole moments prefer an ''outward'' surface direction, pointing into the cell. This results in a splayed state: if one moves along the cell thickness, the direction of polarization is rotated by 180° from one surface to another. This rotation can be either clockwise or anticlockwise. (b) Crossover from the plane-wave modulation of splayed director field at small thickness to the solitonlike modulation for large thickness of the cell. The equilibrium phase profile for different cell thickness was calculated from Eq. (3) for the polarization coherence length $\xi_P = 0.22$ μm, which corresponds to CE-8 ferroelectric liquid crystal with $K = 4.1 \times 10^{-11}$ N [29], $\varepsilon = 3.2$, $P_0 = 5.1 \times 10^{-5}$ A s/m^2, and $\theta_0 = 20°$.

The free-energy density in the constant amplitude (tilt) and equal elastic constant approximation is [19,20]

$$g(x) = g_0 + \tfrac{1}{2} K \theta_0^2 \left(\frac{d\Phi}{dx} \right)^2 - \tfrac{1}{2} \vec{D} \vec{E} - \vec{P} \vec{E}. \tag{1}$$

Here $\Phi(x)$ is the phase profile across the cell, $\vec{D}(x) = \varepsilon \varepsilon_0 \vec{E}(x)$ is the electric displacement field, ε is the dielectric constant of a liquid crystal at high frequencies, and $\vec{P} = P_0(\cos \Phi, \sin \Phi, 0)$ is the polarization. We have taken the dielectric constant at high frequencies in order to exclude the contribution of the permanent dipoles. The second term represents the elastic energy due to deformation of the director profile. The third term represents the electrostatic energy of induced dipoles and the last term represents the electrostatic energy of permanent dipoles in the local electric field. Here the local electric field can originate either from external sources or from the induced charges. By introducing the electric potential $\vec{E}(x) = -\vec{\nabla} \varphi(x)$ and minimizing the free energy $(1/d) \int g(x) dx$ with respect to $\Phi(x)$ and $\varphi(x)$ we obtain a set of coupled equations for the stationary phase profile and the electric potential:

$$\frac{d^2\Phi_0}{dx^2} + \frac{P_0}{K\theta_0^2} \sin \Phi_0 \frac{d\varphi}{dx} = 0, \tag{2a}$$

$$\frac{d^2\varphi}{dx^2} + \frac{P_0}{\varepsilon \varepsilon_0} \sin \Phi_0 \frac{d\Phi_0}{dx} = 0. \tag{2b}$$

Here the second equation is in fact the well-known Poisson equation, which relates the charge distribution to the corresponding potential, $\varepsilon \varepsilon_0 \vec{\nabla}^2 \varphi(\vec{r}) = -\rho(\vec{r}) = \mathrm{div}\, \vec{P}(\vec{r})$. The above set of equations can be solved analytically in the case of zero external field and numerically for the case of a finite field [19,20]. After integrating Eq. (2b), we obtain the sine-Gordon equation for the stationary phase profile $\Phi_0(x)$:

$$\frac{d^2\Phi_0}{dx^2} + \frac{P_0^2}{2\varepsilon \varepsilon_0 K \theta_0^2} \sin 2\Phi_0 = 0, \tag{3}$$

whereas the local electric field is $E_x = d\varphi/dx = (P_0/\varepsilon \varepsilon_0)\cos \Phi_0$. The above equations show that the presence of a splayed spontaneous polarization generates an internal local electric field. This field has the same role as an external homogeneous magnetic or electric field applied to chiral structures. It has a tendency to induce a solitonlike deformation of the phase profile, and finally, for a very high polarization, to flatten the phase profile across the cell. It is easy to understand the origin of this field: any splaylike inhomogeneity of the polarization field results in the appearance of local induced charges. These tend to repel each other via the Coulomb repulsion forces and tend to make the phase profile locally homogeneous. As a result, the induced charges balance in a such a way that the phase profile across the splayed cell obtains a soliton form, which is therefore a profile of minimum elastic and electric energy.

Similarly to the magnetic or electric field induced soliton lattice, the solution of the sine-Gordon equation, which satisfies fixed boundary conditions, $\Phi_0(x = -d/2) = \Phi_0(x = +d/2) = 0$ is a soliton phase profile [21]

$$\sin \Phi_0 = \mathrm{sn}(u, k), \tag{4}$$

where $u = x/\xi_P k + K$ is a reduced coordinate, $\xi_P = \sqrt{\varepsilon \varepsilon_0 K \theta_0^2 / P_0^2}$ is the polarization coherence length, $K(k)$ is the complete elliptic integral of the first kind and modulus k of the Jacobi's elliptic functions is given by the transcendental equation

$$k = \frac{d}{2\xi_P} \frac{1}{K(k)}. \tag{5}$$

Let us note that the magnitude of the modulus k determines the type of solution: for $k \to 0$ we are in the plane-wave regime whereas for $k \to 1$ we are in the soliton regime. We have therefore a crossover from the plane-wave modulation to the solitonlike modulation of the phase of the order parameter in splayed cells. For small electric polarization, the polarization coherence length is very large and we are in the plane-wave regime because $k \to 0$. This means that there is no internal electrostatic field to impose a distortion of the director field. On the other hand, for a very high spontaneous polarization, the polarization coherence length is very small and $k \to 1$. This means that there is a very strong internal electrostatic field that expels the soliton wall to a region close to the surface and sustains rather uniform alignment in the middle of the cell, as shown in Fig. 1(b).

Let us now consider the dynamics of this soliton structure. The nonequilibrium free energy, which determines the dynamics of soliton-splayed structures, depends on both the

elastic distortion energy and the electrostatic energy. We shall use here the so-called adiabatic approximation, where the local electric potential $\varphi(x,t)$ is at all times *in equilibrium with the director field*. This means that the only source of the electric potential in the system is the electric charge distribution, which arises from the divergence of the polarization field. This approximation obviously breaks down if there are ionic impurities in the sample, which follow the motion of the polarization field and screen the resulting local electric potential.

The nonequilibrium phase profile is

$$\Phi(x,t)=\Phi_0(x)+\Psi e^{-t/\tau}, \tag{6}$$

where Ψ is the amplitude of the phase excitation and the equilibrium phase profile Φ_0 satisfies the sine-Gordon equation (3). The linearized Landau-Khalatnikov equation of motion, as deduced from the nonequilibrium free-energy density, appears in the well-known [21] form of the Lamee's equation of order one:

$$\frac{d^2\Psi}{du^2}+[h-2k^2\mathrm{sn}^2\Phi_0]\,\Psi=0. \tag{7}$$

The eigenvalue h of Lamee's equation is

$$h=k^2\left(1+\frac{\gamma\varepsilon\varepsilon_0}{P_0^2\tau}\right) \tag{8}$$

and determines the relaxation rate of the phase excitation. The eigenfunction, which satisfies the fixed boundary conditions, is selected from the general solutions of Lamee's equation [21]

$$\Psi=\mathrm{sn}(u,k)\quad\text{and}\quad h=1+k^2. \tag{9}$$

The corresponding relaxation rate of the lowest-order mode is

$$\tau^{-1}=\frac{P_0^2}{\gamma\varepsilon\varepsilon_0 k^2}=\frac{4K}{\gamma}\frac{K^2(k)}{d^2}. \tag{10}$$

Let us remember that the transverse wavelength of these excitations is equal to a double cell thickness $2d$, i.e., the corresponding wave vector is $q=\pi/d$. The above expression can also be understood as a "dispersion" relation for solitonlike excitations in a cell with variable thickness and is shown in Fig. 2.

One of the surprising features of the dispersion relation for solitary waves in splayed cells with a variable thickness is the appearance of a gap in the long-wavelength limit, $q\approx1/d^2\to0$. This is similar to the magnetic field and electric field induced gaps in helical phases and is related to the symmetry breaking by the external field [21]. In our case, this symmetry breaking field is the Coulomb field of the induced charges, which induces a solitonlike distortion of the stationary phase profile and drives the dynamics of splayed cells into the solitary-wave regime.

The nature of this phenomenon can be further clarified by considering the limiting values of the dispersion [Eq. (10)]. For zero spontaneous polarization we always have the plane-wave regime and the dispersion is parabolic:
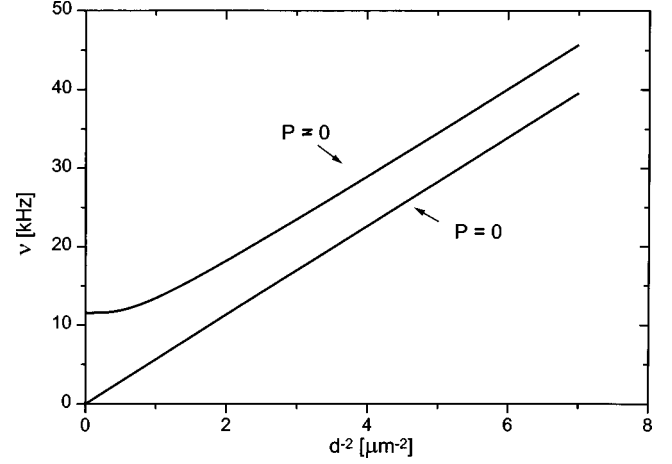

FIG. 2. Thickness dependence of phase excitations in a thin, splayed cell. For zero polarization we have a plane wave limit ($k\to0$) and parabolic dispersion. For large polarization we have a solitonlike limit ($k\to1$) and the appearance of a gap at $q=0$.

$$P_0=0\Rightarrow k=0:\quad\tau^{-1}=\frac{K}{\gamma}\left(\frac{\pi}{d}\right)^2. \tag{11}$$

For a finite polarization P_0 we can have either a plane wave ($k\to0$) or a soliton ($k\to1$) regime, depending on the ratio d/ξ_P. If the thickness of the cell is much smaller than the polarization coherence length, $d\ll\xi_P$, we are in the plane wave regime. The relaxation rates are now renormalized by the spontaneous polarization, which is shown as an additional, thickness independent term in the expression for the relaxation rates:

$$P_0=\mathrm{const},\quad d/\xi_P\to0\quad\text{and}\quad k\to0:$$

$$\tau^{-1}=\frac{K}{\gamma}\left(\frac{\pi}{d}\right)^2+\frac{K}{\gamma}\frac{1}{2\xi_P^2}. \tag{12}$$

On the other hand, for thickness much larger than the polarization coherence length, $d\gg\xi_P$, we have a solitonlike dynamics, which is polarization renormalized. The dispersion has now a finite gap at $q=0$:

$$P_0=\mathrm{const},\quad K(k)\to\infty\quad\text{and}\quad k\to1:$$

$$\tau^{-1}(q=0)=\frac{K}{\gamma}\frac{1}{\xi_P^2}. \tag{13}$$

We see that the presence of a spontaneous polarization always increases the relaxation rates of the excitations. This increase can be interpreted as a renormalization of the elastic constant for bend fluctuations of the director, which represent splay fluctuations of the polarization field. The effect was extensively studied in the past both theoretically [22,23] and experimentally [24]. The elastic constant increases as a square of the spontaneous polarization and this is reflected in the increase of the relaxation rate of phase excitations. This apparent increase of the elastic constant is due to the appearance of a fluctuation-induced electrostatic charge density and a consequent increase of the total free energy of the system due to Coulomb repulsion. This represents an additional ther-

modynamic force, which drives the system back to equilibrium and results in an apparent increase of the liquid crystalline elastic constant.

Let us conclude this section with a brief note on the dynamics of thin ferroelectric liquid crystalline films in the limit of very large measuring fields. In this regime, a switching of a cell takes place, where the electric dipole field is forced to align into the direction of the external electric field. This is a dynamical phenomenon that is substantially different from the intrinsic dynamics discussed so far. The experimental observations [25] and simulations [14] indicate that the switching process in thin ferroelectric liquid crystal cells is of nucleation character and the switching starts at a nucleation site. At this site, the favorable phase is created by the field and then propagates into the unstable phase. The spreading of an energetically favorable state proceeds in a form of a wave front, which propagates into the unstable state. This problem of front propagation into unstable liquid crystalline states has been extensively studied in the past by van Saarlos *et al.* [26], Maclennan *et al.* [14], and others [27]. Maclennan *et al.* [14] have analyzed the nonlinear dynamics of a solitonlike distorted helical smectic structure by numerically solving the equation of motion of the director field for arbitrary electric fields applied in a transverse direction,

$$\eta \frac{\partial \Phi}{\partial t} = K \frac{\partial^2 \Phi}{\partial z^2} - PE \cos \Phi. \qquad (14)$$

Here, the z axis is along the helical axis and the electric field is applied in a transverse direction. They have shown that for very small fields, the response is linear, i.e., the relaxation rate of the excitations is independent of the field and equals $\tau^{-1} = (K/\gamma)q^2$, where q is the wave vector of the elastic distortion. For fields slightly above the threshold for switching, the relaxation rate of excitations grows as $\tau^{-1} = PE/\gamma$ and asymptotically approaches square root dependence $\tau^{-1} \approx \sqrt{E}$, characteristic for solitary wave propagation at very high fields.

III. EXPERIMENTAL RESULTS AND DISCUSSION

In our experiment, we have measured the thickness dependence of the order parameter relaxation rates in the helicoidal and the unwound phases of CE-8. The thickness dependent measurements can be also considered as a measurement of a dispersion relation of order parameter excitations, because the wavelength and correspondingly the wave vector of the eigenmodes is determined by the cell thickness due to the fixed boundary conditions.

The experiment was performed in wedge-type cells, which were nylon coated and undirectionally rubbed with soft velvet. The thickness of the cell was determined by the 4 μm glass spacers on one edge and a close (proximity) contact on the other edge of the glass. Due to the irregularities of the nylon layer, the thinnest part of the cell was approximately 0.3 μm thick. The thickness of each individual cell as a function of the position along the cell was determined by measuring the spectral transmission. A 0.5×2.0 mm^2 slit was used to localize the measuring spot, and it was placed directly on the glass surface of an empty cell. In this way, the
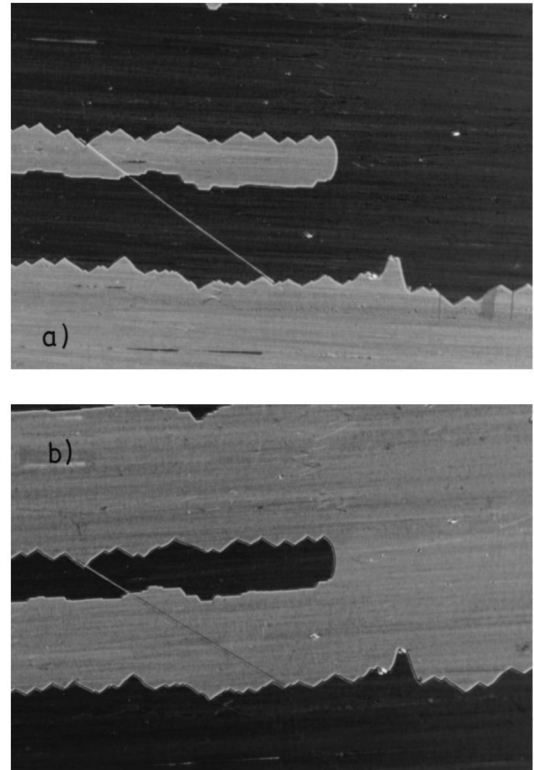

FIG. 3. Photographs of a structure of a ferroelectric liquid crystal CE-8 under a polarizing microscope with polarizer and analyzer in crossed position. The thickness of the sample is ≈ 2 μm. The color of the two kinds of domains depends on the angle between the polarization of the incident light and the rubbing direction. The rubbing direction is nearly horizontal and is observable as a weak modulation of the intensity. (a) polarizer is at $\alpha \approx -3°$ with respect to the rubbing direction. (b) polarizer is at $\alpha = +3°$ with respect to the rubbing direction. Note the inversion of the color contrast of both domains. Field of view: 750×540 μm^2.

local thickness of the cell was determined with an accuracy better than ± 0.05 μm. The resulting wedge angle was of the order of 1.5×10^{-3} and had a negligible effect on the experiment.

After cooling down into the ferroelectric phase, a stripelike texture was observed under a polarizing microscope with cross polarizers. For large thickness, $d > 1$ μm, we could clearly identify two different kinds of stripe-shaped domains with different colors. The domains were rather long, whereas their lateral dimension was of the order of several tens of micrometers. The color of a single domain depended on the orientation of the polarization of the incident light and changed from blue to yellow when the angle of the input polarization was changed from $\alpha \approx -3°$ to $\alpha \approx +3°$ with respect to the rubbing direction. This is illustrated in the two photographs in Figs. 3(a) and 3(b) and suggests that we have a splayed structure, which is a result of a strong polar coupling of the molecules to the nylon surface. A similar kind of structure was discussed, for example, by Maclennan *et al.* [28] and was identified as a splayed chevron structure. The two kinds of domains correspond to either left or right handed twisting of the in-plane director throughout the cell, which both represent splayed polarization field. This was further confirmed by the application of a strong electric field,

which clearly induced an increase of the ''apparent'' tilt angle, thus ''unwinding'' the internal twist of the in-plane director.

The stripe texture disappeared for thicknesses smaller than 1 μm and was replaced by an irregular arrangement of domains of several hundred micrometer sizes. However, even for the lowest thickness of 0.3 μm the angle of maximum color contrast of these domains was close to the rubbing direction. This indicates that the splayed states are still energetically favorable even for such a small thickness because of very strong polar boundary conditions.

In our linear response experiment (which is better known as the ''electroclinic'' experiment, when performed in the smectic-A phase) one measures the real (in-phase) and imaginary (out-of-phase) parts of the linear electro-optic response $\chi(\omega)$ of a sample to a small measuring electric field $\vec{E}=(E_0,0,0)e^{i\omega t}$. This field couples to those collective eigenmodes of the system that have a finite space average of fluctuating electric polarization $\langle \delta \vec{P}(\vec{r},t) \rangle$. Because of the interconnection between the polarization and the director field, this results in a finite value of the space-averaged director field $\langle \delta \vec{n}(\vec{r},t) \rangle$. The dielectric tensor for the optical frequencies ε_{ij} has the same symmetry as the tensor $\bar{n} \otimes \bar{n}$, and the change $\langle \delta \vec{n}(\vec{r},t) \rangle$ will be reflected in the dielectric tensor field. For small external electric fields and consequently small local distortions of the director field, the change of the dielectric tensor ε_{ij} can be expanded in terms of the excess polarization. It is straightforward to show that in the case of a mirror-symmetric director field in a splayed cell, the field induced change of the dielectric tensor is

$$\langle \delta \underline{\varepsilon} \rangle \approx \begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & \langle \delta P_x \rangle \\ 0 & \langle \delta P_x \rangle & 0 \end{bmatrix} \quad (15)$$

and is therefore linear in the electric field. This change of the dielectric tensor can be detected with a suitable optical technique. It is therefore clear that in a linear electro-optic experiment we detect polar eigenmodes of the system, i.e., the dielectric eigenmodes. The method is therefore an optical analog to dielectric spectroscopy.

The linear electro-optic response of wedge-type samples was measured at different thickness and different temperatures and we shall here present the results of the measurements below the phase transition into the ferroelectric phase. The measurements in the smectic-A and close to this phase transition will be reported elsewhere.

In the ferroelectric phase we observe for all thicknesses two relaxation modes, a high- and a low-frequency one, as shown in Fig. 4. We identify them as two phase modes, i.e., the fluctuations of the phase of the director field, and the reason for this will be clear from what follows. The first mode has relatively very high relaxation rates in the kHz region. It is nearly temperature independent except very close to T_c, and shows a very strong thickness dependence. The relaxation rate of this phase mode, as observed in CE-8 filled wedge-type cells in the splayed ferroelectric smectic-C^* phase is shown in Fig. 5 as a function of $1/d^2$. The relaxation rate was here determined from the maximum of the imaginary part of the signal. One can see that, in general, the relaxation rates of this mode increase as $1/d^2$ by decreas-
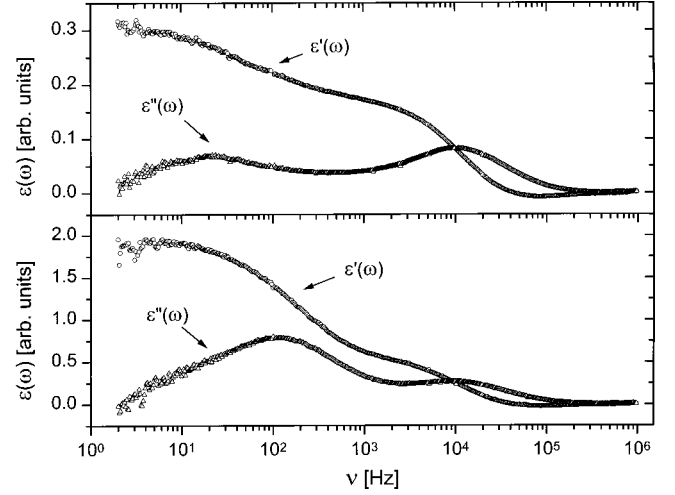

FIG. 4. Real and imaginary parts of the linear electro-optic response in the smectic-C^* phase of CE-8 at $T_c-T=3$ K. The thickness of the cell is 0.7 μm, and the measuring field is $E_0=0.15$ V/μm for the upper and $E_0=0.6$ V/μm for the lower curves. The relaxation rates of the two excitations were determined from the maximum of the imaginary part of the response.

ing the cell thickness. This is therefore the so-called ''thickness'' mode, first reported by Glogarova [16] and corresponds to transverse excitations (i.e., perpendicular to the confining surfaces) of the director phase. The experimental data, however, reveal fine details in this thickness dependence. For small thickness (large $1/d^2$), the relaxation rate follows the predicted $1/d^2$ dependence, as indicated by the upper solid line in Fig. 5, but is shifted to higher frequencies. If this part of the data is extrapolated to $1/d^2 \approx 0$, one can clearly observe a frequency gap of approximately 10 kHz at $1/d^2=0$. There is, however, an obvious crossover at interme-
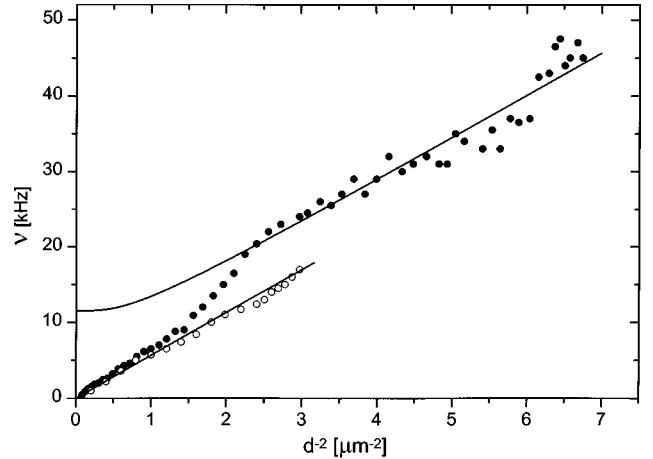

FIG. 5. Relaxation frequency of the fast transverse phase mode (thickness mode) in the smectic-C^* phase of chiral CE-8 in ultrathin cells with splayed phase profile (full dots). Empty dots represent thickness dependence of the relaxation frequency for the transverse phase mode in a mixture of 90% of racemic and 10% of chiral CE-8. The solid lines are the best fit to the Eq. (10) with $K=4.1 \times 10^{-11}$ N, $\varepsilon=3.2$, $P_0=5.1 \times 10^{-5}$ A s/m^2 and $\theta_0=20°$ for the chiral sample and $P_0=0$ for the mixture. A crossover in the dynamics is clearly visible at $d^{-2} \approx 2$ μm^{-2}, which corresponds to a thickness of 0.7 μm.

diate thickness of approximately 1 μm. Here, the observed data approach a line through the origin of the coordinate system, so that there is in fact no frequency gap at $1/d^2 = 0$.

The dynamics at small thickness can be interpreted within the predictions of the theory of solitonlike dynamics. The upper solid line in Fig. 5 represents the best fit to Eq. (10), using the physical parameters that are known [29] for chiral CE-8. The agreement with theory is fairly good for thickness smaller than approximately 0.7 μm. For larger thickness, the data obviously deviate from the predictions of the solitonlike dynamics. This indicates the presence of a physical mechanism that ''screens out'' the induced charges due to the fluctuations of the polarization field. The assumption of a screening mechanism was further confirmed by measuring the thickness dependence of the relaxation rate of this thickness mode in a mixture of a pure and racemic CE-8, which has ten times smaller spontaneous polarization than pure CE-8. The data are shown in Fig. 5 and clearly indicate that there is no detectable renormalization of the relaxation rates in this sample because the spontaneous polarization is very small.

The observed crossover can be explained by the presence of ions, which screen out the electrostatic field in a liquid crystal. The ions follow the collective excitations of the dipolar field in a liquid crystal and screen out the Coulomb field, which is generated by the splaylike excitations of the polarization. If the thickness of the cell is much smaller than the Debye screening length, the screening will not be efficient and we will have a solitonlike, polarization-renormalized dynamics. On the other hand, for large thickness, the screening will be efficient and we will have a plane-wave-like dynamics with no polarization renormalization of the relaxation rates. The expected mobility of the ions is $\mu = d/(\tau E)$, where d is of the order of a Debye screening length, $d \approx 10^{-7}$ m, τ is the relaxation time and E is of the order of the local electric field $P_0/(\varepsilon\varepsilon_0) \approx 1$ V/1 μm. This gives an ionic mobility of $\mu \approx 5 \times 10^{-10}$ m^2 V^{-1} s^{-1}, which is in reasonably good agreement with other experimental data [30].

The relaxation rate of the low-frequency mode is of the order of 20–30 Hz and is nearly independent of the cell thickness. For low measuring fields, the amplitude of this mode is very small and the mode is observable only as a low-frequency ''shoulder'' of the imaginary part of the signal, as shown in Fig. 4(a). By increasing the measuring field, the amplitude of this mode grows first linearly with field, whereas the relaxation rate remains unchanged, as expected for a linear regime. At some critical field E_c, the relaxation rate of this mode starts to increase with a driving field, whereas the amplitude grows nonlinearly. Finally, in the limit of very strong fields, this mode represents a switching of the polarization field over the entire cell. This is noticed by observing the cell under a polarizing microscope and using a very low frequency of the applied electric field. A similar mode was observed recently by Panarin et al. [18].

The observation of *two relaxation modes* in thin cells of ferroelectric liquid-crystalline material has some implications for our understanding of the onset of switching in thin ferroelectric cells. It tells us that there are two independent collective motions, which can couple to an external electric field, applied to a thin ferroelectric liquid-crystalline layer.
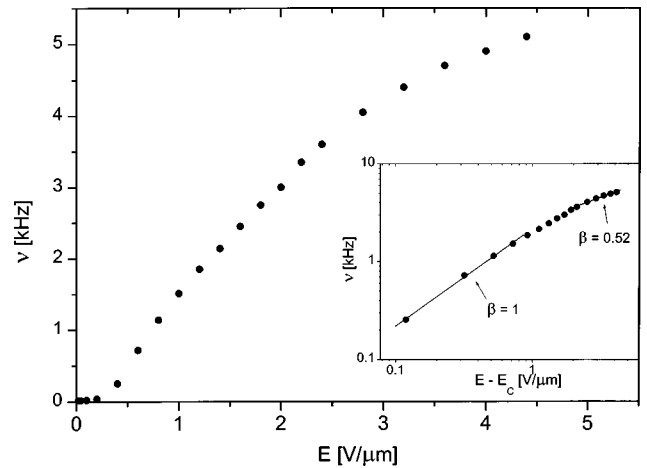

FIG. 6. Relaxation frequency of the slow phase mode as a function of a driving electric field in the smectic-C^* phase of CE-8 in ultrathin splayed cells. Here $d = 0.5$ μm, $T_c - T = 1.5$ K and the threshold for switching is $E_c = 0.28$ V/μm. The inset shows the same data in the log-log scale.

Whereas the first kind of motion is strongly thickness dependent and field independent, the second kind of motion is thickness independent, but depends strongly on the applied field. This suggests that the first motion is just the excitation of transversely confined splayed ferroelectric smectic states, reminiscent of soliton dynamics, whereas the second motion is a mode that gives rise to the switching of polarization of the cell as a whole. In this sense, we conjecture that in the linear regime, the observed low-frequency mode represents small fluctuations of the director field around the nucleation site. In view of rather low frequency of this excitation, it is clear that the site is elastically very weakly coupled to the overall structure. Using a relation $\tau^{-1} = (K/\gamma)q^2$ we get an estimate for the wavelength of this collective excitation of the order of ≈ 10 μm, which is of the order of the domain width. The excitation would then correspond to a slow movement of the domain walls, interacting over a length of several micrometers.

Our measurements clearly indicate nucleation and a crossover from the intrinsic dynamics of a slow mode at small measuring fields to the solitonlike dynamics of this mode at large fields. The dependence of the relaxation rate of the slow mode as a function of the amplitude of a measuring electric field is shown in Fig. 6 for a temperature close to the smectic-A–smectic-C^* phase transition. The relaxation rate is first constant up to a certain threshold electric field and then starts to increase with a driving field. Slightly above the switching threshold, the relaxation rate increases linearly with a driving field and approaches $\sqrt{E}$ dependence at very high fields. The threshold field therefore corresponds to the onset of nucleation of an energetically favorable state and the field dependence of the relaxation rates is in good qualitative agreement with theory. At lower temperatures, the threshold for switching is not so well defined and the critical field increases considerably.

IV. CONCLUSIONS

In conclusion, we have observed two independent order parameter relaxations in ultrathin ferroelectric smectic-C^*

layers. They have been identified as the two phase modes: the first one corresponds to transverse excitations of the phase, pinned by fixed boundary conditions to the bounding walls. These excitations exhibit strong thickness dependence, they are virtually independent of the measuring field, and show a solitonlike character for very small thickness. The second type of relaxation is closely related to the nucleation of switching. This motion is very slow for low measuring fields and clearly shows a threshold field where the intrinsic linear dynamics changes into the collective, nonlinear reori-

entational dynamics of the polarization of a cell as a whole. The fact that this branch of excitations emerges continuously and is present even at very small fields indicates that the nucleation sites for the switching dynamics are inherently present in the structure. We conjecture that these sites are the domain walls, which are observable in our samples. This leads to a conclusion that the switching process is most likely to nucleate at the interface of two domains. The switching seems to proceed throughout the sample via the bulk reorientation with an indication of solitary-wave character at very high strengths of applied electric fields.

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