COMMENTS

Comments are short papers which criticize or correct papers of other authors previously published in the **Physical Review.** *Each Comment should state clearly to which paper it refers and must be accompanied by a brief abstract. The same publication schedule as for regular articles is followed, and page proofs are sent to authors.*

Comment on "Kink switching in ferroelectric free-standing films with high spontaneous polarization"

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(Received 16 May 1997; revised 14 November 1997)

We discuss experiments by Demikhov *et al.* [Phys. Rev. E **52**, 6250 (1995)] on switching dynamics of freely suspended ferroelectric films. The authors have studied the optical response to electric excitation by an in-plane ac field. They perform a theoretical analysis which is not appropriate for the experimental data. We reanalyze the optical response of the film to an oscillating electric field and interpret the experimental data in terms of a uniform azimuthal reorientation of the *c* director (Goldstone mode). Effects of the helical pitch on the interpretation of the experimental results are discussed. We think that the data provide no convincing evidence for the assumption of a "kink-switching" mechanism. [S1063-651X(98)08011-8]

PACS number(s): 64.70.Md, 68.15.+e

INTRODUCTION

The equations which describe the dynamical behavior of chiral ferroelectric S_C^* material in electric fields are well established (see, e.g., [2]). Nevertheless, the mathematical analysis of the switching processes in thin sandwich cells under the action of electric fields is rather difficult because of the complex geometry even in a simple bookshelf sample with planar surface anchoring (see, e.g., [3]). Freely suspended films with lateral electrodes provide a simple experimental geometry. They can be studied by transmission or reflectivity measurements under the influence of in-plane oscillating or rotating electric fields (e.g., [4–10]). The analysis of the frequency dependence of the electrical, optical, or IR response yields information on the switching dynamics and viscous coefficients [11,12].

Demikhov *et al.* [1] report optical investigations of thin freely suspended S_C^* films. The authors have recorded the optical reflection of the films at normal incidence while the film is exposed to a lateral oscillating electric field. As the change of optical contrast in the reflection texture is very weak, the optical data are sampled by means of a lock-in amplifier at the frequency of the applied ac field. The frequency dependence of the optical signal is used to confirm a proposed switching mechanism in the S_C^* phase.

The authors observe a rather low optical signal at high and low frequencies but detect a dramatic increase of their optical signal in the intermediate frequency range, which is reminiscent of a resonant behavior. Such an increase of the optical response with increasing frequency of the driving field seems to be incompatible with the relaxational character of the Goldstone mode. In order to explain this peak in the optical response and its field strength and film thickness dependence, the authors have proposed a so-called kink switching (KS) model. According to that model, the *c* director (the *c* director denotes the projection of the preferred molecular orientation in the tilted S_C^* phase on the layer plane) reorientation occurs in the form of the propagation of a solitary wave ("kink") in the film. Different propagation directions are proposed for thin and thick films. Demikhov *et al.* [1] assume that in the films studied, a lateral propagation of kinks occurs, starting from certain domain boundaries. Their calculations are performed for a different geometry but can be modified correspondingly.

However, no direct evidence for the proposed kink propagation is given, the model is based only on integral optical reflection measurements. For interpretation of their experiment Demikhov *et al.* calculate a scattering intensity which is the time and space average of some correlation function of the *c*-director components. However, they neglect the fact that their lock-in amplifier suppresses exactly this time average but measures a reflectivity modulation at the first harmonics of the excitation field. Therefore the theoretical curves in [1] do not describe the experiment. We reconsider the data analysis and it turns out that the apparent resonance peak is a simple consequence of the optical detection method. Most features of the experimental curves can be interpreted without the concept of solitary waves.

We address two points in this comment. First we propose a correct description of the observed optical data. Second we compare the effects expected from a uniform *c*-director reorientation and the kink-switching model. We analyze other factors which are relevant for the experiment and discuss which evidence for the kink-switching model is actually contained in the data.

THEORY

We consider first a description of the experimental data in [1] with the simple model of a uniform *c*-director reorientation in the layer plane, driven by the electric field and damped by a viscous term. The qualitative predictions are as follows: In the described experiment, the time dependent director response to ac excitation is not measured directly. The lock-in signal sampled at the first harmonics represents only a special part of the dynamic response, viz., the contribution of the respective mode in a time Fourier expansion of the optical signal. An oscillating electric field of frequency ω is applied in the plane of a free-standing smectic film. At low ω, the reorientation of the sample is very fast compared to the excitation period $T=2\pi/\omega$, and the switching process occurs during a very short interval in each half cycle of the driving field. The optical reflectivity curves are modulated only during these short switching periods; most of the time the spontaneous polarization is (almost) aligned to the field. Only narrow peaks appear in the time dependent optical response function, and the first harmonics of such curves are correspondingly small. Higher harmonics of the driving frequency are excited, but filtered out by the lock-in amplifier. When the increasing ac frequency ω approaches the characteristic frequency of the Goldstone mode, the response signal becomes more sinusoidal and the first harmonics in the Fourier expansion increase. This leads to an increased optical contrast near the relaxation frequency when data are sampled with the lock in. At further increasing ω, viscous friction becomes dominant, reorientation of the c director remains incomplete, and the response signal modulation gradually decays to zero. The similarity of increased lock-in signal near the critical frequency to a resonance curve of a linear oscillator phenomenon is only superficial. The c-director response to the oscillating driving field is strongly nonlinear particularly at low frequencies.

In order to put these considerations on a quantitative level, we analyze the underlying mathematical equations. We consider a spatially uniform sample and assume that the film is sufficiently thin such that the S_C^* helix is unwound. The azimuthal reorientation is described by [13]

$$\gamma\theta^2\dot{\varphi}=-PE\sin\varphi+\epsilon_a\epsilon_0E^2\sin\varphi\cos\varphi, \tag{1}$$

with time dependent electric field $E(t)$, spontaneous polarization $P=\mu\theta$, rotational viscosity γ, and in-plane dielectric anisotropy ϵ_a, the azimuthal angle φ (between $\vec{P}$ and the direction in which the electric field is applied), and the tilt angle θ ($\sin\theta\approx\theta$) of the S_C^* phase. Equation (1) can be written in the simplified form

$$\dot{\varphi}=\omega_0(-\sin\varphi\cos\omega t+\epsilon\sin\varphi\cos\varphi\cos^2\omega t) \tag{2}$$

by substitution of $\omega_0=PE_0/\gamma\theta^2$ and $\epsilon=\epsilon_a\epsilon_0E_0/P$. Solutions of Eq. (1) for triangular or square wave electric fields are discussed in literature [10,11]. For a sinusoidal electric field $E(t)=E_0\cos\omega t$, we will first neglect the dielectric term $|\epsilon|\ll|1|$. (This condition is usually fulfilled in S_C^* films.) The analytical solution of Eq. (2) with $\epsilon=0$ for an initial orientation $\varphi(0)=\varphi_0$,

$$\tan\frac{\varphi}{2}=\tan\frac{\varphi_0}{2}\exp\left(-\frac{\omega_0}{\omega}\sin\omega t\right), \tag{3}$$

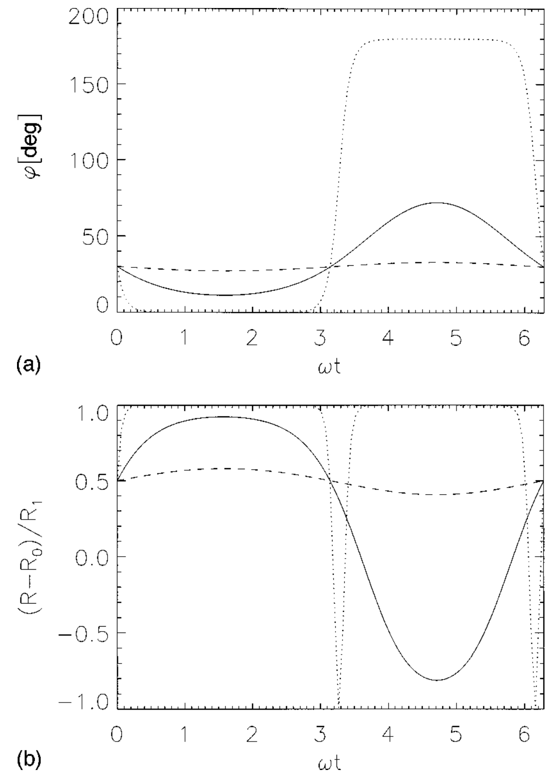

FIG. 1. Characteristic curves for the variation of (a) the azimuthal angle $\varphi(t)$ and (b) the resulting response signal $R(t)$ with $R_0=0$, $R_1=1$. The initial angle φ_0 was chosen to be 30°. Low frequency $\omega/\omega_0=0.1$ (dotted); $\omega/\omega_0=1$ (solid); high frequency $\omega/\omega_0=10$ (dashed).

describes the azimuthal reorientation in an oscillating electric field. It is strictly periodic with $T=2\pi/\omega$; after each period T the c director is at its initial orientation again.

For a description of the optical reflectivity, IR dichroism, or similar experimental observables of thin films we introduce the generalized periodic function

$$R(\varphi)=R_0+R_1\cos2\varphi,$$

which we will call response function (the choice of modified functionals is trivial). It reflects the relation between the observed time-periodic signal and director azimuth. Inserting Eq. (3), we find

$$R(t)-R_0=R_1\left(1-\frac{8y(t)^2}{[1+y(t)^2]^2}\right),\quad\text{with}\quad y(t)=\tan\frac{\varphi(t)}{2}. \tag{4}$$

Figure 1(a) presents three typical curves $\varphi(t)$ at different frequencies, calculated from Eq. (3), together with the corresponding $R(t)$ obtained from Eq. (4) in Fig. 1(b). In both figures, we have rescaled the time axes with $T=2\pi/\omega$ in order to emphasize the changes in the curve shape. Relevant for the discussion of the experimental data is the first harmonics $|\int R(t)\exp(i\omega t)dt|$ of the curves in Fig. 1(b). We have extracted the frequency dependence of this coefficient numerically by Fourier transformation of the analytical $R(t)$ at different ω, the result is shown in Fig. 2. The frequency dependence is in accordance with our qualitative discussion above. The general trend is a pronounced increase of the

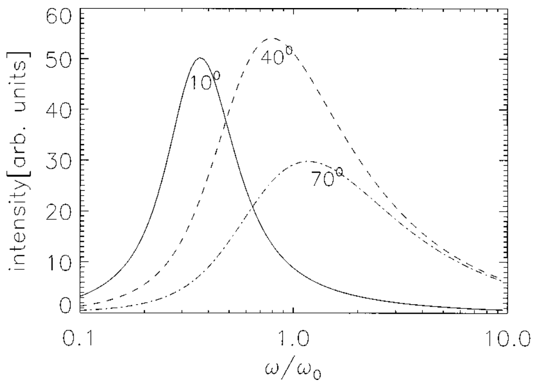

FIG. 2. Frequency dependence of the first harmonics of the response signal $R(t)$ for different φ_0, ω is given in units of ω_0. The choice of different ω_0, i.e., the change in the relaxation time of the sample, simply shifts the curves on the frequency axis.

signal of the first harmonics at frequencies in the vicinity of the Goldstone mode $\omega_{\mathrm{Goldstone}}=\omega_0$. The exact shape of the optical curves depends upon the initial orientation φ_0 as seen in Fig. 2. The signal of a sample which is not monodomain will be a superposition of signals for different $\varphi(0)$. The overall curve shape will be reproduced but details average out and the peak position may change slightly.

A refined model has to consider also the small dielectric interactions due to $\epsilon\neq 0$. The inclusion of the dielectric term leads to a gradual sample alignment. We have solved Eq. (2) for $|\epsilon|\ll 1$ by numerical integration. The reorientation in each field cycle is almost the same as in Fig. 1(a), but with a small drift of the angle φ after each cycle. This case leads theoretically to a final stable solution $\varphi=0$ (or $\varphi=\pi$), and a vanishing lock-in signal. The case $\epsilon_a>0$ finally leads to a symmetric oscillation about $\varphi=\pi/2$, again with the first harmonics of the signal disappearing. In practice, the dielectric interactions are very weak though ($\epsilon<10^{-4}$) many competing effects of walls, defects, film boundaries, and electric field inhomogeneities will influence the c director and prevent a complete sample alignment at $\pi/2$ or 0. In particular, an existence of helical twist along the film normal will counteract the overall alignment $\varphi=0$.

DISCUSSION

In spite of some unknown parameters we will now try a quantitative comparison with data in the paper on which we are commenting. The interpretation of the nature of the optical signal is not problematic. In [1], the description is based on the assumption of light scattering. We think that the azimuthal dependence of the film reflectivity produces the intensity modulation, but for the following discussion this point is peripheral.

In [1], the zero frequency scattering intensity is computed, but light modulation at the frequency of the driving field is measured. There is no indication how both quantities are correlated. Consequently, the qualitative agreement of simulated curves in [1] with the experiment is only coincidental (cf. Figs. 1, 9, and 10 there), and not very satisfactory either. The fit in Fig. 11 of [1] considers only a very small frequency range, and the result $I\propto G(\omega+B)^{-3}$ is not in good accordance with the theoretical prediction $I\propto\omega^{-3}$.

When we compare our simulations with the experimental curves in [1], the overall qualitative agreement (see our Fig. 2 and Fig. 1 in [1]) seems to be much better. Quantitative comparison shows that the intensity peak in our model appears near ω_0 (which is equal to $a\approx 10^4$/s in the notation of Demikhov *et al.*). This corresponds quite well to the experimental result $f\approx 1500$ Hz in Fig. 6 of [1]. In contrast, the kink-switching model yields the intensity peak at $\omega_{\mathrm{ext}}\approx a/10=10^3$/s (see Discussion in [1]) which is too small by one order of magnitude (this is independent of the discrepancies in the optical analysis). We think this is some evidence against KS, and we note that from two competing reorientation mechanisms in a system one should expect that only the faster process (viz., the uniform Goldstone reorientation) is effective.

The rest of the discussion in [1] focuses on the field strength and film thickness dependence of the observed peak frequency. However, in their argumentation the authors completely neglect the existence of a helical twist along the film normal. For the discussion of details of film thickness and frequency dependence of ω_{ext} we have to consider that the free pitch $p_0=2\pi/q_0$ of the material is about 300 nm [14], and a 300 layer film is $\approx 4p_0$ thick (with a smectic layer thickness of roughly 4 nm [14]). At low E_0, the c director forms a helix along the film normal, and the electric field to unwind the helix of about $K_{\mathrm{twist}}\theta^2q_0^2/2P$ is of the order of 10^5 V/m, i.e., rather large on the field strength scale of Fig. 3 in [1]. In fact the elastic term $K_{\mathrm{twist}}\theta^2q_0^2$ is several orders of magnitude larger than the in-plane elastic distortion in the kink. The assumption of an untwisted sample may be justified only at very high electric fields.

The electric field effects on a partially unwound helix are complex, but one can predict the general trend: an increasing optical intensity modulation with progressing helix unwinding, i.e., with increasing E_0. This explains the left hand side of Fig. 3. The dependence on the right hand side of Fig. 3 may be due to the increasing influence of the dielectric term $\propto E_0^2$ which brings the angle φ on average closer to 0°, the first harmonics consequently becoming smaller. The observed film thickness dependence for films between 150 and 400 layers has to be discussed in the light of the different ratios p_0/d. If the films thickness d is a multiple of p_0 (152 layer film) the net polarization at each position in the plane is compensated while for half integer thickness $d\approx 2.5p_0$ (182 layer film) an uncompensated portion of the helix remains. The authors seemingly have not studied the helicity of their films, therefore a detailed discussion of film thickness effects is speculative here. The same holds for the discussion of the field strength independent relaxation frequency presented in Fig. 3 of [1].

CONCLUSIONS

The kink-switching model was originally developed for nonferroelectric S_C cells [3]. In the original application by Schiller the smectic is contained in a thin cell with planar anchoring at the glass plates. The electric reorientation there is of the Fréedericksz type. Due to the degeneracy of the dielectric interaction potential, domains of opposite c-director reorientation may form, separated by walls. These walls can propagate in the cell plane. The bistability of the switched state in S_C is important for the appearance of the kink. Schiller *et al.* [3] have also extended their model to ferroelectric S_C^* samples. In that case, a superimposed large high frequency ac field is employed to maintain the bistability and the dielectric term is dominating.

On the contrary, in S_C^* films with high spontaneous polarization as studied in [1], the polar interaction of $\vec{P}$ with the electric field dominates by far, all other terms including the dielectric interaction ($\epsilon < 10^{-3}$) are negligible, and only one stable orientation exists in the electric field. Therefore the formation of kinks should be rather unlikely. Even if some comparable mechanism exists in the films studied, the paper gives not a single unambiguous evidence for that effect.

Certainly the paper commented on is a valuable contribution to study of the dynamics of freely suspended films in electric oscillating fields. The experimental data provide useful information on the *c*-director dynamics. If we assume that the *c* director in free-standing S_C^* films exposed to laterally oscillating electric fields reorients uniformly in a Goldstone mode, the frequency characteristics as observed by Demikhov *et al.* [1] are well reproduced. A quantitative comparison of peak frequencies seems to support our model. Details of the field strength and film thickness dependence of the relaxation peak frequencies and intensities are difficult to interpret as the phenomena were reported only in films thicker than the free pitch of the substance. Therefore all conclusions drawn from the electric field and film thickness dependence have to take helical twist into consideration.

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[13] $\vec{P}$ is perpendicular to the *c* director in the film plane. We define ϵ_a as the difference of the dielectric permittivities along the *c* director and perpendicular to it in the layer plane. This has consequence for the sign of ϵ_a compared to [1] in Eqs. (1), (6), etc. We have also corrected the sign of the first term in the equation, which is, however, not essential for the comparison of both models.

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