
**Liquid Crystals**
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713926090

# Dielectric response of ferroelectric*$_C$ liquid crystals in a bias electric field

B. Žekš[a,b]; T. Carlsson[c]; I. Muševič[b]; B. Kutnjak-Urbanc[b]
[a] Institute of Biophysics, Medical Faculty, Ljubljana, Slovenia [b] J. Stefan Institute, Ljubljana, Slovenia [c] Institute of Theoretical Physics, Chalmers University of Technology, Göteborg, Sweden

**To cite this Article** Žekš, B. , Carlsson, T. , Muševič, I. and Kutnjak-Urbanc, B.(1993) 'Dielectric response of ferroelectric*$_C$ liquid crystals in a bias electric field', Liquid Crystals, 15: 1, 103 — 111
**To link to this Article: DOI:** 10.1080/02678299308027842
**URL:** http://dx.doi.org/10.1080/02678299308027842

LIQUID CRYSTALS, 1993, VOL. 15, NO. 1, 103–111

# Dielectric response of ferroelectric $S_C^*$ liquid crystals in a bias electric field

by B. ŽEKŠ†‡, T. CARLSSON§, I. MUŠEVIČ‡ and B. KUTNJAK-URBANC*‡

†Institute of Biophysics, Medical Faculty, Lipičeva 2, 61105 Ljubljana, Slovenia

‡J. Stefan Institute, Jamova 39, 61111 Ljubljana, Slovenia

§Institute of Theoretical Physics, Chalmers University of Technology, S-412 96, Göteborg, Sweden

*(Received 10 September 1992; accepted 23 February 1993)*



The dielectric response of the ferroelectric chiral smectic C phase in a bias electric field is studied theoretically within the Landau model using the constant amplitude approximation. It is argued that the response consists of two modes, one of which is related to the unwinding of the helix (the unwinding mode), whereas the other mode is associated with the distortion of the polarization distribution at constant pitch of the helix (the Goldstone mode). The relaxation frequency of the unwinding mode ($f_U$), which is inversely proportional to the square of the sample dimension along the helical axis, is estimated to be of the order of $10^{-4}$ Hz for a sample of length $l \approx 1$ mm along the helical axis. Consequently, in dielectric experiments performed usually at frequencies higher than $f_U$, only the Goldstone mode contribution is detected. In this way, we can explain the behaviour of the measured static dielectric susceptibility which goes to zero as the critical field is approached from below, although the model predicts that the total static dielectric susceptibility, being a sum of the Goldstone mode and the unwinding mode contribution, diverges at the critical field.

## 1. Introduction

In the frequency range below 1 MHz, the dielectric response of ferroelectric chiral smectic C ($S_C^*$) phases consists of two contributions [1, 2]. The soft mode part of the response has an origin in the change of the tilt angle of the molecules, i.e. the change of the amplitude of the order parameter, when a homogeneous electric field is applied perpendicular to the helical axis. The Goldstone mode part, on the other hand, is related to the change in the direction of the tilt, i.e. to the change in the phase of the order parameter. As the change in the tilt of the molecules in tilted smectic phases is energetically expensive [3] except very close to the $S_A \leftrightarrow S_C^*$ phase transition temperature $T_c$, the soft mode dielectric response is small and can be resolved [1, 4] from the Goldstone mode only at temperatures very close to $T_c$ ($T_c - T \lesssim 1$ K). At lower temperatures, the contribution to the dielectric response from the soft mode is negligibly small and only the Goldstone mode contribution is observed in dielectric measurements.

The Goldstone mode contribution is observed also in dielectric measurements in a bias electric field. Excluding a temperature interval $\sim 1$ K below $T_c$, this contribution is the dominant one and this is the case we study in this paper. Studies of

*Author for correspondence.

dielectric properties in a bias field are rare in the literature and only a few reports [5–8] of this type of experiment exist. The general feature of these experiments, performed at low but finite frequencies, is that the Goldstone mode dielectric strength seems to decrease as the bias electric field is increased, going towards zero as the critical unwinding field is approached.

In a recent paper [9] we have presented a theoretical study of the phason contribution to the static dielectric susceptibility in a bias electric field. In this calculation we have shown that the phason mode dielectric strength is expected to increase with the bias electric field, diverging as the field approaches the critical unwinding field. This is in apparent contradiction to the experimental results [5–8]. Here we show that the experimental and theoretical results are consistent if we interpret both results in a proper way.

The outline of this paper is as follows. In § 2, we introduce two modes that contribute to the phason part of the dielectric response of the system, i.e. the Goldstone and the unwinding mode. The relaxation frequency of the unwinding mode at zero field is estimated in § 2.1 and compared to the Goldstone mode frequency. The corresponding dielectric strengths are evaluated as functions of a bias electric field in sub-section 2.2. In conclusion, a brief summary of the results is brought forward and the explanation of experimental dielectric results is given on the basis of the model.



## 2. The Goldstone mode and the unwinding mode

In an unperturbed $S_C^*$ liquid crystal, the molecules are tilted and they precess helicoidally as we go from one smectic layer to another. The spontaneous polarization $\mathbf{P}=(P_x, P_y)$, which lies in a smectic plane and is perpendicular to the director, thus precesses in a similar way. Choosing the $z$ axis as the layer normal, we describe the unperturbed system as

$$P_x = -P_0 \sin \Phi(z), \qquad P_y = P_0 \cos \Phi(z), \tag{1}$$

where $\Phi(z)=q_0 z$ and $q_0=2\pi/p_0$ is the wave vector which corresponds to the pitch $p_0$ of the unperturbed helix, and $P_0$ is the spontaneous polarization. The average spontaneous polarization is determined by the average of $\cos\Phi(z)$ and of $\sin\Phi(z)$ and is equal to zero. By applying an electric field in the $y$ direction, the induced polarization appears in this direction, which is calculated by averaging of $\cos\Phi(z)$. The electric field distorts the pure helicoidal structure, so that the phase $\Phi(z)$ is a more complicated function, describing the tendency of the polarization to align along the field.

In our further discussion, we refer to the previous paper [9], where we have studied the influence of the bias field on the pitch of the helix and on the induced polarization and where we have evaluated the static susceptibility as a function of a bias field. Let us consider the response of the helical system, if we increase the bias field by a small amount. The additional field tends to orient the spontaneous polarization in the field direction and the response of the system can be separated into two processes. The first one corresponds to the polarization reorientation at constant pitch of the helix, whereas the other one is associated with changes in the the helix. The wave vector of the former response is large (equal to the wave vector of the helix) and the corresponding process is expected to be rather fast. The rate of the latter process, which is related to pitch changes, is associated with the length of the sample along the helical axis. The corresponding wave vector is therefore small and the process is expected to be slow compared to the former.

We have therefore introduced two modes which contribute to the dynamic response of the helicoidal system in a bias electric field. The question arises, what is the contribution of both modes to the dielectric response of the sample? Dielectric experiments are performed at low but finite frequencies, usually not lower than about 10 Hz. In § 2.1, the pitch relaxation rate is estimated and it is shown that the relaxation frequency is much lower than 10 Hz. As a consequence, the frequencies used in a dielectric experiment are too high to change the pitch. Hence only the fast mode can be observed in dielectric measurements and it is usually named the Goldstone mode, in analogy to the zero field case. By the name unwinding mode, we will denote the slow mode associated with pitch changes. This mode is usually not observed dielectrically, but is expected to contribute to the dielectric relaxation because of the linear response of the pitch to a small perturbing field in the presence of the bias field.

### 2.1. *Estimation of the relaxation time of the unwinding mode*

In this sub-section, the relaxation rate $f_U$ of the unwinding mode is estimated at zero bias field. We assume that we have a sample of length $l$ along the helical axis and that the molecular director is free to rotate at both boundaries, at $z=0$ and at $z=l$. We are interested in such a fluctuation of the phase which changes the pitch of the helix and costs the least energy. The amplitude of this fluctuation is maximal at both boundaries, and is equal to zero in the middle of the sample. Its wavelength is therefore $\lambda=2l$. The corresponding dynamical process is associated with the wavevector $q=q_0\pm\pi/l$, (see figure 1), whereas the Goldstone mode deformation is related to the wave vector $q=0$. As the phason dispersion is parabolic with the minimum at the critical wave vector $q_0$, at which the phason relaxation frequency is

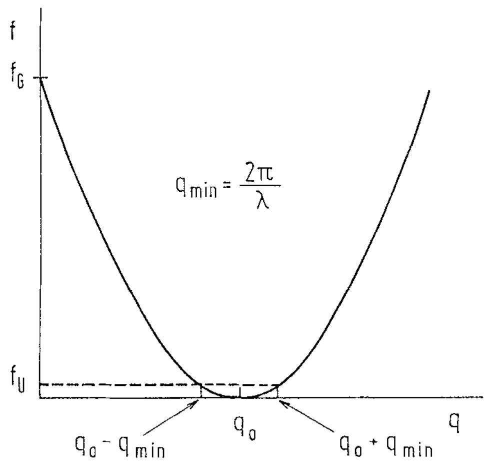


Figure 1. Estimation of the relaxation rate of the unwinding mode at zero bias field. Relaxation frequency ($f$) of phase fluctuations as a function of the wave vector $q$. The wave vector $q$, which corresponds to the unwinding process, is $q=q_0\pm q_{\min}$, where $q_{\min}=\pi/l$ ($\lambda=2l$ is the wavelength of the director distortion). The ratio $f_U/f_G$ is equal to $(q_{\min}/q_0)^2$, leading to equation (2).

zero due to the presence of a continuous helical symmetry in the system, the unwinding mode frequency ($f_U$) and the Goldstone mode frequency ($f_G$) can be compared

$$\frac{f_U}{f_G}=\frac{1}{4}\frac{p_0^2}{l^2}\propto\frac{1}{l^2}. \tag{2}$$

The phason relaxation frequency is shown in figure 1 as a function of the wavevector $q$. The relaxation frequency of the unwinding mode is schematically presented as well. A typical value of the helical period $p_0$ is about 1 $\mu$m. If the size of the sample is $l=1$ mm and the Goldstone mode frequency $f_G\approx 10^3$ Hz, the relaxation frequency of the unwinding process is $f_U\approx 10^{-4}$ Hz, which is a very small value compared to the Goldstone mode frequency. Even at samples with length $l\approx 10\,\mu$m along the helical axis, we see that the relaxation frequency of the unwinding mode increases only to $f_U\sim 1$ Hz. Thus the relaxation frequency of the unwinding mode is expected to be below the lowest frequencies used in a dielectric experiment, and for this reason, the unwinding mode cannot be observed in most experiments. The observability of this mode is also expected to be difficult due to the presence of ionic impurities which give a dominant contribution to the low frequency dielectric response. These two contributions might be resolved, because of their different frequency dependences, in thin samples where the unwinding mode frequency is large enough.

### 2.2. *Dielectric strengths of the Goldstone and the unwinding mode*

In this sub-section we derive the Goldstone and the unwinding mode dielectric strengths, i.e. their static contributions to the dielectric susceptibility, as functions of a bias electric field. Let us introduce dimensionless quantities in the following way. The dimensionless polarization $\tilde{P}$ and the reduced field $\varepsilon$ are defined according to

$$\tilde{P}=\frac{P_{\text{ind}}}{P_0},\qquad \varepsilon=\frac{E}{E_c}, \tag{3}$$

so that the dimensionless induced polarization $\tilde{P}$ is expressed in units of the spontaneous polarization $P_0$ and the field $\varepsilon$ in units of the critical unwinding field $E_c$, given by [9]

$$E_c=\frac{\pi^2 K_3 \Theta_0^2 q_0^2}{16 P_0}, \tag{4}$$

where $\Theta_0$ is the magnitude of the tilt. The static dielectric susceptibility $\chi$ of the system is related to a dimensionless susceptibility $\tilde{\chi}$ by the relation

$$\chi=\frac{P_0}{E_c}\tilde{\chi}, \tag{5}$$

so that

$$\tilde{\chi}=\frac{d\tilde{P}}{d\varepsilon}=\frac{d\langle\cos\Phi\rangle}{d\varepsilon}. \tag{6}$$

In a previous paper [9] we have calculated the polar ordering $\langle\cos\Phi\rangle$ as a function of the reduced field $\varepsilon\in(0,1)$. The result of this calculation is shown in figure 2. The corresponding susceptibility $\chi$ is presented in figure 3 as a function of the bias field $\varepsilon$.

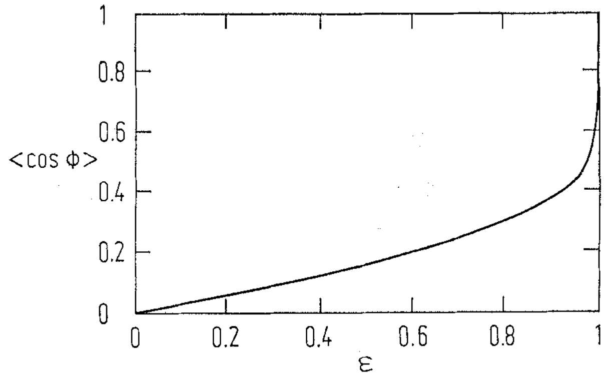


Figure 2. The polar ordering $\langle\cos\Phi\rangle$ as a function of the reduced bias electric field $\varepsilon$.

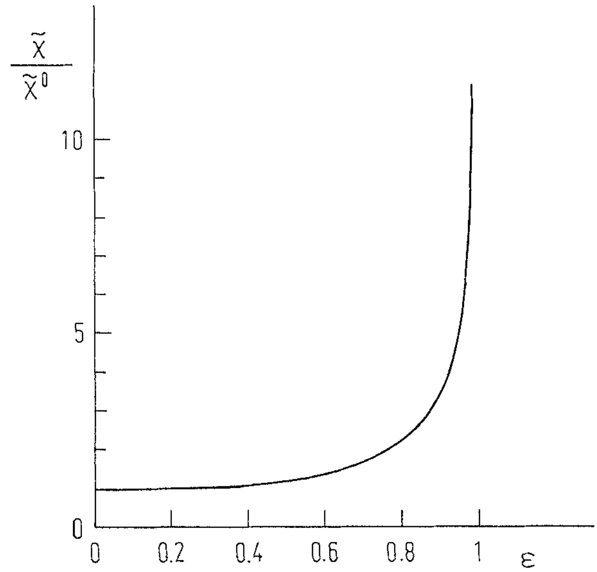


Figure 3. The sum of the Goldstone mode and the unwinding mode dielectric strengths, $\tilde{\chi}=d\langle\cos\Phi\rangle/d\varepsilon$ (normalized to the dielectric constant $\tilde{\chi}^0=\pi^2/32$) as a function of the bias electric field $\varepsilon$.

Since this is the total static response, the susceptibility includes both modes, the Goldstone mode and the unwinding mode

$$\tilde{\chi}=\tilde{\chi}_G+\tilde{\chi}_U. \tag{7}$$

The total susceptibility $\tilde{\chi}$ diverges at the critical field, where $\varepsilon\to 1$, as shown in figure 3. From the definition of the Goldstone mode contribution we deduce the corresponding susceptibility $\tilde{\chi}_G$

$$\tilde{\chi}_G=\left.\frac{d\langle\cos\Phi\rangle}{d\varepsilon}\right|_{p=\mathrm{const}}, \tag{8}$$

where $p$ denotes the pitch of the helix as a function of the bias field $\varepsilon$. This definition has to be understood in the following way. At a given bias field, $\varepsilon$, the field is increased by a small amount $d\varepsilon$. To obtain the Goldstone mode contribution $\tilde{\chi}_G$ of the susceptibility, the additional field $d\varepsilon$ is not allowed to change the pitch. In this way the unwinding mode contribution $\tilde{\chi}_U$ is defined as a difference between the total susceptibility $\tilde{\chi}$ and the Goldstone mode susceptibility $\chi_G$.

In order to derive dielectric strengths $\tilde{\chi}$, $\tilde{\chi}_G$ and $\tilde{\chi}_U$, we start with the balance of torque equation [10, 11]

$$K_3\Theta^2 \frac{\partial^2\Phi}{\partial z^2} - EP_0 \sin\Phi = 0. \tag{9}$$

Introducing the reduced electric field $\varepsilon$, defined by equations (3) and (4), and also introducing a dimensionless length scale $u = z/z^*$, where $z^* = \sqrt{(K_3\Theta^2/E_c P_0)}$, the equation we have to solve is a sine-Gordon equation

$$\frac{d^2\Phi}{du^2} - \varepsilon \sin\Phi = 0. \tag{10}$$

For a given value of the reduced field, $\varepsilon$, we can use this equation to calculate [9] the pitch $p$ over the pitch $p_0$ at zero field and the average induced polarization $\langle\cos\Phi\rangle$ in dimensionless units, respectively,

$$\frac{p}{p_0} = \left(\frac{2}{\pi}\right)^2 \frac{h}{\sqrt{\varepsilon}} \mathbf{K}(h), \tag{11}$$

$$\langle\cos\Phi\rangle = \frac{2}{h^2}\left[1 - \frac{\mathbf{E}(h)}{\mathbf{K}(h)}\right] - 1. \tag{12}$$

In these equations $\mathbf{K}(h)$ and $\mathbf{E}(h)$ are complete elliptic integrals [12] of the first and the second kind, respectively, and the parameter $h$ is related to the reduced electric field $\varepsilon$ through the transcendent equation

$$h = \sqrt{\varepsilon}\mathbf{E}(h). \tag{13}$$

The total static response given by equation (6) can be expressed as

$$\tilde{\chi} = \frac{d\langle\cos\Phi\rangle}{d\varepsilon} = \frac{dh}{d\varepsilon}\frac{d\langle\cos\Phi\rangle}{dh}. \tag{14}$$

Using the properties [12] of complete elliptic integrals we calculate the derivative

$$\frac{d\langle\cos\Phi\rangle}{dh} = \frac{2}{h^3}\left[\frac{1}{1-h^2}\frac{\mathbf{E}(h)^2}{\mathbf{K}(h)^2} - 1\right]. \tag{15}$$

Distinguishing between the total dielectric strength in equation (6) and the Goldstone mode dielectric strength in equation (8) which has to be calculated at a constant pitch, determined by a bias field, we derive the expressions for the quantities $dh/d\varepsilon$ and $dh/d\varepsilon|_{p=\mathrm{const}}$, respectively. The first quantity, $dh/d\varepsilon$, which allows the small measuring field to change the pitch is calculated as the total derivative of equation (13), whereas the second quantity, $dh/d\varepsilon|_{p=\mathrm{const}}$, which refers to the change in the parameter $h$ at constant pitch, is obtained from the total derivative of equation (11),

$$\frac{dh}{d\varepsilon}=\frac{1}{2h}\frac{\mathbf{E}(h)^3}{\mathbf{K}(h)}, \tag{16a}$$

$$\left.\frac{dh}{d\varepsilon}\right|_{p=\text{const}}=\frac{1-h^2}{2h}\mathbf{E}(h)\mathbf{K}(h). \tag{16b}$$

By substituting equations (15) and (16) into equation (14) we derive the final result

$$\tilde{\chi}=\frac{1}{h^4}\frac{\mathbf{E}(h)^3}{\mathbf{K}(h)}\left[\frac{1}{1-h^2}\frac{\mathbf{E}(h)^2}{\mathbf{K}(h)^2}-1\right], \tag{17a}$$

$$\tilde{\chi}_G=\frac{1-h^2}{h^4}\mathbf{E}(h)K(h)\left[\frac{1}{1-h^2}\frac{\mathbf{E}(h)^2}{\mathbf{K}(h)^2}-1\right]. \tag{17b}$$

The total dielectric strength $\tilde{\chi}$ depicted in figure 3 diverges at the critical field ($\varepsilon\to 1$, $h\to 1$), whereas the Goldstone mode dielectric strength $\tilde{\chi}_G$ decreases with a bias field and goes to zero at the critical field as shown in figure 4. The dielectric strength $\tilde{\chi}_U$ of the unwinding mode, presented in figure 5, is by definition (7) a difference between the total strength and Goldstone mode strength. It is zero in the absence of a bias field, increases with the field monotonously and diverges at the critical field.



## 3. Conclusion

In the present paper we show that the phason part of the dielectric response of the $S_C^*$ phase in a bias electric field consists of two modes. One of these, the unwinding mode, is related to the unwinding of the helix, characterized by the wave vector $q=q_0\pm\pi/l$, $l$ being the length of the sample along the helical axis. The corresponding relaxation frequency $f_U$ is inversely proportional to the square of the

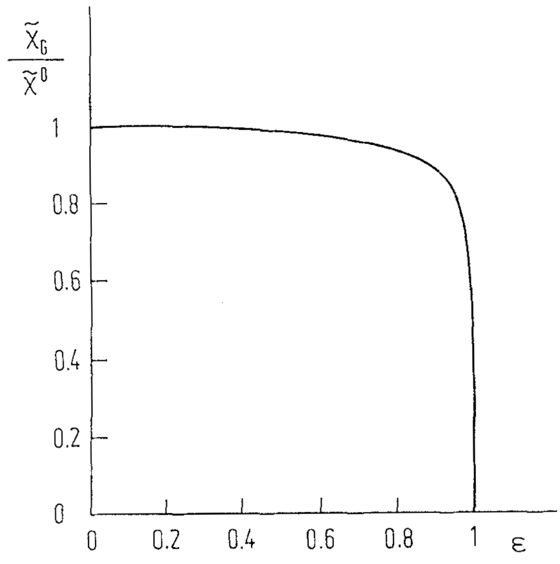


Figure 4. The Goldstone mode dielectric strength $\tilde{\chi}_G=d\langle\cos\Phi\rangle/d\varepsilon|_{p=\text{const}}$ as a function of the bias electric field $\varepsilon$, normalized to the dielectric constant $\tilde{\chi}^0$ at zero bias field, $\tilde{\chi}^0=\pi^2/32$.

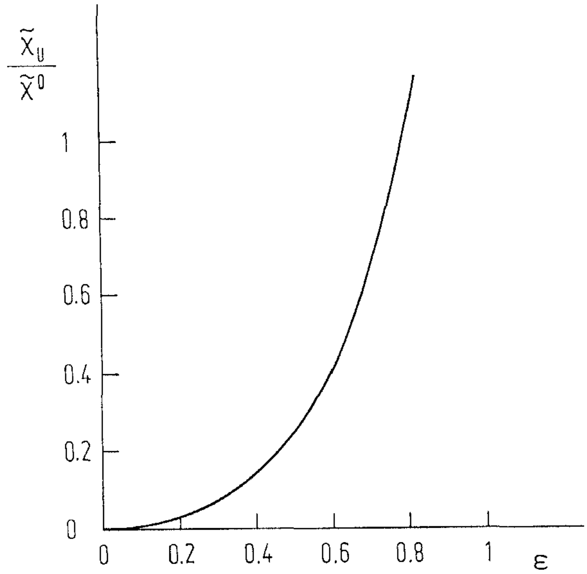


Figure 5. The unwinding mode dielectric strength $\tilde{\chi}_U = \tilde{\chi} - \tilde{\chi}_G$ as a function of the bias electric field $\varepsilon$, normalized to the dielectric constant $\tilde{\chi}^0$ at zero bias field, $\tilde{\chi}^0 = \pi^2/32$.



sample length along the helical axis. The unwinding mode frequency is estimated to be less than 1 Hz (c.f. equation (2)). The other mode, the Goldstone mode, is associated with the deformation of the polarization profile at constant pitch, i.e. the corresponding wave vector is equal to the wave vector of the helical pitch. The Goldstone mode frequency [1, 4–6, 13] is of the order of a few hundred Hz.

In dielectric experiments, the measuring field is used with a frequency which is higher than the unwinding mode frequency $f_U$. As a consequence, only the Goldstone mode is detected in these experiments. We show that the dielectric strength of the Goldstone mode decreases with a bias field and goes to zero at the critical field (see figure 4). This behaviour is in a qualitative agreement with what is observed in experiments [5–8].

The unwinding mode dielectric strength $\chi_U$ being zero in the absence of a bias field increases with the field and diverges at the critical field (see figure 5). However, as already discussed, the slow unwinding mode does not contribute to the measured dielectric susceptibility unless very low frequencies are used in the experiment. This mode could be observed, if the helix is confined to a small domain (for example, [14]) and the relaxation frequency is shifted into an experimentally accessible range.

It should also be pointed out that recent observations [15] of more than two modes (the soft) and the Goldstone mode) in ferroelectric liquid crystals can be explained on the basis of general symmetry arguments. External fields or restricted geometries break the continuous helicoidal symmetry of the bulk $S_C^*$ phase, so that additional modes are expected, which recover the broken symmetry.

Ozaki and Yoshino [16], who have studied the dynamic response of second harmonic generation in a ferroelectric liquid crystalline system to an applied, stepwise electric field, have detected two response times, one of the order of about a second and the other one of the order of about a millisecond. Recently also, a calculation by Hornreich and Shtrikman [17] shows that the dynamic response of the helicoidal cholesteric phase consists of two relaxations with well-separated relaxation times $t_{fast}/t_{slow} \approx 10^{-4}$. There exists therefore both experimental and theoretical evidence in the literature to support the model, presented in this paper.

This work was supported by the National Science Foundation through the United States-Yugoslav Joint Fund for Scientific Cooperation under Grant No. NSF JF 845. Support from the Ministry of Science and Technology of the Republic of Slovenia is also acknowledged.

## References

[1] LEVSTIK, A., CARLSSON, T., and ŽEKŠ, B., 1987, *Phys. Rev.* A, **35,** 3527.
[2] CARLSSON, T., ŽEKŠ, B., FILIPIČ, C., and LEVSTIK, A., 1990, *Phys. Rev.* A, **42,** 877.
[3] CARLSSON, T., and DAHL, I., 1983, *Molec. Crystals liq. Crystals,* **95,** 373.
[4] FILIPIČ, C., CARLSSON, T., LEVSTIK, A., ŽEKŠ, B., BLINC, R., GOUDA, F., LAGERWALL, S. T., and SKARP, K., 1988, *Phys. Rev.* A, **38,** 5833.
[5] BAHR, CH., HEPPKE, G., and SHARMA, N. K., 1987, *Ferroelectrics,* **76,** 151.
[6] PAVEL, J., GLOGAROVA, M., and BAWA, S. S., 1987, *Ferroelectrics,* **76,** 221.
[7] KUTNJAK, Z., and LEVSTIK, A. (personal communication).
[8] GOUDA, F. (personal communication).
[9] URBANC, B., ŽEKŠ, B., and CARLSSON, T., 1991, *Ferroelectrics,* **113,** 207.
[10] HANDSCHY, M., and CLARK, N. A., 1984, *Ferroelectrics,* **59,** 69.
[11] CARLSSON, T., and ŽEKŠ, B., 1989, *Liq. Crystals,* **5,** 359.
[12] JAHNKE, E., EMDE, F., and LÖSCH, F., 1960, *Tafeln Höherer Funktionen* (B. G. Teubner Verlagsgesellschaft).
[13] LEVSTIK, A., KUTNJAK, Z., FILIPIČ, C., LEVSTIK, I., BREGAR, Z., ŽEKŠ, B., and CARLSSON, T., 1990, *Phys. Rev.* A, **42,** 2204.
[14] BERESNEV, L. A., LOSEVA, M. V., CHERNOVA, N. I., KONONOV, S. G., ADOMENAS, P. V., and POZHIDAYEV, E. P., 1990, *Pis'ma Zh. éksp. teor. Fiz.,* **51,** 457.
[15] WROBEL, S., BIRADER, A. M., and HAASE, W., 1989, *Ferroelectrics,* **100,** 271. WROBEL, S., HAASE, W., PFEIFFER, M., BERESNEV, L., and GEELHAAR, T., 1992, *Molec. Crystals liq. Crystals,* **212,** 335.
[16] OZAKI, M., and YOSHINO, K., 1989, *Jap. J. appl. Phys.,* **28,** L1830.
[17] HORNREICH, R. M., and SHTRIKMAN, S., 1991, *Phys. Rev.* A, **44,** R3490.