Dielectric relaxation in a ferroelectric liquid crystal

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Résumé. — Nous présentons le calcul de la distorsion d'un cristal liquide ferroélectrique au voisinage de la transition A → C* sous l'effet d'un champ électrique alternatif parallèle aux couches smectiques. Cette distorsion se décompose en quatre modes normaux ; chacun de ces modes est une combinaison de la polarisation électrique et de la direction des molécules. Notre hypothèse originale par rapport au travail de Blinc et Zeks est que la viscosité ne couple pas les modes normaux. Dans ce cas, nous trouvons que le *mode mou* de la transition A → C* tend loin de celle-ci vers le mode diélectrique de ces deux phases. L'effet flexoélectrique modifie les fréquences de relaxation des modes dans la phase C* près de la transition. Cet effet change l'amplitude relative des contributions des quatre modes à la constante diélectrique ; la valeur de celle-ci à fréquence nulle n'en dépend cependant pas.

Abstract. — We present an analytic calculation of the normal modes of distortion induced by a small transverse A.C. electric field on a ferroelectric liquid crystal close to its transition temperature T_c between the smectic A and C* phases. These normal modes are superpositions of electric polarization and angular molecular distortion. Compared to a previous work of Blinc and Zeks, we assume that the losses are diagonal for the normal modes. The new prediction is that the soft mode in the A and C* phase goes continuously into pure polarization modes far from T_c. Flexoelectric effects eventually could be observed close to T_c, in the amplitude and frequencies of the A.C. contribution of the normal modes to the dielectric constant, although for zero frequency the sum of these contributions remains zero.

1. **Introduction.** — Ferroelectric liquid crystals possess in their ordered C* phase an increase of the transverse dielectric constant [1], which is attributed to the so-called « Goldstone » mode, i.e. to the rotation of the spontaneous polarization toward the applied electric field. On the basis of simple arguments [2, 3], the relaxation frequency for this process has been identified with the mechanical relaxation frequency of the helical texture. In fact, an electric field also changes the tilt of the molecules inside the smectic layers, so that four variables are involved in the problem, two for the molecular orientation and two for the transverse polarization [4].

A more complete calculation is needed. Such a calculation has been made by Blinc et al. [5, 6], who have given the amplitude and frequencies of the corresponding four normal modes of the problem. As previously shown [7], this treatment was incomplete because it gave to the flexoelectric component of the spontaneous polarization a role in the dielectric response, although it has none at zero frequency.

In addition, dissipation was introduced for the tilt and polarization variables, and not for the normal modes.

In this work, we describe the dynamics of E field induced distortions in a ferroelectric smectic liquid crystal, with a dissipation function diagonal for the eigenmodes of the distortion. This assumption allows us to relate the properties of the « soft » mode close to the transition temperature (in particular the electroclinic [8] effect), with the standard relaxation of the transverse dielectric constant far from T_c.

The geometry of our system and the complex notation are the same as described in reference [7]. The helical axis of the C* texture is the Z axis. The field **E** is applied along OX. As usual we call K_3 the layer twist elastic constant, q the wave vector of the spontaneous helix, C the electroclinic coefficient, $a[a = \alpha(T - T_c)]$ and b the first coefficients of the Landau expansion of the free energy density.

In [7], we had written an expression for the dissipation function as :

$$\frac{1}{2\Gamma_1} \dot{\theta}\dot{\theta}^* + \frac{1}{2\Gamma_2} \dot{P}\dot{P}^* + \frac{j}{2\Gamma_3} [\dot{P}\dot{\theta}^* - \dot{P}^* \dot{\theta}] .$$

In principle, $\Gamma_1 \Gamma_2 \Gamma_3$ could be chosen arbitrarily, so that the dissipation function could be non diagonal if represented on the basis of the eigenvector of the free energy. This would represent a situation where the losses would couple the modes of distortion. This coupling is probably very weak due to the very large number of thermal modes to which each distortion mode is coupled, as implicitly written in the dissipation function. It seems reasonable to describe the dynamics of the system by neglecting this possible friction coupling between the E field distortion modes. This amounts to writing a dissipation function diagonal on the eigenvectors defining the modes. As in references [5] and [6], we also neglect inertia effects, i.e. we restrict our analysis to the purely damped regime, typically below the lowest relaxation frequency of the transverse dielectric constant $\varepsilon_\perp$. This frequency is in the MHz range.

We start from the free energy expression [4, 5]

$$f = \frac{1}{2} a\theta\theta^* + \frac{1}{4} b(\theta\theta^*)^2 + j\frac{A}{2}\left(\theta\frac{\partial\theta^*}{\partial z} - \theta^*\frac{\partial\theta}{\partial z}\right) +$$

$$+ K_3 \frac{\partial\theta}{\partial z}\frac{\partial\theta^*}{\partial z} + \frac{1}{2\chi}PP^* - \frac{\mu}{2}\left(P\frac{\partial\theta^*}{\partial z} + P^*\frac{\partial\theta}{\partial z}\right)$$

$$+ j\frac{C}{2}(P\theta^* - P^*\theta) - \frac{1}{2}(P + P^*)E . \qquad (1)$$

The equilibrium (Euler) equations are :

$$\frac{P}{\chi} - \mu\frac{\partial\theta}{\partial z} - jC\theta = E \qquad (2)$$

$$\mu\frac{\partial P}{\partial z} + jCP - K_3\frac{\partial^2\theta}{\partial z^2} - 2jA\frac{\partial\theta}{\partial z} + (a + b\theta\theta^*)\theta = 0 . \qquad (3)$$

2. Electric induced distortion in the smectic A phase.

— Let us first discuss the case of the smectic A phase, where $\theta\theta^*$ is zero. After a Fourier transform we find the normal modes by diagonalizing. We note with a subscript k the amplitude of any component of wave vector k varying like $\exp(jkz)$.

Equations (2) and (3) are written simply as :

$$P_k - j\left(1 + \frac{\mu k}{C}\right)\chi C\theta_k = \chi E_k \qquad (2')$$

$$j\left(1 + \frac{\mu k}{C}\right)P_k + \left[A_k + \left(1 + \frac{\mu k}{C}\right)^2\right]\chi C\theta_k = 0 \qquad (3')$$

$$A_k = \frac{\alpha(T - T_c) + K(k - q)^2}{\chi C^2} \text{ is the dimensionless}$$

temperature; $K = K_{33} - \chi\mu^2$ is the renormalized [4] twist constant. As the field E is uniform, we are only interested in the $k = 0$ components. We must now diagonalize (2'), (3'). The eigenvalue equation is :

$$r^2 - r\left[1 + A_k + \left(1 + \frac{\mu k}{C}\right)^2\right] + A_k = 0 ,$$

where the flexoelectric term $\mu k/C$ is retained just to point out that $k \neq 0$ modes have different eigenvalues (see later the discussion on flexoelectricity in the C* phase; the new Fourier component which appears in addition to $k = 0$ is $k = 2q$; this results in the same A). We now drop the flexo term for $k = 0$. The eigenfunctions are :

$$\varphi_{1,0} = P_0 - j\chi C\theta_0(r_1 - 1)$$

$$\varphi_{2,0} = P_0 - j\chi C\theta_0(r_2 - 1) .$$

From now on, we suppress the obvious subscripts.
The diagonalized equations are now

$$r_1 \varphi_1 = \chi E_0 = \chi E \qquad (2'')$$

$$r_2 \varphi_2 = \chi E . \qquad (3'')$$

The eigenvalues are :

$$r_1 = \tfrac{1}{2}[2 + A - (4 + A^2)^{1/2}]$$

$$r_2 = \tfrac{1}{2}[2 + A + (4 + A^2)^{1/2}]$$

r_1 corresponds to the soft mode (« in phase » mode of reference [5]);
r_2 corresponds to the hard mode (« out of phase »). With the previously discussed assumption, the dynamical equations associated with (2'') and (3'') are :

$$r_1 \varphi_1 + \tau_1 \dot{\varphi}_1 = \chi E$$

$$r_2 \varphi_2 + \tau_2 \dot{\varphi}_2 = \chi E$$

τ_1 and τ_2 are the only two friction coefficients (with dimensions of time) which remain in the chosen approximation. τ_1 and τ_2 are presumably of the same order of magnitude, comparable to a transverse dielectric relaxation time. Assuming $E = E \exp(j\omega t)$, we find :

$$\varphi_1(\omega) = \frac{\chi E}{r_1 + j\omega\tau_1} \qquad \varphi_2(\omega) = \frac{\chi E}{r_2 + j\omega\tau_2} .$$

To calculate $\varepsilon_\perp$, we derive $P(E)$ of the form :

$$P_0(\omega) = \frac{\chi}{r_1 - r_2}\left[\frac{1 - r_2}{r_1 + j\omega\tau_1} - \frac{1 - r_1}{r_2 + j\omega\tau_2}\right]E .$$

The two terms in the bracket represent the contribution to the dielectric constant from the two normal modes. For $\omega = 0$, these two terms recombine to give back the result of our previous calculations [7]

$$\Delta\varepsilon_\perp = \frac{4\pi\chi}{A} = \frac{4\chi C^2 \chi^2}{Kq^2 + \alpha(T - T_c)} .$$

On figure 1, we have plotted the dependence of r_1 and r_2 versus A (i.e. versus the temperature). We have also plotted (Fig. 2) the corresponding amplitudes of the two relative contributions to $P_0(\omega)$, i.e.

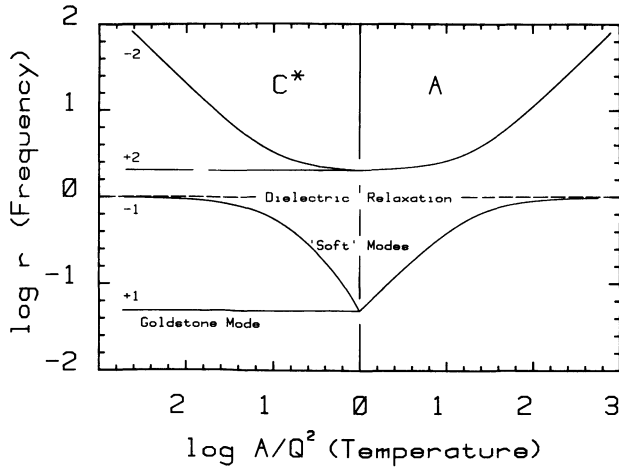$$\frac{1}{r_1[1 + (r_1 - 1)^2]} \text{ and } \frac{1}{r_2[1 + (r_2 - 1)^2]} .$$

Fig. 1. — Relaxation frequencies of the four modes (smectic C* without flexo coupling) and of the two modes (smectic A) of distortion induced by a transverse electric field, versus the reduced temperature. T_c corresponds to zero, smectic A on the right.

On the « soft mode » branch, r_1 goes from almost zero close to T_c ($A \to 0$), to 1 at high temperature. In our notations, T_c corresponds to $A = Kq^2/\chi C_i^2 = Q^2$. For $A \to 0$, the corresponding amplitude diverges like $1/2\ A$. For high temperature, the amplitude saturates down to 1. The relaxation frequency of the soft mode contribution is r_1/τ_1. Close to T_c, there should be a slowing down, limited at the value

$$A/2\ \tau_1 \sim (Kq^2/C^2)\frac{1}{2\ \tau_1} .$$

At high temperature, the relaxation frequency saturates at $1/\tau_1$.

On the « hard mode » branch r_2 goes from 2 (close to $A = 0$) to infinity at high temperature, where it diverges as A/τ_2. The corresponding amplitude goes from 1/4 to 0 (as A^{-3}) as shown on figure 2. The relative amplitude of the hard mode contribution remains weak compared to that of the soft mode. The relaxation frequency of the hard mode
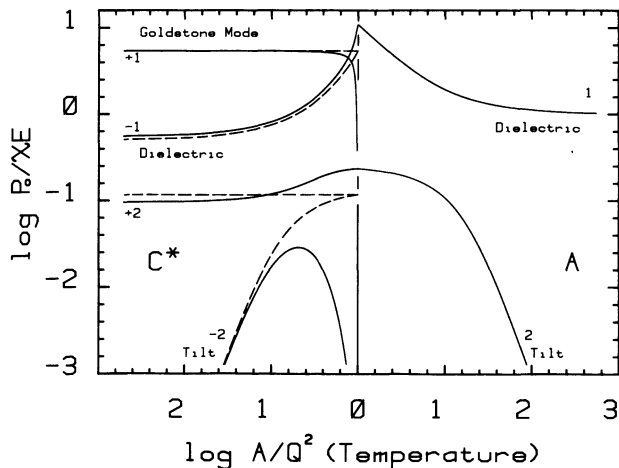

Fig. 2. — Amplitude of the relative uniform polarization versus the reduced temperature. The dashed lines represent the case $\beta = 0$ (no flexo coupling) the full lines correspond to $\beta = 0.05$.

is r_2/τ_2. Since $r_2 > r_1$ and $\tau_1 \sim \tau_2$, the hard mode relaxation frequency is expected to be larger than the soft mode relaxation frequency r_1/τ_1.

Knowing r_1 and r_2, we can discuss the physical nature of the two modes. We can see on figures 2 and 3 the amplitudes of $P/\chi E$ and $C\theta/E$ for the normal modes. Close to T_c ($A \sim 0$), φ_1 and φ_2 are modes of coupled tilt and polarization. For large A, at high temperature, φ_1 tends to a pure polarization mode, and φ_2 tends to a pure tilt mode. θ and P are decoupled. $1/\tau_1$ appears as the transverse dielectric relaxation frequency of a standard smectic A (in the MHz range). $r_2/\tau_2 \sim A/\tau_2$ appears as the pure tilt relaxation, observed for instance in light scattering experiments [9] above a $S_A \to S_C$ transition. Note, however, that this high temperature limit is obviously not valid close to T_c, where θ and P are strongly coupled. In that range of temperature ($A \sim 0$), the low frequency relaxation of the mode 1 must be identified with the relaxation observed in the electroclinic effect.
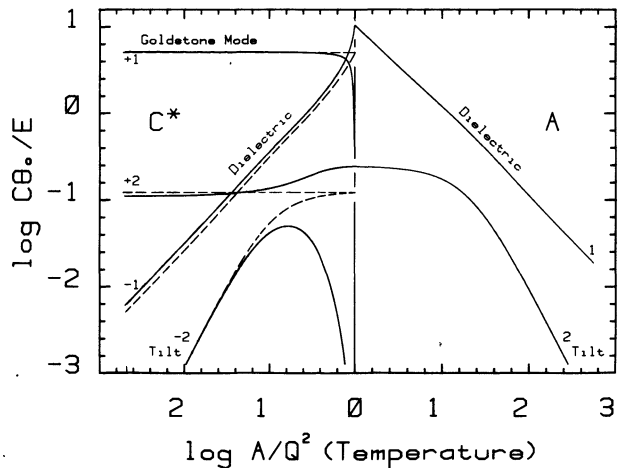

Fig. 3. — Amplitude of the relative uniform tilt versus the reduced temperature. The dashed lines represent the case $\beta = 0$ (no flexo coupling). The full lines correspond to $\beta = 0.05$. Dielectric means that, far from T_c, the mode tends toward a pure dielectric mode.

3. Electric induced distortions in the smectic C* phase. — We now discuss the more complicated case of the smectic C*. In the absence of an E field, the minimization of f gives the spontaneous polarization and tilt :

$$P_q = j\left(1 + \frac{\mu q}{C}\right) \chi C\theta_q$$

$$\theta_q\,\theta_q^* = A_q\frac{\chi C^2}{b} = -\frac{\alpha(T - T_c)}{b} .$$

To calculate the normal modes of distortion induced by the field E, one must go back now to the small amplitude approximation and write the various equations which give the amplitudes P_k and θ_k excited by the field E_k. As previously seen in the case of smectic A, with a uniform E, we excite P_0 and θ_0. The non linear term $b\theta\theta^*\ \theta$, when expressed with the θ_k, gives

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a coupling between the various k components. We keep only in this term those which are linear in the small distortion θ_k. This results, in the equation for the $k = 0$ Fourier component, in a change in the coefficient of θ_0 and in a coupling of θ_0 with θ_{2q}^*, which

is thus also indirectly excited. The eigenmodes must be linear combinations of the θ_0, P_0 and θ_{2q}^* and P_{2q}^*, as first noted by Zeks and Blinc [5]. To explicitly do the calculation, we write the equations (2) and (3) to be diagonalized, in the matrix form :

$$\begin{vmatrix} 1 & -j & 0 & 0 \\ j & 1+A & 0 & (A-Q^2)\exp(2jqz) \\ 0 & 0 & 1 & j(1+2\beta) \\ 0 & (A-Q^2)\exp(-2jqz) & -j(1+2\beta) & 1+A+4\beta(1+\beta) \end{vmatrix} \begin{vmatrix} P_0 \\ \chi C\theta_0 \\ P_{2q}^* \\ \chi C\theta_{2q}^* \end{vmatrix} = \begin{vmatrix} \chi E \\ 0 \\ 0 \\ 0 \end{vmatrix}.$$

In this equation, A is now

$$\frac{Kq^2 + b\theta_q\,\theta_q^*}{\chi C^2} = \frac{Kq^2 + \alpha(T_c - T)}{\chi C^2}$$

and again measures the temperature scale. β is now $\mu q/C$. The eigenvalue equation for r is now the quadratic equation :

$$(r^2 - r(2+A) + A)\,(r^2 - r(2+A+4\,\beta(1+\beta)) + A) - $$
$$- (1-r)^2(A-Q^2)^2 = 0\,.$$

The eigenmodes are the vectors :

$$P_0 = 1$$
$$\chi C\theta_0 = j(r-1)$$
$$P_{2q}^* = \frac{(1+2\,\beta)}{(A-Q^2)\,(r-1)}\,(1+(r-1)\,(1+A-r)) \times$$
$$\times \exp(-2jqz) \quad (4)$$
$$\chi C\theta_{2q}^* = \frac{-j}{A-Q^2}\,(1+(r-1)\,(1+A-r))\,\exp(-2jqz)\,.$$

Note that, in general, whatever may be the eigenvalues r, because they are real, the various complex k components of P and θ remain « perpendicular », as were the spontaneous P_q and θ_q. The only difference between the different modes comes from the possible varying sign of $(r - 1)$ which may lead to various « in phase » and « out of phase » geometries. For the two modes $\pm$ 1, $r - 1$ remains negative. The P and θ distortions remain « in phase » i.e. with the same relative orientation as the spontaneous P_q and θ_q. The modes $\pm$ 2 are the « out of phase » combinations.

To simplify the discussion, let us take first the simple case $\beta = 0$ (no flexoelectricity).
In this case, the eigenvalue equation decomposes into two equations :

$$r^2 - r(2 + A) + A \pm (1 - r)\,(A - Q) = 0\,.$$

The two values r_+ (associated with the $+$ sign) are temperature independent :

$$r_{+1} = 1 + \frac{Q^2 - (4 + Q^4)^{1/2}}{2}$$

$$r_{+2} = 1 + \frac{Q^2 + (4 + Q^4)^{1/2}}{2}\,.$$

For low Q^2 (typically, we expect $Q^2 \sim 10^{-1}$-10^{-3}) r_{+1} is very close to $Q^2/2 = Kq^2/2\,\chi C^2$ and r_{+2} is very close to 2. Again calling $\tau_{\pm,1/2}$ the damping time associated with each normal mode, the relaxation frequencies of these $+$ modes are of the order of $Q^2/2\,\tau_{+1}$ and $2/\tau_{+2}$. The two other eigenvalues r_- are temperature dependent :

$$r_{-1} = 1 + \frac{2\,A - Q^2 - ((2\,A - Q^2)^2 + 4)^{1/2}}{2}$$

$$r_{-2} = 1 + \frac{2\,A - Q^2 + ((2\,A - Q^2)^2 + 4)^{1/2}}{2}\,.$$

At T_c, the two eigenfrequencies r_{-1}/τ_{-1} and r_{-2}/τ_{-2} are equal to the corresponding frequencies of the $+$ modes, and of course to the two eigenfrequencies of the 1 and 2 modes of the smectic A phase. The reason is that, at T_c, the modes 1 and 2 decompose into 1 $\pm$ and 2 $\pm$. This implies that $\tau_{\pm 1} = \tau_1$ and $\tau_{\pm 2} = \tau_2$. We shall assume that this remains valid even in presence of flexoelectric effects, in the $\beta \neq 0$ case. Far from T_c, the relaxation frequency r_{-1}/τ_1 tends to the constant $1/\tau_1$, the dielectric relaxation frequency of the smectic A (since we have neglected the dielectric anisotropy to describe the properties of the C* phase). The other frequency r_{-2}/τ_2 diverges as $2\,A/\tau_2$, i.e. linearly in $T_c - T$.

To understand the physical nature of the modes, let us look at the P and θ components of the $\pm$ 1,2 eigenvectors. Recombining the $k = 0$ and $k = 2\,q$ components, we find for the $+$ 1 mode :

$$P_{+1} \simeq \frac{\chi E}{Q^2}\cos qz\,\exp(jqz)\,,$$

and

$$\theta_{+1} \simeq \frac{-jE}{CQ^2}\cos qz\,\exp(jqz)\,.$$

Remember that the undisturbed spontaneous helix is defined by $\theta_q = |\,\theta_q\,|\exp(jqz)$. The $+$ 1 distortion is then a pure rotation of the spontaneous θ_q and P_q.

With the small Q^2 in denominator, the amplitude of this « Goldstone » mode is by far the largest of the four modes. The relative amplitude of the distortion (i.e. the quantities $P_0/\chi E$, $C\theta_0/E$, $P_{2q}/\chi E$, $C\theta_{2q}/E$, which are the relative susceptibilities of the polarizability and of the electroclinic effects) are plotted on figures 2, 3, 4, 5. The $- 1$ mode corresponds to an « in phase » change in the moduli of the spontaneous P_q and θ_q. The amplitudes are :

$$P_{-1} \sim -jE \frac{\exp(jqz)\sin qz}{r(1+(1-r)^2)} \qquad \theta_{-1} \sim \frac{E\exp(jqz)\sin qz}{Cr(1+(1-r)^2)}.$$

Close to T_c, the relative susceptibilities have the same amplitude, although far below T_c, as shown on figures 2 and 3 the $- 1$ mode becomes a pure polarization mode, the θ distortion beeing quenched by the large rigidity of the smectic layers. This mode is the « soft » mode of the C* phase.
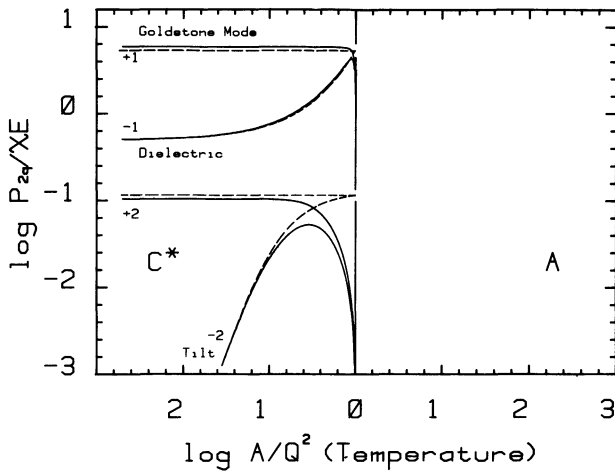

Fig. 4. — Amplitude of the $k = 2\,q$ relative polarization, *versus* the reduced temperature. Other details as in figure 3.
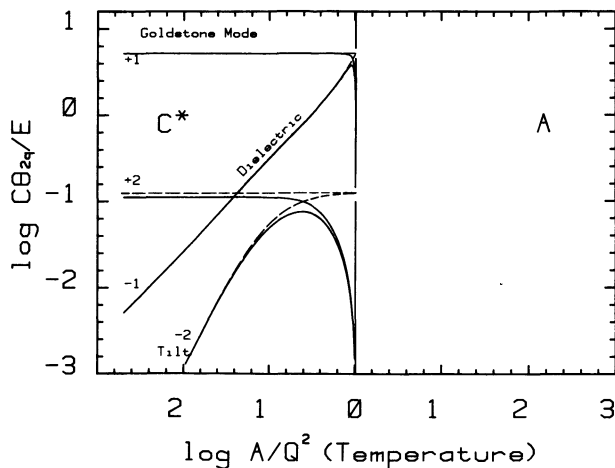

Fig. 5. — Amplitude of the $k = 2\,q$ relative tilt, *versus* the reduced temperature. Other details as in figure 3.

The amplitudes of the mode ± 2 are plotted on figures 2, 3, 4, 5. The $+ 2$ mode corresponds to equal

rotations, in opposite senses, of P_q and θ_q. The amplitudes are

$$P_{+2} \sim \frac{\chi E}{4}\exp(jqz)\cos qz$$

and

$$\theta_{+2} \sim \frac{jE}{4C}\exp(jqz)\cos qz.$$

They are $\sim Q^2/4$ times smaller than the one of the dominant $+ 1$ « Goldstone » mode, and temperature independent. The $- 2$ mode corresponds to « out of phase » change in the moduli of P_q and θ_q. The amplitudes are :

$$P_{-2} \sim \frac{-j\chi E\exp(jqz)\sin qz}{r(1+(1-r)^2)(r-1)}$$

$$\theta_{-2} \sim \frac{E\exp(jqz)\sin qz}{Cr(1+(1-r)^2)}.$$

For high temperature, as shown on figures 2, 3, this mode becomes a pure tilt mode.

Let us now discuss the case where the flexoelectric coupling is included. The eigenvectors now take the complicated form given by equation (4). We have seen in the C* phase that, to first order in field E, the distortion of the texture can be described as a superposition of $k = 0$ and $k = 2\,q$ components of P and θ, i.e. are represented by four normal modes ; linear combination of these four variables. On the other hand, in the A phase, only two modes are excited, a combination of P_0 and θ_0 excited by E. P_{2q} and θ_{2q} have zero amplitude. Just at the transition, the 4 modes of the C* must go continuously into the corresponding modes of the A phase, at spatial frequencies 0 and $2\,q$. In the previous special case of no flexoelectric coupling, the smectic A modes at $k = 0$ and $k = 2\,q$ had same eigenvalues (and frequencies). This implied that the four C* modes had to merge two by two at the transition, so that the relaxation frequencies were continuous with those of the A phase. Turning on the flexo-
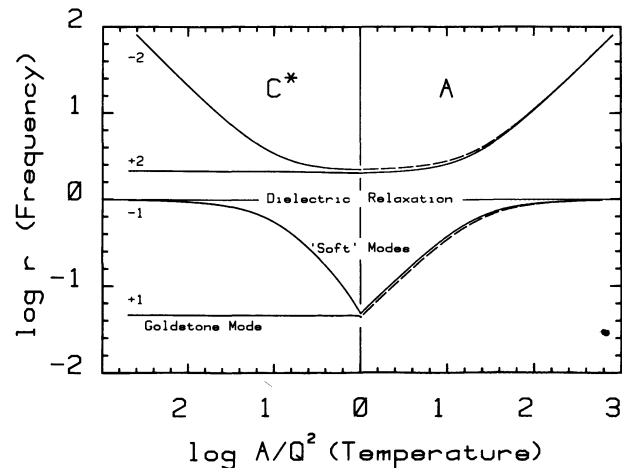

Fig. 6. — Relaxation frequencies of the E induced distortion modes, in the presence of flexoelectric coupling $\beta = 0.05$, *versus* reduced temperature. The dashed lines represent the $k = 2\,q$ mode relaxation frequencies which have zero amplitude in the smectic A phase.

electric coupling results in different eigenvalues (and frequencies) for the corresponding 0 and 2 q modes of the A phase. The four modes of the C* phase at T_c thus have, in general, different eigenvalues and frequencies, as shown on figure 6. Since we keep the requirement that the amplitude of the 2 q modes in the A phase must be zero, on going to T_c from the C* phase, the amplitude of all the four 2 q components, and of the two $k = 0$ components for the two modes which go continuously toward the 2 q mode of the A phase must vanish. We have computed the relative amplitudes of $P/\chi E$ and $C\theta/E$ at $k = 0$ and $k = 2 q$ for the four different modes of the C* phase (see Figs. 2, 3, 4, 5) versus the reduced temperature A. These curves are computed for a typical value of the twist energy $Q^2 = 0.1$. Note that the flexo coupling $\beta = \mu q/C$ cannot be too large. Its maximum value is that which suppresses the curvature elasticity, because of the K renormalization $K = K_3 - \chi\mu^2$, i.e. $\beta_{Max} = Q \sim 0.3$.

We have chosen $\beta = 0.05$. One sees on figures 2, 3, 4, 5, that the amplitude of each mode depends on the flexo coupling on a temperature scale of the order of Q^2. For low temperature, the only excited mode remains the $+ 1$ « Goldstone » mode. The one component which is sensitive to the flexo coupling far below T_c is the $(P_{+1})_{2q}$ component, which varies as $(1 + \beta)$. In fact, a dielectric measurement would measure only the amplitudes and relaxation frequencies of the P_0 components of these modes. P_{2q} is not easy to observe. The simplest observable consequences of the flexo coupling are that : the relaxation frequencies at T_c are no longer doubly degenerate; and that, far below T_c, the relaxation frequency of the dominant « Goldstone » mode depends now on the flexoelectric coupling, from both the β dependence of r and the β renormalization of K_3.

To summarize, we can write formally the general expression giving $P_0(E, \omega)$ from the contribution of the four modes, as :

$$P_0(E, \omega) = \sum_{i=1}^{4} \frac{\chi E}{r_i + j\omega\tau_i} \times \left[1 + (1 - r_i)^2 + \left[\frac{(1 - r_i)^2 - A(1 - r_i)^{-1}}{Q^2 - A} \right]^2 \left(\left(\frac{1 + 2\beta}{1 - r_i} \right)^2 + 1 \right) \right]^{-1}$$

where $i = 1$ to 4 corresponds to the four ± 1, ± 2 modes. This bulky expression in fact gives $\chi(\omega)$, from which we can simply derive $\Delta\varepsilon_\perp(\omega)$ (from the A phase) by $\Delta\varepsilon_\perp(\omega) = 4 \pi\chi(\omega)$. Using the eigenvalue equation, one can verify that, for $\omega = 0$, β vanishes from $\chi(0)$, as previously shown [7].

At the end of this calculation, we must examine the starting hypothesis of the pure damped regime to describe the dynamics. If, as we assumed, the two damping frequencies $1/\tau_1$ and $1/\tau_2$ are comparable, the maximum acceptable frequency is of the order of $1/\tau$. We must keep only the ± 1 modes to describe the relaxation. If $1/\tau_1$ and $1/\tau_2$ can be very different, the four mode analysis may retain some physical interest. In that case, however, the very small relative amplitudes of the high frequency ± 2 modes limit any practical interest of the calculation to the temperature range $A \sim Q^2$ close to T_c.

It is interesting to compare our predictions with existing data on the C* transverse dielectric constant. The most important parameter is Q^2, the relative twist to piezo energies. Dielectric measurements [10] indicate an increase in dielectric constant, from the « Goldstone » mode, of the maximum value of

$$\frac{2 \pi\chi}{Q^2} \Big/ (1 + 4 \pi\chi) \sim 11 .$$

In the A phase, $\varepsilon_\perp = 1 + 4 \pi\chi$ is of the order of 5, resulting in $Q^2 \sim 0.2$. A direct estimate of Q^2 can be made from the polarization measurement. Assuming a weak flexo coupling we take from $P = \chi C\theta$ (see Refs. [10] and [11]) the estimate

$$P/\theta = \chi C \sim 4 \times 10^{-2} \text{ Debye/rad.}$$

This results in $Q^2 \sim 10^{-2}$, with $\chi \sim 0.3$ and $K \sim 5 \times 10^{-7}$ cgs. In the A phase, we know from reference [8] that the lower relaxation frequencies of the electroclinic effect are in the kHz range. Since the transverse dielectric relaxation frequency in the A phase is in the MHz range, from our calculation this results in $Q^2 \sim 10^{-3}$. These independent measurements give two to three orders of magnitude dispersion for Q^2. There is a clear inconsistency between dielectric and spontaneous polarization measurements. Note that a recent report [12] from a Japanese group seems to indicate a larger value of $\Delta\varepsilon_\perp$, reaching 80. This corresponds to $Q^2 = 5 \times 10^{-2}$, in better agreement with the polarization measurement. Additional experiments would be useful to clarify this point.

4. Conclusion. — To conclude, we have studied the supposedly purely damped dynamics of the dielectric response of a C* ferroelectric liquid crystal. Instead of introducing friction on the physical variables describing the tilt (θ) and the polarization (P) as previously [5] made by Zeks and Blinc, we have introduced a damping diagonal for the normal modes (i.e. linear combinations of θ and P). We have given an analytic expression for the dielectric constant. In the A

phase, the relaxation of the electroclinic effect goes continuously to the simple transverse dielectric relaxation, far above T_c. In the C* phase, one again finds the dominant « Goldstone » mode, almost temperature independent. The other « soft » mode, as in the A phase, rapidly becomes very « rigid » below T_c and also merges into the simple dielectric relaxation

mode. The influence of the flexoelectric coupling should be visible close to T_c, where careful dielectric relaxation measurements could eventually detect it. At zero frequency, apart from the renormalization of K_3 and q, the flexoelectric coupling vanishes exactly from the dielectric constant, in both the A and C* phases.

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