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## Molecular Crystals and Liquid Crystals Incorporating Nonlinear Optics

Publication details, including instructions for authors and subscription information: <a href="http://www.tandfonline.com/loi/gmcl17">http://www.tandfonline.com/loi/gmcl17</a>

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Version of record first published: 20 Apr 2011.

To cite this article: Kamensky Viacheslav (1990): Dynamics of Frederiks Transition, Molecular Crystals and Liquid Crystals Incorporating Nonlinear Optics, 179:1, 277-284

To link to this article: <a href="http://dx.doi.org/10.1080/00268949008055376">http://dx.doi.org/10.1080/00268949008055376</a>

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Mol. Cryst. Liq. Cryst., 1990, Vol. 179, pp. 277-284 Reprints available directly from the publisher Photocopying permitted by license only © 1990 Gordon and Breach Science Publishers S.A. Printed in the United States of America

## DYNAMICS OF FREDERIKS TRANSITION

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Abstract A model describing the dynamics of the Frederiks transition in an inhomogeneous case is proposed. Formation and motion of structural walls is investigated.

The Frederiks transition is one of the well-known and well-investigated phenomena in liquid crystals. This is accounted for, on the one hand, by a relatively simple experimental technique of the study, and, on the other hand, by practical significance of the effect. It is natural that the study of the static case is most complete. The study of the dynamics of the effect is less complete and the existing papers (see [1] and references therein) describe processes of formation of the orientation structure only along the direction, orthogonal to the plates, restricting a nematic liquid crystal, i.e., in one-dimensional case. Then the mean-quadratic fluctuation of the angle of deviation of the director from the equilibrium in the moment when the field is switched on, is chosen as the initial condition, homogeneous in the plane, parallel to the plates.

However it is quite natural that for real specimen due to the presence of defects, boundaries, etc., this requirement of homogeneity is not fulfilled. Accordingly, characteristic times of formation of the structure will be determined by other mechanisms whose study require a more detailed description.

Take a case of the original homeotropic orientation of the director. Chose the z-axis in the direction of the original orientation (and, consequently, orthogonal to the plates, restricting the crystal), the x-axis - in the direction of the magnetic field, imposed parallelly to the plates and assume inhomogeneity along the y-axis. The equations of motion of an incompressible nematic become two-dimensional. The equations of motion for the velocity of a nematic  ${\bf v}$ , pressure  ${\bf v}$  and angle of the deviation of the director from the initial position are obtained in a standard fashion [1] and are not reproduced here because of their bulky form. Note only that it is assumed in these equations that  ${\bf v}$  1 and the main nonlinear terms are retained.

The next step is the study of the instability of the obtained system of the equations of motion. Linearization of these equations (in the initial moment  $\Psi=0$ ) permits to exclude  $\Psi$  and P exactly. Thus, it is necessary to investigate the stability of only the equation for  $\Psi$ . The dispersion relation for perturbations  $\delta \Psi \sim \exp\left(ikx + iqz + \omega t\right)$  in this case is

$$\omega(k,q) = (\gamma_a H^2 - K_1 k^2 - K_3 q^2) / \gamma^*$$
 (1)

where  $K_{i}$  are the Frank constants,  $\chi^* = \left[ \chi_1(d_4 + d_5) - d_2 \chi_2 \right] \times \left( d_4 + d_5 - d_2 \right)$  is the effective viscosity,  $d_i$  are Leslie coefficients, H is the magnetic field and  $\chi_a$  is the anisotropic part of the magnetic susceptibility.

If to assume that thereoccurs a strong coupling of nematic molecules with the restricting plates, the wave vector q may take only discrete values  $q = \pi m/d = mq_o$  (here d is the size of the sample in the direction Z,

 $m=1,2,\ldots$ ). Then as is clear from (1), for each harmonics m there is a certain critical field  $H_{mc}$  at which  $\omega(0,mq_o)$  becomes zero and the system loses its stability relative to the m-th harmonics. Simple estimates show that in real experiments there is always a sufficiently broad region of fields  $2H_c > H > H_c / H_c = (K_3/\chi_a)^{1/2} \mathfrak{N}/d/$ , where the first harmonics with  $q=q_o$  becomes unstable. This is the case we shall consider henceforth. Then the wave vectors k, corresponding to the unstable state, must obey the inequality  $k^2 \le \delta_o \, q_o^2 \, K_3 / K_4$ , where  $\delta_o = (H^2 - H_c^2)/H_c^2$  is the instability parameter. It is obvious that perturbations with k=0 increase with the largest increment.

Thus, in the case of small  $\delta_{\mathbf{o}} < 1$  , in the linear approximation in the system an instability for modes with the respective small wave vectors  $\mathbf{k}$  and with a small increment  $\omega$ starts developing. Increasing perturbations cease to satisfy the linear theory and to describe them it is necessary to employ the total system of nonlinear equations of motion. An exact solution of this system is not possible. Yet, as the analysis of the system and the successive comparison of the results with the assumptions show, in the case when  $\delta_o$  <1, the total system of equations of motion can also be reduced to an equation for  $\,$  . This equation in dimensionless variables  $z=Vq_0x$ ,  $\eta=q_0z$ ,  $\tau=K_3q_0^2t/y^*$ ,  $\nu=(K_3/K_4)^{1/2}is$  $\Psi_{\tau} = \Psi_{\xi\xi} + \Psi_{\eta\eta} + (1 + \delta_0) \Psi - \frac{2}{3} \Psi^3 - \frac{$  $-\frac{K_{1}-K_{3}}{K_{2}}\left[\varphi(\varphi_{\eta}^{2}-\nu^{2}\varphi_{3}^{2})+\varphi^{2}(\varphi_{\eta\eta}-\nu^{2}\varphi_{33})-\nu\varphi_{3}\varphi_{\eta}-2\varphi\varphi_{3\eta}^{(2)}\right]$ 

Due to the nonlinearity in the solution there emerge all harmonics, higher in  $9 \circ$  , for which the first

harmonic is the source, therefore the solution of Eq.(2) will have the form  $\varphi = \sum_n A_n(\xi,\tau) \sin(n\eta)$ . However, insertion of this expression into (3) and the analysis of equations for each harmonic shows that the amplitude  $A_n$  of higher harmonics is much smaller than the amplitude of the first harmonic, therefore they can be neglected.

Thus, the solution of Eq.(2) in the main approximation will have the form  $\varphi=A(\xi,\tau)\sin\eta$ , where  $A(\xi,\tau)$  is the function weakly dependent on  $\xi$  and  $\tau$  ( $\partial A/\partial \xi$ ,  $\partial A/\partial \tau \ll 1$ ) due to the small values of  $\omega$  and k. The small values of the derivatives of A over  $\xi$  make it possible to simplify the form of the nonlinearity in Eq.(2). As a result, it becomes

$$W_{T} = W_{XX} + W - W^{3} \tag{3}$$

where 
$$T = \delta_0 \tau$$
,  $X = \delta_0^{1/2} \xi$ ,  $A = (2 \delta_0)^{1/2} \nu W$ 

Investigation of equations of this type with localized initial perturbations was first performed in [2] and developed in [3]. The basic results of these works can be formulated as follows. In systems, described by Eq.(3), the transition from an unstable into a stable stationary state occurs due to propagation of the wavefront connecting the regions of these states, the form of the front is determined by the solution of the ordinary second order differential equation and the velocity of the front is constant with logarithmic accuracy and is equal to 2. In its complete form the solution of Eq.(3) reads

$$W(x,T) = W\left[x - 2T + \ln\left(\frac{T}{\Im t}\widetilde{W}^{-1}(i)\right)\right], \quad (4)$$

where  $\widetilde{W}(k)$  is the Fourier transform of the initial condition W(X,T=0). In dimensionless variables the velocity of the front  $C=2\delta_o^{1/2}(K_1K_3)^{1/2}\pi/d\chi^*=\delta_oC_o$  and with typical sizes  $d\sim 10^{-3}-10^{-2}$  cm  $C_o\sim 10^{-2}-10^{-3}$  cm/sec. The characteristic size of the front is of the order

 $d(K_1/K_3)^{1/2} \delta_0^{-1/2}$ . The magnitude of the amplitude of the stable state is  $A \sim \delta_0^{1/2} (K_3/K_1)^{1/2}$ . The characteristic time of the stable state formation comprises the two parts. The first part  $T_1$  is the time of growth of the fastest mode (with K=0) of the initial perturbation:  $T_1 \sim -\ln |\widetilde{W}(0)|$ . The second part is the time the wave covers in the direction  $\infty$  the distance of the order of the sizes of the sample  $\ell$ :  $T_2 \sim 1/C = 1/2$ 

order of the sizes of the sample  $\ell: T_2 \sim L/c = L/2$ , where L is the langth of the sample in dimensionless units. Thus the entire time of the stable state formation is  $T_0 \sim L/2 - \ln |\widetilde{W}(0)|$ .

As the initial condition for the homogeneous case one usually takes the mean quadratic fluctuation of the value of the angle  $\varphi$  in the plane of the layer, i.e.,  $\langle (\Delta W)^2 \rangle$ . Thus,

$$W^{2} = \left[1 + \left(\frac{\Delta W}{2}\right)^{2} - 1\right) \exp(-2T)\right]^{-1}$$
(5)

where in dimensional variables  $\langle (\Delta W)^2 \rangle = \langle (\Delta A)^2 \rangle / \delta_o v_{\sim}^2 / \delta_o V_{\sim}^2$ 

Here  $T_K$  is the temperature,  $k_B$  is the Boltzmann constant. Since for not too large  $\delta_o$  and for the sizes d, typical of the experiment,  $\langle (\Delta W)^2 \rangle$  <<1, the characteristic time of the fluctuation mechanism, as it follows from (5)

$$T_{se} \sim -\frac{1}{2} \ln \langle (\Delta W)^2 \rangle \approx \frac{1}{2} \ln \left( \frac{2 \cdot 10^{-8} |\ln 2\delta_0|}{\delta_0 d} \right)_{(6)}$$

It is evident that the initial state of the specimen, homogeneous in  $\infty$ , is a great simplification of the model. In real conditions there are always inhomogeneities caused by properties of the restricting plates or by edge effects. Therefore the both mechanisms of formation of the stable structure may be in principle realized. To find the contributions coming from these mechanisms it is necessary to compare the times  $T_{\mathbf{o}}$  and  $T_{\mathbf{fe}}$ . Let us estimate these times using the following values of the characteristic parameters for MBBA:  $K_4$  = 6,0 10<sup>-7</sup> dyn,  $K_3$  = 7.5 10<sup>-7</sup> 10<sup>-7</sup> dyn,  $\chi^*$  = 19·10<sup>-2</sup> Poise,  $T_K$  = 300K. Assuming that the size of the specimen is  $\ell \sim d \cdot 10^n$ , the size of the perturbation region is  $\sim d \cdot 10^{-m}$  (remember that the amplitude of the initial state is  $A < \delta_0^{1/2} V < 1$ ), we get in the result  $T_4 \approx -\ell n \left( 4 \delta_0 10^{-m-z} \right)$ ,  $T_2 \approx 1,7 \delta_0^{1/2} 10^n$ . At  $d = 10^{-2}$ cm, m = 1,  $\chi \approx 1$   $A_{in} \sim \delta_0^{1/2} V \cdot 10^{-v}$ 

$\delta_{ullet}$	T <sub>se</sub>	T,	T <sub>2</sub>
10 <sup>-2</sup>	3.6	3.9	1.8 $10^{n-1}$
5·10 <sup>-2</sup>	4.6	3.1	3.9 10 <sup>n-1</sup>
10 <sup>-1</sup>	5.2	2.7	5.5 10 <sup>n-1</sup>

This estimate shows that at  $N\sim 1$  all the times are comparable with each other. At increasing  $\delta_o$  and d, the value of  $T_{\xi\ell}$  also grows, whereas  $T_{\ell}$  descreases. As for  $T_{2}$ , at the same values of the parameters its value is of the order of the magnitude of  $T_{\xi\ell}$ . It should be mentioned, however, that the estimate of  $T_{2}$  holds for the case when the initial perturbation was unique and was localized at one of the edges of the

specimen. Yet, in reality in a specimen there may be a considerable number of perturbation centers generating the corresponding waves. Then  $T_2$  will descrease proportionally to this number. Thus, these estimates show that there can possibly exist certain conditions when formation of the finite structure in the Frederiks transition results from formation of the stable structure in the finite regions of the specimen and from further expansion of these regions.

In the result of the interaction of wavefronts in the system walls can be formed. Their characteristic size will evidently be of the order of the size of the front (i.e.,  $d_{\rm w} \sim d/\nu \delta_{\rm o}^{4/2}$ ) and must grow with decreasing  $\delta_{\rm o}$ This dependence was also obtained theoretically from energy considerations in [4,5]. Such walls were observed in experiment and reported in [6] where the dependence  $d_{\mathbf{w}}^{-2} \sim \delta_{\mathbf{o}}$  was observed and the sizes of the walls were in good agreement with the values predicted in this work. In this paper the values of the velocity  ${\cal S}$ were measured depending on the values of  $\delta_{\mathbf{o}}$ found that at  $\delta_a > 0.1$ ,  $S \approx 3.10^{-5}$  and at decreasing it noticeably grows. In terms of the proposed mechanism, the motion of walls can be described in the following fashion. When two oppositely moving waves form a wall, then in the main approximation since the velocities of the motion of the fronts are identical and equal 2 in dimensionless variables, its center must be non-moving,

fixed. Yet, as is clear from (4), there are also logarithmic additions to the velocity, associated with the initial perturbation. It is at the expense of the difference of these logarithms and of the difference of the initial

conditions for each front that a non-zero, much smaller than the value of the velocity of the front, velocity of the wall occurs. Actually S << C. This can also qualitatively account for the growth of S at decreasing

- $\delta_o$  . Since corrections to the velocity of the front have a factor  $T^{-1}$  before the logarithm, their difference is  $T^{-1}$ -proportional. At the same time in virtue of the definition of T in dimensional variables, it is  $\delta_o$ -proportional which actually generates the corresponding divergence in S .
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