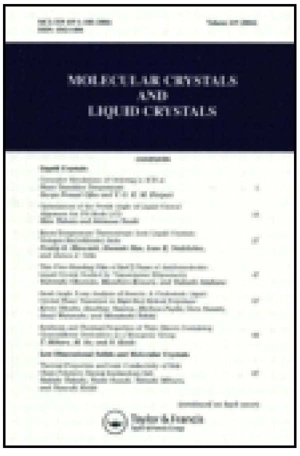

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Freedericksz Transition in a Film of Nematic Liquid Crystal in the Magnetic Field with Pretilt Angle

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The Freedericksz transition in the magnetic field with pretilt angle is theoretically investigated. Assuming the amplitude of director distortion ξ_m is small, three characteristic fields H_{ci} ($i = 1, 2, 3$) caused by nontrivial value of the pretilt angle θ_0 are given as functions of θ_0 and the elastic constants. Drawing of free energy as a function of ξ_m leads to clear understanding of these characteristic fields and the dynamics of director states. The relaxation laws of ξ_m after switching the field are expressed in the concise form.

1. INTRODUCTION

The Freedericksz transition in a film of nematic liquid crystal, induced by an applied magnetic field, is usually discussed in the simple geometry where the initial director orientation is chosen to be homogeneous or homeotropic and the field is applied in the direction normal to the initial director configuration. With increase in the field strength in this geometry, the initial uniform state becomes unstable at a threshold field H_c, and the state with distorted director field will be stabilized. It is interesting to note here that two independent modes of distortion with opposite signs are possible at the fields $H > H_c$: the molecular axis tends to be reoriented parallel to the applied field by tilting its head or tail. However, the free energy for these two modes is degenerate, and the second order transition will be expected at $H = H_c$.

The situation becomes considerably different when we introduce a pretilt bias angle in the initial director configuration. In the presence of a magnetic field normal to this director configuration, the system may show the Freedericksz transition also in this case. However the presence of the pretilt angle yields the symmetry breaking terms in the free energy, and the degeneracy of the two modes of distortion will be lifted up. This means that one of the two modes becomes more stable than the other, and the Freedericksz transition here acquires the first order nature with two different threshold fields.

Nehring[1] was the first to derive the expression of one of these threshold field (to be called H_{c1} in our following treatment). Onnagawa *et al.*[2,3] pointed out the presence of the second threshold field H_{c2} slightly below H_{c1}. Furthermore these authors performed a series of important and interesting experiments to confirm the presence of two different modes of distortion.

With these results in mind, it seems us necessary to have a theory, not restricting just in the vicinity of the threshold field H_{c1}, by taking account of the effects of the higher order terms in the free energy. It would be also interesting to study the dynamical characteristics of this system when the strength of the applied field is abruptly switched from one value to the other.

In the next section (§2), we summarize the results of analysis of the static properties with a special stress on the analytical expressions of the quantities like H_{c2}, characterizing the phase diagram of our system. A relaxation equation for the maximum amplitude of distortion, induced by a switching of the field, is derived in §3, and this is applied to deduce the time constants, characterizing the processes of growth or relaxation of distortions. The results of the paper are briefly summarized and discussed in the final section.

2. BASIC EQUATIONS AND STATIC PROPERTIES

Let us consider a layer of nematic liquid crystal of thickness d confined between the boundary plates at $z = 0$ and d. In the absence of the applied field, the director field $\mathbf{n}_0$ is assumed to be uniform and lies in the x-z plane, spanning an angle θ_0 with x-axis lying in the plane parallel to the boundary plate

$$\mathbf{n}_0 = [\cos\theta_0, 0, \sin\theta_0].$$

This configuration will be driven to a distorted state

$$\mathbf{n}(z) = [\cos\theta(z), 0, \sin\theta(z)]$$

in the presence of an applied field $\mathbf{H}$ in the direction normal to $\mathbf{n}_0$

$$\mathbf{H}(t) = H(t)[-\sin\theta_0, 0, \cos\theta_0].$$

The director angle θ becomes time dependent, when we consider the distortion in the director just after switching the field.

The motion of distortion in the director angle $\xi(z,t) = \theta(z,t) - \theta_0$ is described by

$$\begin{aligned}\gamma_1\, \partial\xi/\partial t &= -\delta F/\delta\xi \\ &= f(\xi)\,\partial^2\xi/\partial z^2 + \tfrac{1}{2}\partial f/\partial\xi(\partial\xi/\partial z)^2 + \tfrac{1}{2}\chi_a H^2 \sin 2\xi\end{aligned} \tag{2.1}$$

where F is the free energy functional, measured from the value in the absence of the field

$$F = \int_0^d dz\left[\tfrac{1}{2}f(\xi)(\partial\xi/\partial z)^2 - \tfrac{1}{2}\chi_a H^2 \sin^2\xi\right]. \tag{2.2}$$

The function $f(\xi)$ is introduced by

$$\begin{aligned}f(\xi) &= K_1\cos^2(\xi + \theta_0) + K_3\sin^2(\xi + \theta_0) \\ &= K_3\left[1 - \kappa\cos^2(\theta_0 + \xi)\right],\end{aligned} \tag{2.3}$$

where the parameter $\kappa = (K_3 - K_1)/K_3$ describes the competition between the splay and bend components in our distortion. In these equations, $\gamma_1(>0)$ is the rotational viscosity and χ_a is the anisotropy of diamagnetic susceptibility of the sample nematic liquid crystal.

Let us confine ourselves to the static distortion $\partial\xi/\partial t = 0$ throughout this section. In this case, the first integral of Eq. (2.1) is easily found

$$f(\xi)(d\xi/dz)^2 = \chi_a H^2\left[\sin^2\xi_m - \sin^2\xi\right], \tag{2.4}$$

where it is assumed that $\xi = \xi_m$ and $d\xi/dz = 0$ at $z = d/2$. The equilibrium director distortion $\xi(z)$ is determined by the condition

$$\begin{aligned}&z(\chi_a/K_3)^{1/2}H \\ &= \operatorname{sgn}(\xi_m)\int_0^{\xi} d\xi'\left\{\left[1 - \kappa\cos^2(\theta_0 + \xi')\right]/\left[\sin^2\xi_m - \sin^2\xi'\right]\right\}^{1/2}\end{aligned} \tag{2.5}$$

at a given magnetic field. Putting $z = d/2$ and $\xi = \xi_m$, we get

$$(d/2)(\chi_a/K_3)^{1/2}H$$
$$= \operatorname{sgn}(\xi_m)\int_0^{\xi_m} d\xi'\left\{\left[1 - \kappa\cos^2(\theta_0 + \xi')\right]/\left[\sin^2\xi_m - \sin^2\xi'\right]\right\}^{1/2} \quad (2.6)$$

which relates the parameter ξ_m to the field H. In particular, the first critical field H_{c1} is obtained by taking the limit $\xi_m \to 0$

$$H_{c1} = H_{c0}\left[1 - \kappa\cos^2\theta_0\right]^{1/2} \quad (2.7)$$

in accord with Nehring's result[1] where $H_{c0} = (\pi/d)(K_3/\chi_a)^{1/2}$ is the critical field in the homeotropic geometry $\theta_0 = \pi/2$.

Considering the case where the field H is not so far from H_{c1} and ξ_m is still small, one can expand the r.h.s. of Eq. (2.6) in powers of ξ_m. To do this, it is convenient to introduce a new variable ψ by $\sin\psi = \sin\xi/\sin\xi_m$ and rewrite the Eq. (2.6) in the form

$$\left(1 - \kappa\cos^2\theta_0\right)^{1/2}(\pi/2)(H/H_{c1})$$
$$= \int_0^{\pi/2} d\psi\left\{\left[1 - \kappa\cos^2(\theta_0 + \xi)\right]/\left[1 - \sin^2\xi\right]\right\}^{1/2} \quad (2.8)$$

Retaining the terms up to the second order, we obtain the result

$$\begin{aligned}(H - H_{c1})/H_{c1} &= \beta\xi_m + \gamma\xi_m^2\\ &= \gamma(\xi_m + \beta/2\gamma)^2 - \beta^2/4\gamma,\\ \beta &= (1/\pi)\left[\kappa\sin 2\theta_0/\left(1 - \kappa\cos^2\theta_0\right)\right],\\ \gamma &= (1/4)\left[(1 - \kappa)/\left(1 - \kappa\cos^2\theta_0\right)^2\right]. \quad (2.9)\end{aligned}$$

Based on the above expression, we plot in Figure 1 a phase diagram, representing the relation between the maximum amplitude of equilibrium distortion ξ_m and applied field H. This tells us that two branches of distortion I and II with maximum amplitudes ξ_I and ξ_{II} are possible once the field exceeds the second critical field H_{c2}, given by

$$\begin{aligned}(H_{c2} - H_{c1})/H_{c1} &= -\beta^2/4\gamma\\ &= -(1/\pi^2)\kappa^2\sin^2 2\theta_0/(1 - \kappa). \quad (2.10)\end{aligned}$$

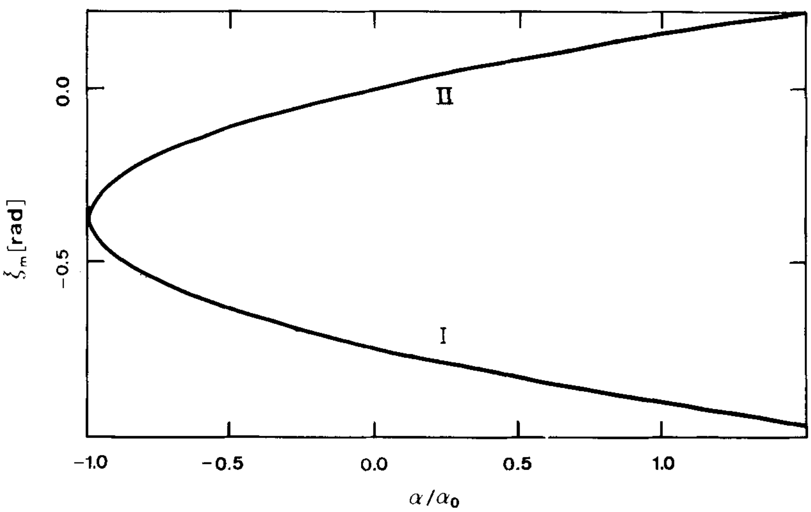

FIGURE 1 The maximum amplitude of distortion ξ_m as a function of the reduced field $\alpha/\alpha_0 = (H - H_{c1})/(H_{c1} - H_{c2})$: $\theta_0 = \pi/8$ and $\kappa = 0.65$.

As is seen from Eq. (2.9), the explicit forms of ξ_{I} and ξ_{II} are given by

$$\xi_{\text{I}} = -\beta/(2\gamma) - \left[\left(\beta^2/4\gamma + H/H_{c1} - 1\right)/\gamma\right]^{1/2},$$

$$\xi_{\text{II}} = -\beta/(2\gamma) + \left[\left(\beta^2/4\gamma + H/H_{c1} - 1\right)/\gamma\right]^{1/2}. \quad (2.11)$$

It is seen that two branches are degenerate at $H = H_{c2}$, and ξ_{I} vanishes at $H = H_{c1}$.

We are going to calculate the free energy of each branch as a function of the external field in order to see their stability. To do this, we make use of the first integral (2.4) to rewrite the free energy in (2.2) only in terms of variable ξ. Then we expand it in powers of ξ_m, in a similar way as we did in Eq. (2.6). Retaining the terms up to the fourth order, we obtain the result

$$\left[\chi_a H_{c1}^2(d/\pi)\right]^{-1} F = -(1/6)\beta\xi_m^3\left[1 + (3\gamma/2\beta)\xi_m\right]. \quad (2.12)$$

We show in Figure 2 how the free energy of the branch I or II depends on the applied field with help of Eqs. (2.10) and (2.12). It is evidently seen that the uniform undistorted state is stable up to a field H_{c3} which is slightly larger than and very close to H_{c2}; it is easily seen that $(H_{c1} - H_{c3})/(H_{c1} - H_{c2}) = 8/9$ from the conditions $F = 0$ and $\partial F/\partial \xi_m = 0$. With further increase in the field the distorted branch I

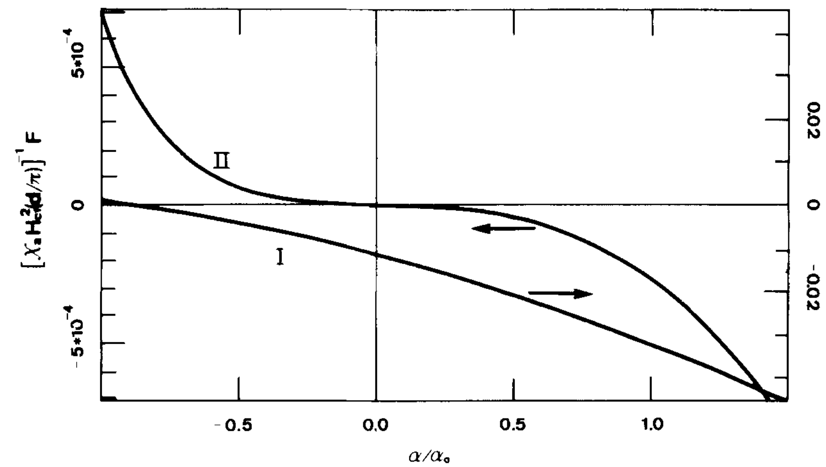

FIGURE 2 The free energy of the branches I and II as functions of α/α_0: $\theta_0 = \pi/8$, $\kappa = 0.65$.

becomes stable in place of undistorted state which is now metastable. When the field exceeds H_{c1}, the second branch II becomes metastable state in place of the undistorted state. We show in Figure 3 how the characteristic fields H_{ci} $(i = 1, 2, 3)$ depend on the bias angle θ_0.

It is instructive to note that the free energy F in Eq. (2.12) is expressed in the form

$$\Psi = \left[\chi_a H_{c1}^2 (d/\pi)\right]^{-1} F = -\alpha \xi_m^2/2 + \beta \xi_m^3/3 + \gamma \xi_m^4/4 \quad (2.13)$$

with aid of Eq. (2.9) where $\alpha = (H - H_{c1})/H_{c1}$. This enables us to draw Ψ as a function of ξ_m at a given value of the applied field, as is shown in Figure 4. The asymmetry in this plot is entirely due to the third order term induced by the presence of the pretilt angle θ_0, as is stressed in Introduction.

Let us briefly comment on the capacitance C for a weak electric field perpendicular to the boundary.

$$1/C = (2/\pi)(H_{c1}/H)\int_0^{\xi_m} d\xi \left\{\left[1 - \kappa \cos^2(\theta_0 + \xi)\right]/\left[\sin^2\xi_m - \sin^2\xi\right]\right\}^{1/2}\left[1 + \epsilon \sin^2\theta_0\right]/\left[1 + \epsilon \sin^2(\theta_0 + \xi)\right]. \quad (2.14)$$

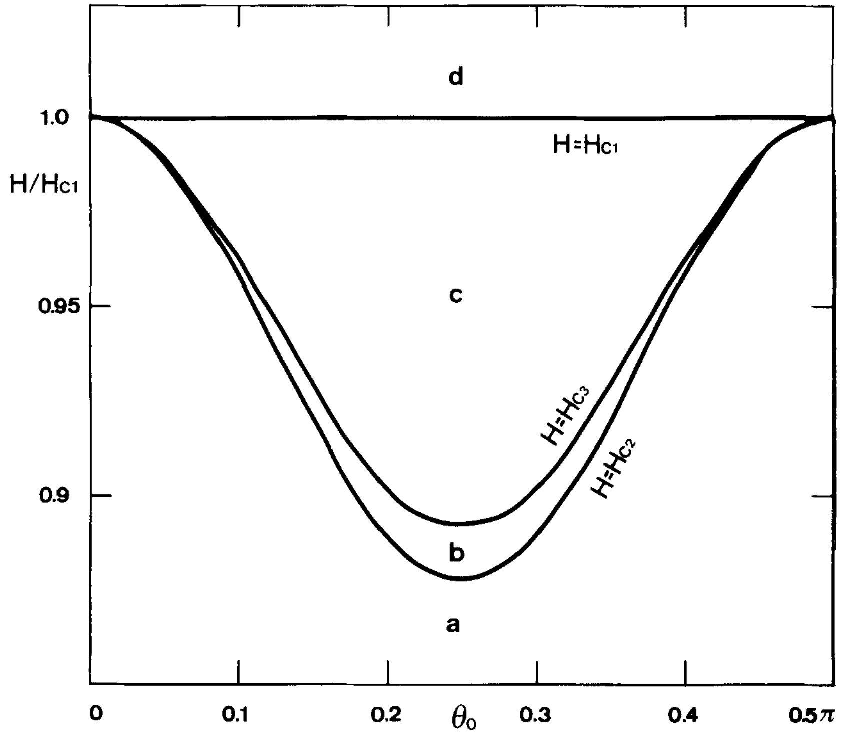

FIGURE 3 Stability of various director configurations as the function of magnetic field and tilt angle ($\kappa = 0.65$). The plane is divided into four regions: (a) uniform state is stable and no distorted states can exist, (b) uniform state is stable and distorted state along branch I is metastable, (c) uniform state is metastable and branch I stable, (d) uniform state is unstable, branch I stable and branch II metastable.

Here C is normalized by the value in the absence of field and $\epsilon = (\epsilon_2 - \epsilon_1)/\epsilon_1$; ϵ_1 and ϵ_2 represent the dielectric constants, measured along (ϵ_1) or normal (ϵ_2) to the nematic axis. With the help of Eq. (2.9) we find

$$1/C = 1 - C_1(H_{c1}/H)(C_2\xi_m + C_3\xi_m^2) \tag{2.15}$$

where

$$C_1 = \epsilon/(1 + \epsilon\sin^2\theta_0)$$
$$C_2 = (2/\pi)\sin 2\theta_0$$
$$C_3 = (1/2)\cos 2\theta_0 + (1/4)\pi\beta\sin 2\theta_0 - (\sin^2 2\theta_0/2)C_1.$$

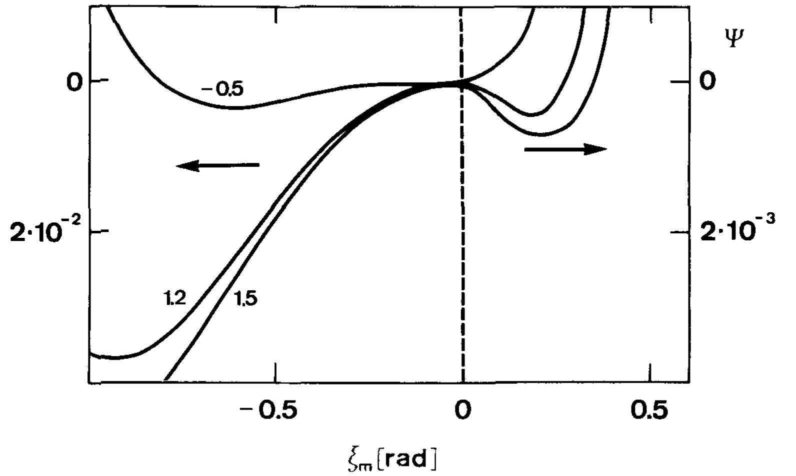

FIGURE 4 The free energy $\Psi = [\chi_a H_{c1}^2(d/\pi)]^{-1}F$ as a function of ξ_m with fixed field strength (a) $\alpha/\alpha_0 = -0.5$ (b) 1.2 (c) 1.5: $\theta_0 = \pi/8$, $\kappa = 0.65$.

As is clearly seen in Eq. (2.15), the pretilt angle gives rise to the contribution linear in ξ_m which takes the different value for the branches I and II.

In closing this section, let us briefly touch upon the space dependence of distortion $\xi(z)$ which is to be determined from Eq. (2.5). Again we can expand the integral there in powers of ξ_m. Retaining the lowest order correction, we get the result

$$\xi(z) = \xi_m\{\sin(\pi z/d) + \tfrac{1}{2}\pi\beta\xi_m\cos(\pi z/d)[\cos(\pi z/d) + (2z/d - 1)]\} \tag{2.16}$$

where use has been made of Eq. (2.9) to eliminate the magnetic field. It is seen that the above expression of $\xi(z)$ satisfies all boundary conditions imposed on it, as it should be.

3. DYNAMICAL PROPERTIES

In this section, we are going to study a dynamical motion of director field, induced by an abrupt switching of the external field. In order to fix our idea, we suppose that the system is initially in equilibrium on the stable branch I (or on the metastable branch II) at a given field H'. Here the maximum amplitude of distortion ξ_m takes the value $\xi_{\mathrm{I}}(H')$ or $\xi_{\mathrm{II}}(H')$, as is given in Eq. (2.10). When the field is abruptly switched to a new value H at $t = 0$, the above director configuration is no longer in equilibrium and a relaxation motion of ξ_m tending to a new equilibrium value will be induced.

To derive the relaxation equation for $\xi_m(t)$, we assume that the space and time dependence of the quantity $\sin\xi(z, t)$ can be decomposed in the form $\sin\xi(z, t) = \sin\xi_m(t)\sin\psi(z)$. This is equivalent to saying that the space structure of the director field is kept in the form as in the static case even in the relaxation process. Then we can derive from Eq. (2.1) the following equation.

$$\begin{aligned}\gamma_1 \ln\left[(1-\sin^2\xi)/(1-\sin^2\xi_m)\right] d(\ln|\sin\xi_m|)/dt \\ +f(\xi)(\partial\xi/\partial z)^2 - \chi_a H^2(\sin^2\xi_m - \sin^2\xi) = 0. \quad (3.1)\end{aligned}$$

We have imposed the condition $[\partial\xi/\partial z]_{z=1/2d} = 0$ at any time $t > 0$. Now we limit ourselves in the case of small $\xi_m(t)$, and cast Eq. (3.1) in the form

$$f(\xi)(\partial\xi/\partial z)^2 - \left[\chi_a H^2 - \gamma_1(d\xi_m/dt)/\xi_m\right](\sin^2\xi_m - \sin^2\xi) = 0. \quad (3.2)$$

Separating the space dependence and integrating over z or equivalently over ψ, we obtain

$$\begin{aligned}&\left[(H/H_{c1})^2 - \tau_0(d\xi_m/dt)/\xi_m\right]^{1/2} \\ &= (2/\pi)(1-\kappa\cos^2\theta_0)^{-1/2}\int_0^{\pi/2} d\psi \\ &\quad\times\left\{\left[1-\kappa\cos^2(\xi+\theta_0)\right]/(1-\sin^2\xi)\right\}^{1/2}, \quad (3.3)\end{aligned}$$

where a characteristic time $\tau_0 = \gamma_1/(\chi_a H_{c1}^2)$ is introduced. The integral on the r.h.s. is expanded in powers of ξ_m, and the l.h.s. is also expanded to yield the following relaxation equation for $\xi_m(t)$

$$\begin{aligned}d\xi_m/dt &= -2(H/H_{c1})\xi_m(-\alpha + \beta\xi_m + \gamma\xi_m^2)/\tau_0 \\ &= -2(H/H_{c1})\gamma\xi_m(\xi - \xi_{\mathrm{I}})(\xi - \xi_{\mathrm{II}})/\tau_0 \quad (3.4)\end{aligned}$$

Here the expressions of quantities α, β and γ as well as the new equilibrium values of maximum distortion $\xi_{I,II} = \xi_{I,II}(H)$ are given in the last section.

We have assumed that our system was initially in equilibrium in the field H' on the branch I or II. Thus the initial value $\xi_0 = \xi_m(0)$ is given by $\xi_I(H')$ or $\xi_{II}(H')$. We can integrate Eq. (3.4) under this condition to obtain the result

$$(1/\xi_I)\ln[(1 - \xi_I/\xi_m)/(1 - \xi_I/\xi_0)] \\ -(1/\xi_{II})\ln[(1 - \xi_{II}/\xi_m)/(1 - \xi_{II}/\xi_0) \\ = -2(H/H_{c1})\gamma(\xi_I - \xi_{II})(t/\tau_0). \quad (3.5)$$

In Figure 5, we plot the relaxation profile of $\xi_m(t)$ which is initially on the branch I in the reduced field $\alpha = (H_{c1} - H)/H_{c1} = 1.5\alpha_0$ $[\alpha_0 = (H_{c1} - H_{c2})/H_{c1}]$ and switched to a new equilibrium value in the field $\alpha = 1.2\alpha_0$ or $-0.5\alpha_0$. As is clearly seen from these plots and Eq. (3.5), the distortion $\xi_m(t)$ obeys the relaxation laws

$$\xi_m(t) = \xi_{I,II}[1 + (\xi_{I,II}/\xi_0 - 1)\exp(-t/\tau_{I,II})]^{-1}, \quad (3.6)$$

in the long time limit. It should also be noted that these asymptotic laws are valid in the whole stage of relaxation, when the magnitude of change in the field is small (see Figure 5a). The relaxation times $\tau_{I,II}$ are expressed by

$$1/\tau_I = 2(H/H_{c1})\gamma\xi_I(\xi_I - \xi_{II})/\tau_0 \quad (3.7a)$$

$$1/\tau_{II} = 2(H/H_{c1})\gamma\xi_{II}(\xi_I - \xi_{II})/\tau_0 \quad (3.7b)$$

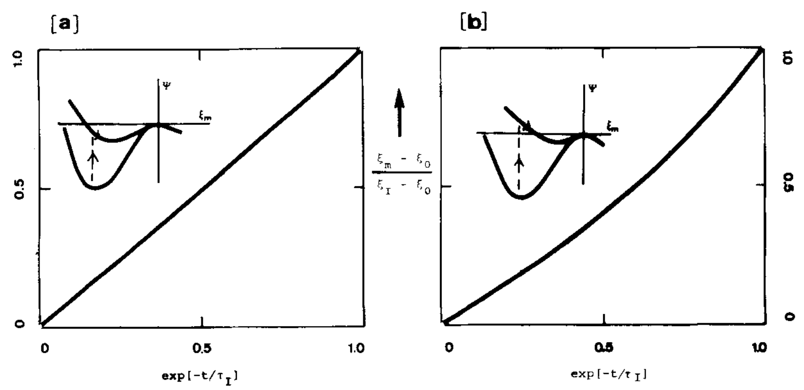

FIGURE 5 Relaxation of ξ_m after switching field strength (a) $\alpha/\alpha_0 = 1.5 \rightarrow 1.2$, (b) $\alpha/\alpha_0 = 1.5 \rightarrow -0.5$ both for $\theta_0 = \pi/8$, $\kappa = 0.65$.

which characterize the time scales of transition between different equilibrium states at the fields H' and H. It is interesting to note that the transition to a state in the vicinity of H_{c2} along the branch I takes a very long time. This is an example of the phenomena of critical slowing down familiar in the physics of critical phenomena: To be more explicit, we note that $1/\tau_{\mathrm{I}}$ is expressed in the form

$$1/\tau_{\mathrm{I}} \simeq 4(1/H_{c1})[(H_{c1} - H_{c2})(H - H_{c2})]^{1/2}/\tau_0 \qquad (3.8a)$$

in the limit $H \to H_{c2}$. Similarly, the relaxation along the second branch II shows the critical slowing down in the limit $H \to H_{c1}$

$$1/\tau_{\mathrm{II}} \simeq 2(1/H_{c1})(H - H_{c1})/\tau_0. \qquad (3.8b)$$

Finally, we briefly comment on the case where the initial field H' ($> H_{c1}$) is switched to a new value H ($< H_{c2}$). In this case, the distortion $\xi_m(t)$ tends to 0 according to the relaxation law

$$\mathrm{Re}\ln[\xi_0(\xi_m - \xi_{\mathrm{I}})/\xi_m(\xi_0 - \xi_{\mathrm{I}})]/\xi_{\mathrm{I}}(\xi_{\mathrm{I}} - \xi_{\mathrm{I}}^*) = -2\gamma(H/H_{c1})(t/\tau_0) \qquad (3.9)$$

as is derived from Eq. (3.5). Here ξ_{I} stands for a complex number $\xi_{\mathrm{I}} = -[\beta + i(4\gamma\alpha - \beta)^{1/2}]/2\gamma$. Asymptotically, the relaxation law (3.9) tends to the form

$$\xi_m(t) \simeq \xi_0 \exp[-t/\tau]$$

where the relaxation time τ is given by $\tau = \tau_0/(2\alpha H/H_{c1})$.

4. SUMMARY

We have studied static and dynamical characteristics of magnetic field induced Freedericksz transition in a nematic liquid crystal film with a pretilt bias angle in initial director orientation.

Throughout the paper, the amplitude of director distortion is assumed to be small and various characteristics are expressed in the form of expansion in power of this quantity. This approach is particularly useful to pick out the essential physical role of pretilt angle as a source of symmetry breaking force in the present problem. This is of course only possible in the sacrifice of quantitative validity of our theory which is limited in the vicinity of the threshold field H_{c1} or H_{c2}. (A numerical but more quantitatively thorough analysis of the present problem has been reported by Motooka *et al.*[4])

In particular, we have derived the expression of free energy as a function of the maximum amplitude of distortion ξ_m and the field strength H. This enables us to construct a phase diagram of our system in the plane of ξ_m and H, and further to obtain analytical expressions of various characteristic fields H_{ci} ($i = 1, 2, 3$) with following clear-cut interpretation of their physical meaning. The field H_{c2} characterizes the nucleation of the branch I distortion ξ_{I} which becomes the most stable state at H_{c3} in place of undistorted state. At this point of field strength, the value of maximum amplitude ξ_m jumps from 0 to a finite value $-(2/3)(\beta/\gamma)$, resulting in the first order transition instead of the second order one in the ordinary case without pretilt angle. The field H_{c1} is characterized by the fact that the second branch distorted state ξ_{II} becomes metastable instead of undistorted state.

It should also be remarked that the free energy expression (2.13) again characterizes the asymptotic relaxation times of our system for an abruptly switched magnetic field. It is easy to check that the inverse relaxation times $1/\tau_{\text{I, II}}$ in Eq. (3.7) are proportional to the curvatures of free energy $\partial^2\Psi/\partial\xi_m^2$ calculated at the local minima $\xi_{\text{I, II}}$ in the new field. These curvatures vanish at the field H_{c1} or H_{c2}, resulting in the critical slowing down as is discussed in the last section. It will be very interesting to check such behavior experimentally.

Finally let us comment on the dependence of the asymmetry of phase diagram $H_{c1} - H_{c2}$ on the elastic constants. As is seen in Eq. (2.11), this becomes large when the parameter κ approaches to unity. It is known that the bend constant K_3 becomes very large compared with the splay constant K_1 when the system is cooled down[5] to the transition temperature T_{NS} between nematic and smectic phases. Thus we can expect that effect of the pretilt angle will be enhancedly observed in the vicinity of T_{NS}. However this statement must be complemented by a proviso that the Eq. (2.11) does not hold in its form in the limit $\kappa \to 1$. In this limit, the splay distortion may be very easily induced in the vicinity of the walls over the distances far shorter than the film thickness d. Thus the director configuration is dominantly governed by the external field over most part of the sample, resulting in the breakdown of our small amplitude approximation. The details of what happens in this limit are discussed in Appendix. The results of this calculation are summarized in Figure 6. It is clearly seen that H_{c2} can take values less than half of H_{c1} over a wide range of bias angle θ_0 if $\kappa = 1$.

In conclusion, the Freedericksz transition in the nematic liquid crystal film with a pretilt angle provides a lot of interesting properties in common with thermodynamical first order phase transition.

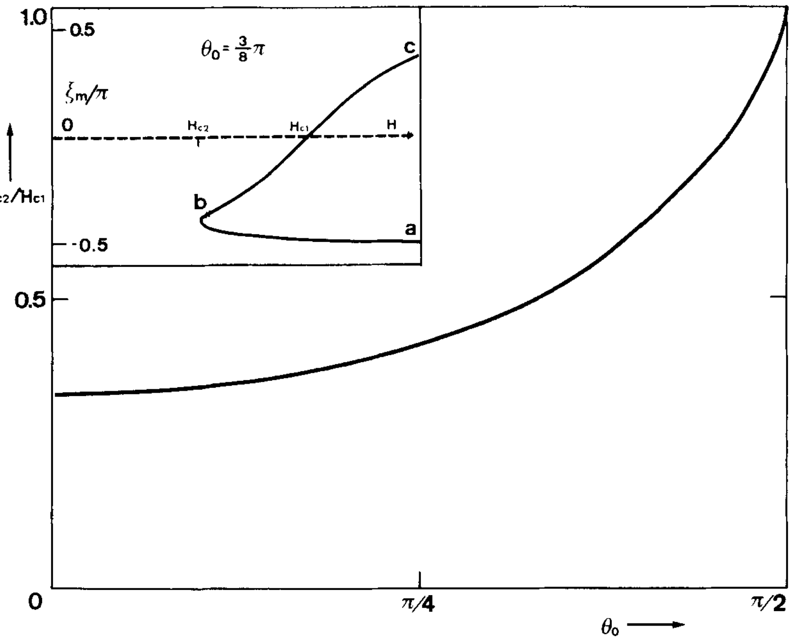

FIGURE 6 The second critical field H_{c2} as a function of θ_0 in the limit of $\kappa = 1$. Inset shows an asymmetric phase diagram with $\theta_0 = (3/8)\pi$ [cf. $\kappa < 1$ case, Figure 1].

APPENDIX

In this Appendix, we are going to discuss the phase diagram in ξ_m-H plane in a special limit $\kappa = 1$. To do this, we first put $\kappa = 1$ in Eq. (2.6), and find that the integral in this equation may be calculated in an elementary manner. The result is written down in the form

$$H/H_{c1} = 1 + (2/\pi)\cot\theta_0 \operatorname{sgn}\xi_m \ln[1 + \sec\xi_m + \tan|\xi_m|] \quad (A.1)$$

for $\xi_m + \theta_0 > 0$, and

$$H/H_{c1} = -(4/\pi)\arcsin(\sin\theta_0/\sin\xi_m) - 1 + (2/\pi)\cot\theta_0\left\{2\ln\left[\cos\theta_0 + \left(\sin^2\xi_m - \sin^2\theta_0\right)^{1/2}\right] - \ln(\sec\xi_m - \tan\xi_m)\right\} \quad (A.2)$$

for $\xi_m + \theta_0 < 0$. These formulae yield a phase diagram as is shown in the inset of Figure 6. The part bc of the curve is obtained from (A.1) while the part ab from (A.2). From this figure and Eq. (A.3), we see that the characteristic field H_{c2} in the present limit is realized for ξ_m such as $\sin \xi_m = -2 \sin \theta_0 / A$ with $A = (3 + \sin^2\theta_0)^{1/2}$, and expressed in the form

$$\begin{aligned} H_{c2}/H_{c1} = {} & (4/\pi)\arcsin(A/2) - 1 \\ & + (2/\pi)\cot\theta_0\left[\ln(\cos\theta_0) + 2\ln(A + \sin\theta_0)\right. \\ & \left. - \ln(\alpha + 2\sin\theta_0) - (1/2)\ln 3\right]. \end{aligned} \tag{A.3}$$

In Figure 6, we show how the ratio H_{c2}/H_{c1} depends on the pretilt angle θ_0. It is clearly seen from this plot that the quantity $H_{c1} - H_{c2}$ becomes comparable to H_{c1} itself and the corresponding value of ξ_m is also of the order of unity, suggesting that the distortion at the second threshold field cannot be treated in the scheme of small amplitude approximation.

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