Flow-alignment instability and slow director oscillations in nematic liquid crystals under oscillatory flow

A.P. Krekhov* and L. Kramer
Physikalisches Institut, Universität Bayreuth, D-95440 Bayreuth, Germany
(Received 23 October 1995)

The nonlinear response of a nematic slab subjected to a rectilinear low-frequency oscillatory Couette and Poiseuille flow is investigated theoretically. We find that under Poiseuille flow and with appropriate alignment conditions by surface anchoring and/or magnetic field a state with slow, spontaneous director rotation appears. This may provide a model for the slow director rotations observed in high-frequency Couette flow.

PACS number(s): 61.30.Cz, 61.30.Gd, 64.70.Md, 47.20.−k

I. INTRODUCTION

In nematic liquid crystals the coupling between the preferred molecular orientation (director $\hat{\mathbf{n}}$) and the velocity field leads to interesting flow phenomena. In a steady velocity field $u(z)$ along the x axis (rectilinear plane shear flow, typically of the Couette or Poiseuille type) the director will, in the absence of other orienting effects, tend to align in the x-z plane (shear plane) at the Leslie angle $\theta_{fl}=\pm\tan^{-1}(\alpha_3/\alpha_2)^{1/2}$ with the x axis for positive/negative shear rate $\partial u/\partial z$ if $\alpha_3/\alpha_2>0$. Here α_3,α_2 are Leslie viscosities [1]. In typical low-molecular weight materials with rodlike molecules one has $\alpha_3/\alpha_2\approx 0.01$. In the usual layer geometry the director is anchored at the boundaries and then one may have interesting instabilities and transitions that have been studied in the past; see, e.g., [1–5]. Also the nonaligning case $\alpha_3/\alpha_2<0$, which is found in some materials, in particular, near a nematic-smectic transition, has been studied. Then one finds a transition to tumbling motion which has also attracted much attention [6–8].

Here we are concerned with oscillatory flow where $u(z,t)$ oscillates symmetrically around zero, which has properties quite different from the steady case. We will consider only situations where the director lies initially in the shear plane. Most typically one deals with a planar (in the plane of the layer, also called homogeneous) or homeotropic (perpendicular to the layer plane) orientation. For low-frequency Couette flow (viscous penetration depth $\sqrt{\eta/\rho\omega}$ larger than the cell thickness d) with its linear velocity profile (uniform shear rate) the director oscillates initially between two positions, which in the flow-aligning case are bounded by the alignment angles $\pm\tan^{-1}(\alpha_3/\alpha_2)^{1/2}$ as the oscillation amplitude becomes large [9]. With increasing flow amplitude one then tends to find experimentally transitions to roll states [10–12], which are not understood very well [12,13] (in the theory the elastic coupling has been neglected, which is a questionable approximation at low frequencies). Actually a simpler mechanism arises when the shear is made elliptical (or circular as a special case), by applying oscillations $x(t)=x_0\sin\omega t$, $y(t)=y_0\cos\omega t$ to one of the confining plates (or by applying the two rectilinear components to the two plates), and this situation has been studied intensively in the past [5,14–17]. The threshold calculated there diverges when the elliptic excitation degenerates into a rectilinear one ($y_0=0$ or $x_0=0$).

In Couette flow with ultrasonic frequencies, where one has strong deviations from the linear velocity profile, a transition from the homeotropically aligned state to a state with a slowly rotating planar component of the director and various wave phenomena (''autowaves'') have been found experimentally [18,19], which are still not understood. We are not aware of experiments on flow-alignment instabilities in oscillatory Poiseuille flow.

Here we will consider theoretically low-frequency Couette and Poiseuille flow and confine ourselves to states that are spatially uniform in the plane of the layer. Thus roll transitions are excluded (they will be discussed elsewhere). In previous work we have analyzed the time-averaged (over the oscillation period) torques acting on the director in spatially homogeneous situations, i.e., with neglect of boundary conditions [20,21]. For low-frequency Couette flow there are no torques whereas for Poiseuille flow there are torques directed away from the flow-alignment angles and, for $\theta>\theta_{fl}$, away from the shear plane. Besides the weakly stable planar state $\hat{\mathbf{n}}=\hat{\mathbf{x}}$ (for flow-aligning materials) there exists a stationary attractor out of the shear plane. It was confirmed by numerical simulations that with homeotropic boundary conditions above a critical flow amplitude an out-of-plane transition indeed occurs leading to the new stationary state [21].

Our present work confirms that homogeneous transitions occur only under Poiseuille flow (and more general flows with nonuniform shear rate) and, for the standard material MBBA (4-methoxybenzylidene-4′-n-butylaniline), only with nonplanar alignment (homeotropic or oblique). After formulating the problem in Sec. II in terms of the standard Erickson-Leslie equations we start out in Sec. III with the simplest possible situation, namely, oblique alignment at one of the flow-alignment angles (under oscillatory flow the two angles correspond to equivalent states). This orientation corresponds to the only in-plane solution with time-independent director. One can show *analytically* that this state remains stable for (low-frequency) Couette flow but suffers an in-plane instability under Poiseuille flow at a critical flow amplitude A_c.

*Permanent address: Physics Department, Bashkirian Research Center, Russian Academy of Sciences, 450025 Ufa, Russia.

From numerical simulations, whose presentation we defer to Sec. V, we find that the bifurcation corresponding to the instability is subcritical (the attractor collides with a saddle) and the time-averaged (over the flow period) director can move towards a (nearly) homeotropic orientation, which, however, is unstable with respect to out-of-plane fluctuations [21]. In fact, the system relaxes to a stationary out-of-plane state, which exists already for amplitudes below the critical one. In this subcritical regime, for decreasing amplitude, the out-of-plane state loses stability through a Hopf bifurcation giving rise to a slow limit-cycle solution with rotating director. The limit cycle disappears via a homoclinic bifurcation from the in-plane saddle-point solution that exists for $A<A_c$.

Unfortunately this interesting scenario can only be traced numerically. However, a much simpler but analogous situation arises when the prealignment is induced by a magnetic field instead of surface anchoring. Disregarding boundaries the previously introduced time-averaging approach can then be used [20,21] and is presented in Sec. IV. The resulting ordinary differential equations allow a phase space analysis and also exhibit the scenario with the slow director rotation.

As discussed in Sec. VI, our results provide a mechanism to understand the slow director rotation found in high-frequency experiments [18,19]. Moreover, one can devise experimentally more accessible situations for low-frequency Poiseuille flow where the combined action of planar surface anchoring and an oblique magnetic field also lead to the slow oscillations.

II. BASIC EQUATIONS

We consider the nematic layer of thickness d to be confined between two infinite parallel plates. If one of the plates is fixed and the other plate oscillates periodically in a parallel direction one obtains oscillatory Couette flow. Oscillatory Poiseuille flow is realized when an alternating pressure gradient is applied in a direction parallel to the layer. We look for solutions of the nematodynamic equations where the director $\hat{\mathbf{n}}$ and the velocity $\hat{\mathbf{v}}$ are functions only of the distance z from the boundaries and time t and then one can write

$$n_x=\cos\theta\,\cos\phi,\quad n_y=\sin\phi,\quad n_z=\sin\theta\,\cos\phi,$$

$$v_x=u(z,t),\quad v_y=v(z,t),\quad v_z=0\,. \tag{1}$$

Clearly the incompressibility condition and normalization $\hat{\mathbf{n}}^2=1$ are satisfied. $\theta(z,t)$ is the angle with respect to the x axis within the flow plane (x-z plane) and $\phi(z,t)$ is the out-of-plane angle.

We use a length scale d and a time scale $1/\omega$ with $\omega/2\pi$ the frequency of oscillatory flow, so that the dimensionless variables are

$$\tilde{z}=z/d,\quad \tilde{t}=\omega t,\quad \tilde{u}=u/d\omega,\quad \tilde{v}=v/d\omega. \tag{2}$$

The equations governing the alignment and the flow taking into account a magnetic field $\hat{\mathbf{H}}=H(m_x,m_y,m_z)$ with $\hat{\mathbf{m}}^2=1$ can be written as [1,22]

$$\theta_{,t}-K(\theta)u_{,z}-\frac{\lambda}{1-\lambda}\cos\theta\,\tan\phi v_{,z}$$
$$=\epsilon[a_1(\theta,\phi)\theta_{,zz}+a_2(\theta,\phi)\theta_{,z}^2+a_3(\theta,\phi)\phi_{,zz}$$
$$+a_4(\theta,\phi)\phi_{,z}^2+a_5(\theta,\phi)\theta_{,z}\phi_{,z}+a_6(\theta,\phi,\hat{\mathbf{m}})h^2], \tag{3}$$

$$\delta u_{,t}=-p_{0,x}+\partial_z\{-(1-\lambda)K(\theta)\cos^2\phi\,\theta_{,t}$$
$$-(1-\lambda)K'(\theta)M(\phi)\phi_{,t}+2c_2(\theta,\phi)\cos\theta M$$
$$\times(\phi)v_{,z}+[c_1(\theta,\phi)+c_2(\theta,\phi)\cos^2\theta\,\cos^2\phi]u_{,z}\}, \tag{4}$$

$$\phi_{,t}-K'(\theta)M(\phi)u_{,z}-\sin\theta\frac{\cos^2\phi-\lambda\,\sin^2\phi}{1-\lambda}v_{,z}$$
$$=\epsilon[b_1(\theta,\phi)\theta_{,zz}+b_2(\theta,\phi)\theta_{,z}^2+b_3(\theta,\phi)\phi_{,zz}$$
$$+b_4(\theta,\phi)\phi_{,z}^2+b_5(\theta,\phi)\theta_{,z}\phi_{,z}+b_6(\theta,\phi,\hat{\mathbf{m}})h^2], \tag{5}$$

$$\delta v_{,t}=\partial_z\{-2\lambda\,\cos\theta M(\phi)\theta_{,t}-\sin\theta(\cos^2\phi-\lambda\,\sin^2\phi)\phi_{,t}$$
$$+2c_2(\theta,\phi)\cos\theta M(\phi)u_{,z}+[c_1(\theta,\phi)$$
$$+c_2(\theta,\phi)\sin^2\phi]v_{,z}\}, \tag{6}$$

where the tildes have been omitted and

$$K(\theta)=\frac{\lambda\,\cos^2\theta-\sin^2\theta}{1-\lambda},\quad \lambda=\frac{\alpha_3}{\alpha_2},$$

$$M(\phi)=\frac{1}{2}\sin\phi\,\cos\phi,\quad \delta=\frac{d^2}{l^2},\quad \epsilon=\frac{1}{\tau_d\omega},\quad l=\sqrt{\frac{-\alpha_2}{\rho\omega}},$$

$$\tau_d=\frac{\gamma_1 d^2}{K_{11}},\quad \gamma_1=\alpha_3-\alpha_2,\quad h=\frac{H}{H_F},\quad H_F=\frac{\pi}{d}\sqrt{\frac{K_{11}}{\mu_0\chi_a}}. \tag{7}$$

Here the Parodi relation [23]

$$\alpha_6-\alpha_5=\alpha_3+\alpha_2 \tag{8}$$

has been used. The notation $f_{,i}\equiv\partial f/\partial i$, $f'(g)\equiv\partial f/\partial g$ has been used throughout and the coefficients $a_i(\theta,\phi)$, $b_i(\theta,\phi)$, $c_i(\theta,\phi)$ are given in Appendix A. H_F is the splay–Fréedericksz-transition field, l the viscous penetration depth, and τ_d the (splay) director relaxation time.

Boundary conditions for the velocities u and v for the oscillatory Couette flow are

$$u(z=+1/2)=a\,\cos t,\quad u(z=-1/2)=0,$$
$$v(z=\pm 1/2)=0\,, \tag{9}$$

where $a=A/d$ with A the displacement amplitude. The (dimensionless) pressure gradient $p_{0,x}$ in Eq. (4) is zero. In the case of Poiseuille flow one has a dimensionless pressure gradient $p_{0,x}=d/(-\alpha_2\omega)(\Delta P/\Delta x)\cos t$ ($\Delta P/\Delta x$ is the applied pressure gradient in physical units) and

$$u(z=\pm 1/2)=0\,,\quad v(z=\pm 1/2)=0\,. \tag{10}$$

This is to be supplemented by the boundary conditions for the director.

III. STABILITY OF THE FLOW-ALIGNMENT SOLUTION

Let us first consider the case of zero magnetic field ($h=0$). Analytic progress is possible for the special case $\theta=\theta_{fl}$, $\phi=0$ at $z=\pm 1/2$, where θ_{fl} is the flow-alignment angle ($\tan^2\theta_{fl}=\lambda$). Then one has the simple flow-alignment solution $\theta=\theta_{fl}$, $u=u_0(z,t)$, $\phi=0$, $v=0$ of Eqs. (3)–(6), where $u_0(z,t)$ (our basic state) satisfies the equation

$$\delta u_{0,t}=-p_{0,x}+Q(\theta_{fl})u_{0,zz}, \tag{11}$$

with

$$2(-\alpha_2)Q(\theta)=\alpha_4+(\alpha_5-\alpha_2)\sin^2\theta + (\alpha_3+\alpha_6+2\alpha_1\sin^2\theta)\cos^2\theta. \tag{12}$$

In order to analyze the stability of the basic state Eqs. (3)–(6) are linearized with respect to the in-plane and out-of-plane perturbations:

$$\theta=\theta_{fl}+\theta_1(z,t),\quad u=u_0(z,t)+u_1(z,t),\quad \phi=\phi_1(z,t),\quad v=v_1(z,t). \tag{13}$$

In the low-frequency range to be considered here one has $\delta\ll 1$ ($\rho\approx 10^3$ kg/m^3, $-\alpha_2\approx 10^{-1}$ N s/m^2, and $d\approx 10^{-4}$ m gives $\delta<1$ for frequencies $f<1$ kHz) and it seems reasonable to neglect the inertial terms in Eqs. (4), (6), and (11). Then from Eq. (11) one has for the basic state in Couette flow

$$u_0=a(z+1/2)\cos t,\quad u_{0,z}=a\cos t, \tag{14}$$

and in Poiseuille flow

$$u_0=a(4z^2-1)\cos t,\quad u_{0,z}=a8z\cos t. \tag{15}$$

In both cases a is the maximal flow amplitude. Eliminating the velocity component v_1 one is left with (θ_1, u_1 do not couple to ϕ_1, v_1)

$$\theta_{1,t}-K'(\theta_{fl})u_{0,z}\theta_1=\epsilon P_1(\theta_{fl})\theta_{1,zz}, \tag{16}$$

$$\phi_{1,t}-\frac{\frac{1}{2}K'(\theta_{fl})R(\theta_{fl})(1-\lambda)-N(\theta_{fl})\sin\theta_{fl}}{R(\theta_{fl})(1-\lambda)-\sin^2\theta_{fl}}u_{0,z}\phi_1 = \epsilon P_2(\theta_{fl})\frac{R(\theta_{fl})(1-\lambda)}{R(\theta_{fl})(1-\lambda)-\sin^2\theta_{fl}}\phi_{1,zz}, \tag{17}$$

where

$$P_1(\theta)=\cos^2\theta+k_3\sin^2\theta,\quad P_2(\theta)=k_2\cos^2\theta+k_3\sin^2\theta,$$
$$2(-\alpha_2)R(\theta)=\alpha_4+(\alpha_5-\alpha_2)\sin^2\theta,$$
$$2(-\alpha_2)N(\theta)=(\alpha_3+\alpha_6+2\alpha_1\sin^2\theta)\cos\theta \tag{18}$$

and the boundary conditions are

$$\theta_1(z=\pm 1/2)=0,\quad \phi_1(z=\pm 1/2)=0. \tag{19}$$

Equations (16) and (17) are uncoupled, so they give two independent criteria for stability with respect to θ_1 and ϕ_1 perturbations. Since Eqs. (16) and (17) are of the same form we consider the general problem

$$Y_{,t}-Bu_{0,z}(z,t)Y=CY_{,zz},\quad Y(z=\pm 1/2)=0 \tag{20}$$

with constants B and C (>0). From Floquet theory the solution of (20) can be written in the form $Y=\exp(\sigma t)y(z,t)$ where σ is the Floquet exponent, which plays the role of the growth rate for the perturbation, and $y(z,t)$ is a 2π-periodic function in t.

The solution for the case of oscillatory Couette flow (14), where $u_{0,z}$ is independent of z, has the form

$$Y=\exp(-C\pi^2 t)\exp(Ba\sin t)\cos(\pi z). \tag{21}$$

Since the constant $C>0$ one has negative growth rate for both perturbations at all amplitudes of the flow. Therefore the solution $\theta=\theta_{fl}$, $\phi=0$ (basic state) is linearly stable.

Let us now consider the more general case when $u_{0,z}=af(z)\cos t$. In particular, $f(z)=8z$ for Poiseuille flow. The marginal stability condition $\sigma=0$ corresponds to the situation where the equation

$$y_{,t}-aBf(z)\cos t\, y=Cy_{,zz},\quad y(z=\pm 1/2)=0 \tag{22}$$

has a 2π-time periodic solution. The coefficient C is proportional to $\epsilon=1/\tau_d\omega$ [see Eqs. (16) and (17)] and is therefore very small for frequencies large compared to the inverse director relaxation time τ_d ($\tau_d\approx 10^2$ s for $\gamma_1\approx 10^{-1}$ N s/m^2, $d\approx 10^{-4}$ m, and $K_{11}\approx 10^{-11}$ N). Thus we will search for a solution of (22) in the form $y=y_0+Cy_1+\cdots$, $C\ll 1$ with 2π-time periodic y_i satisfying the boundary conditions for y. At lowest order C^0 one has

$$y_0=Y_0(z)e^{aBf(z)\sin t}, \tag{23}$$

where the function $Y_0(z)$ remains undetermined at this order. From the boundary conditions follows $Y_0(z=\pm 1/2)=0$. At order C^1 one has

$$y_{1,t}-aBf(z)\cos t\, y_1=y_{0,zz}, \tag{24}$$

which gives

$$y_1=e^{aBf\sin t}\{[Y_0''+\tfrac{1}{2}(aBf')^2Y_0]t-2aBf'Y_0'\cos t - aBf''Y_0\cos t-\tfrac{1}{4}(aBf')^2Y_0\sin 2t\}. \tag{25}$$

From the periodicity condition for y_1 follows that the function $Y_0(z)$ must satisfy the equation

$$Y_0''+\frac{1}{2}(aBf')^2Y_0=0,\quad Y_0(z=\pm 1/2)=0. \tag{26}$$

It is well known that there exists a smallest (real and positive) eigenvalue a for the nontrivial solution of the problem with given function $f'(z)\neq 0$. Therefore for any flow with $u_{0,zz}\neq 0$ one expects instability of the basic state at some critical value of a.

For Poiseuille flow one has $f'=8$ and from Eq. (26) follows $Y_0=\cos kz$, where $k=8aB/\sqrt{2}$. From the boundary conditions one has $k=\pi$ and therefore

$$a_c = \frac{\pi\sqrt{2}}{8B}. \quad (27)$$

Above the critical amplitude a_c the solution of Eq. (20) will grow in time and therefore the flow alignment solution $(\theta=\theta_{fl}, \phi=0)$ becomes unstable. With standard MBBA material parameters (see Appendix B) one finds $B=K'(\theta_{fl})=0.20$ and $B=0.06$ for the θ_1 and ϕ_1 perturbations, respectively, so that the lowest critical amplitude $a_c=2.78$ corresponds to the θ_1 (in-plane) perturbation.

From the above linear stability analysis follows that the flow alignment $(\theta=\theta_{fl},\ \phi=0)$ induced by boundary conditions is stable for low-frequency oscillatory Couette flow. On the other hand, for oscillatory Poiseuille flow this solution becomes unstable at some critical amplitude as well as for a general flow with $u_{0,zz}\neq 0$.

The critical amplitude a_c for the instability of the solution $(\theta=\theta_{fl},\ \phi=0)$ can be also obtained in the framework of our recently developed time-averaging method [20,21]. By introducing a "slow" time $T=\epsilon t$ that modulates the periodic behavior on the "fast" time scale t, so that $\theta=\theta(z,t,T)$, $\phi=\phi(z,t,T)$, one can formulate a systematic perturbation expansion

$$\theta=\theta_0+\epsilon\theta_1+\ \dots, \qquad \phi=\phi_0+\epsilon\phi_1+\ \dots, \quad (28)$$

where all functions $\theta_i,\ \phi_i$ are periodic in t. At order ϵ^0, corresponding to neglect of the elastic coupling, one has the solutions

$$\theta_0=-\tan^{-1}\left\{\lambda^{1/2}\tanh\left[\frac{\lambda^{1/2}}{1-\lambda}g(z,t)+\frac{1}{2}\ln\left|\frac{\lambda^{1/2}+\tan\eta}{\lambda^{1/2}-\tan\eta}\right|\right]\right\}, \quad (29a)$$

$$\phi_0=\tan^{-1}\left\{\tan\chi\left[\frac{K(\theta_0)}{K(\eta)}\right]^{1/2}\right\}, \quad (29b)$$

where $g(z,t)=\int_0^t u_{,z}dt$ and $\theta_0=\theta_0(\eta,g)$, $\phi_0=\phi_0(\chi,\eta,g)$ are periodic functions in t. Thus, from Eqs. (29), one has a continuous two-parameter family of periodic oscillations of θ and ϕ parametrized by their values η and χ at $t=2\pi n/\omega$, which are now allowed to depend on z and slow time T ("slow angles") and are undetermined at this order. Note that θ_0 oscillates around η and ϕ_0 around χ, but, in general, $\langle\theta_0\rangle\neq\eta$ and $\langle\phi_0\rangle\neq\chi$, i.e., the director oscillation is in general not symmetric around its position at $t=2\pi n/\omega$ where the flow displacement reverses. However, for small oscillation amplitude the difference becomes inessential. From the solvability conditions for the inhomogeneous linear equations at first order in ϵ one obtains evolution equations for the slow angles η and χ [see Eqs. (17) and (18) in Ref. [21]]. The linearization of the evolution equations around the flow-alignment angle $\eta=\theta_{fl}+\eta_1(z)$, $\chi=\chi_1(z)$ gives with $u=u_0$ from Eq. (15) for the case of oscillatory Poiseuille flow

$$\eta_{1,zz}+32a^2K'^2(\theta_{fl})\eta_1=0, \qquad \eta_1(z=\pm 1/2)=0. \quad (30)$$

(The equation for χ_1 is omitted.) The solution of Eq. (30) is $\eta_1=\cos\pi z$ and the corresponding amplitude $a_c=\pi\sqrt{2}/8K'(\theta_{fl})$ is exactly the same as Eq. (27) with $B=K'(\theta_{fl})$ for the θ_1 perturbation.

The result obtained here is not unexpected in view of our previous results showing that Poiseuille-type oscillatory flow always exerts a destabilizing torque on the (nearly) flow-aligned director. The present analysis complements the previous one by giving the threshold in a situation with stabilizing boundary conditions. What will the full nonlinear evolution of the director be at amplitudes a of order of the critical one? Within the flow plane it has mainly a tendency towards the homeotropic (time-averaged) position [20], but then there is the possibility of an out-of-plane transition, which could occur already at substantially lower amplitudes [21].

IV. NONLINEAR BULK OSCILLATIONS WITH MAGNETIC FIELD

In order to gain some understanding of the nonlinear director evolution we first replace boundary conditions by a magnetic field of strength h applied in the appropriate direction characterized by $\hat{\mathbf{m}}$. For oscillatory Poiseuille flow one can then resort to a much simpler spatially homogeneous situation. Taking the prescribed velocity field (15) (valid for low frequencies) and following our time-averaging procedure (see Ref. [21]) one obtains from Eqs. (3) and (5) for the slow angles η, χ, which are now spatially uniform, the evolution equations

$$\eta_{,T}=B_1(\eta,\chi,h,\hat{\mathbf{m}}), \quad (31a)$$

$$\chi_{,T}=B_2(\eta,\chi,h,\hat{\mathbf{m}}), \quad (31b)$$

where $T=\epsilon t$ and the functions

$$\begin{aligned}B_1=a^2 32\{&[a_1(\eta,\chi)+a_5(\eta,\chi)M(\chi)]K'(\eta)K(\eta)\\&+a_3(\eta,\chi)M(\chi)[K''(\eta)K(\eta)+K'^2(\eta)M'(\chi)]\\&+a_2(\eta,\chi)K^2(\eta)+a_4(\eta,\chi)[K'(\eta)M(\chi)]^2\}\\&+h^2a_6(\eta,\chi,\hat{\mathbf{m}}),\end{aligned} \quad (32a)$$

$$\begin{aligned}B_2=a^2 32\{&[b_1(\eta,\chi)+b_5(\eta,\chi)M(\chi)]K'(\eta)K(\eta)\\&+b_3(\eta,\chi)M(\chi)[K''(\eta)K(\eta)+K'^2(\eta)M'(\chi)]\\&+b_2(\eta,\chi)K^2(\eta)+b_4(\eta,\chi)[K'(\eta)M(\chi)]^2\}\\&+h^2b_6(\eta,\chi,\hat{\mathbf{m}})\end{aligned} \quad (32b)$$

are obtained from the general expressions in the approximation of lowest-order time-Fourier expansion for the "fast" director oscillations up to the second harmonic of the response to oscillatory flow. The functions a_i, b_i are defined in Appendix A. We remind the reader that θ oscillate around η and ϕ around χ (if there is out-of-plane motion, i.e., if $\chi\neq 0$) and $\theta=\eta$, $\phi=\chi$ whenever the flow displacement goes through zero. The coupled ordinary differential equations (31) exhibit an interesting bifurcation scenario with a regime of slow limit-cycle oscillations when the magnetic field is applied in the flow plane at the flow-alignment angle

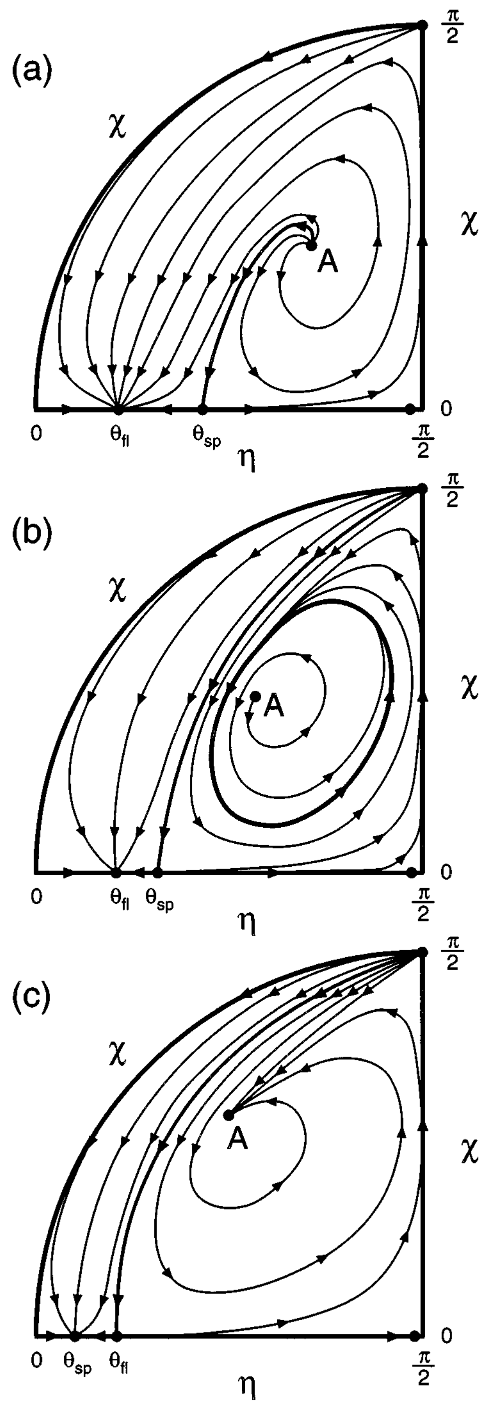

FIG. 1. Plot of trajectories in (η,χ) phase space for the solutions of the evolution equations (31) for $a/h=1$ (a), $a/h=2$ (b), and $a/h=3$ (c). Magnetic field h at the flow-alignment angle θ_{fl}. Note that the director performs rapid oscillations with the external frequency $\omega/2\pi$ around the position (η,χ); see Eqs. (29).

θ_{fl}. The trajectories of the system (31) in (η,χ) phase space are plotted schematically for this case in Fig. 1 for different values of a/h [from Eqs. (32) one sees that this is the relevant control parameter when time is rescaled appropriately]. For values of $a/h<1.9$ (for MBBA parameters) [Fig. 1(a)], corresponding to a strong effect of the magnetic field compared to the influence of the oscillatory flow, one has only one attractor $(\eta=\theta_{fl},\ \chi=0)$, corresponding to the flow-alignment solution $(\theta=\theta_{fl},\ \phi=0)$ whereas all the other fixed points are unstable. In particular, there is a saddle point $(\eta=\theta_{sp},\ \chi=0)$ and an unstable spiral point A in an out-of-plane position (note that by the symmetry $\eta\rightarrow-\eta$ and, separately, $\chi\rightarrow-\chi$, one has twofold and fourfold degeneracy). With increasing a/h the flow-alignment solution becomes unstable and a large stable limit cycle appears through a homoclinic bifurcation from the saddle point at $a/h=1.9$ [Fig. 1(b)]. This limit cycle corresponds to a slow-time periodic out-of-plane motion of the time-averaged (over the oscillatory flow period) director orientation. Further increase of a/h leads to a reduction of the limit cycle and increase of its frequency. It disappears at $a/h=2.27$ through a forward Hopf bifurcation from the spiral point A (we have followed the bifurcation in the reverse sense). Beyond that point A is stable [Fig. 1(c)] and one has a constant time-averaged out-of-plane director orientation which is characterized (approximately) by η_A, χ_A. In addition, θ_{sp} and θ_{fl} cross through each other at $a/h=2.52$ (for MBBA parameters), thereby exchanging their stability properties.

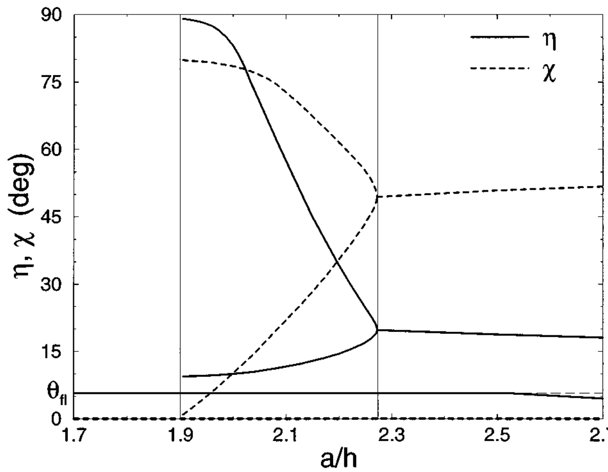

FIG. 2. Bifurcation diagram for the solutions $(\eta,\ \chi)$ of the system (31). Magnetic field h at the flow-alignment angle θ_{fl}.

The bifurcation diagram for the stable solutions (η,χ) of the system (31) is plotted in Fig. 2. In the region of the slow-time oscillations the minima and maxima of η and χ are given. The phenomenon of slow-time director oscillations exist in a narrow region of the parameter a/h and is very sensitive to the elastic constant anisotropy. Thus, in the one-constant approximation $(K_{11}=K_{22}=K_{33})$ one has no out-of-plane attractors.

V. NUMERICAL SIMULATIONS

Direct simulations for oscillatory Couette and Poiseuille flow were performed using central finite differences for the spatial derivatives and the predictor-corrector scheme for the time discretization. All calculations were made for MBBA material parameters (see Appendix B) and flow frequencies 5 Hz$\leq f\leq$100 Hz.

For flow-alignment boundary conditions $\theta(z=\pm1/2)=\theta_{fl}$, $\phi(z=\pm1/2)=0$ the calculations for the Couette case confirm that there are no homogeneous in-plane and out-of-plane instabilities of the solution $\theta=\theta_{fl}$, $\phi=0$ up to large flow amplitudes $(a=10)$. Also for homeotropic and planar

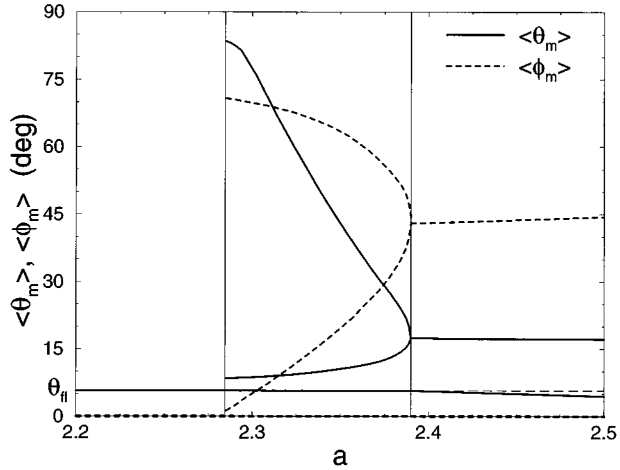

FIG. 3. Bifurcation diagram for $\langle \theta_m \rangle$, $\langle \phi_m \rangle$. Flow alignment boundary conditions; frequency of Poiseuille flow $f = 10$ Hz.

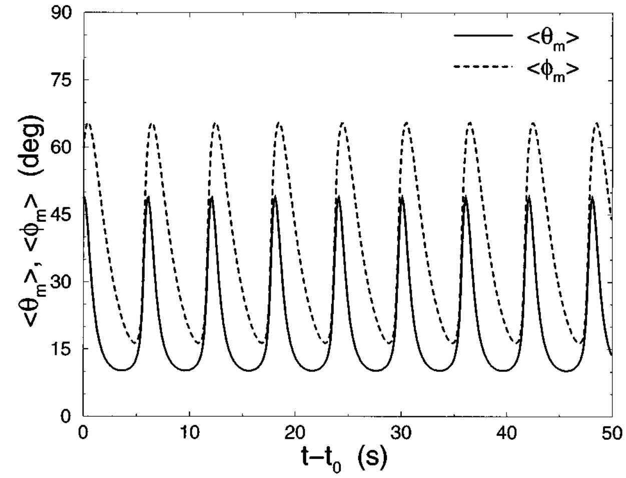

FIG. 4. Slow-time out-of-plane director oscillations. Frequency of Poiseuille flow $f = 10$ Hz; amplitude $a = 2.325$.

boundary conditions no instabilities were found, in agreement with the predictions of our recent analysis [20,21].

The stability properties of in-plane oscillations with respect to out-of-plane motion under oscillatory Poiseuille flow have already been studied for homeotropically oriented nematics [21]. In the frequency range considered we did not find an essential difference in the critical amplitude between the results obtained using the prescribed velocity field [21] and full numerical simulations with the self-consistent velocity field. Note that for steady Poiseuille flow there is also an out-of-plane transition in the homeotropic configuration [8]. The velocity threshold there was found to be lower than for the oscillatory flow.

For planar boundary conditions the (small) in-plane director oscillations (between $\pm \theta_{fl}$) are found to be stable under oscillatory Poiseuille flow with respect to out-of-plane distortions up to large values of the flow amplitude.

We find that the flow-alignment solution $\theta = \theta_{fl}$, $\phi = 0$ becomes unstable in the case of oscillatory Poiseuille flow at some critical amplitude corresponding to a critical pressure gradient $\Delta P/\Delta x$. The critical amplitude for the instability decreases slightly with increasing frequency ($2.5 \geqslant a_c \geqslant 2.3$ for 5 Hz$\leqslant f \leqslant$100 Hz) which is near the value $a_c = 2.78$ obtained from the linear stability analysis.

The in-plane and out-of-plane director distortions can be described by the averaged (over the oscillatory flow period) angles $\langle \theta_m \rangle$ and $\langle \phi_m \rangle$, respectively, taken at the midplane of the nematic layer ($z = 0$). Starting with different initial director distributions the stable long-time solutions of the system (3)–(6) have been computed. The bifurcation diagram as a function of the oscillatory Poiseuille flow amplitude a is shown in Fig. 3. In a narrow region of the flow amplitudes slightly below the out-of-plane instability threshold the solutions with the slow-time oscillations of the director distortions exist (the minima and maxima of $\langle \theta_m \rangle$ and $\langle \phi_m \rangle$ are plotted). Clearly the situation is analogous to that discussed in the preceding section, where the boundary conditions are replaced by a magnetic field, but the range of existence of the slow-time oscillations here is smaller. The typical temporal evolution of $\langle \theta_m \rangle$, $\langle \phi_m \rangle$ is shown in Fig. 4. The period of the oscillations is of the order of the director relaxation time τ_d (in physical units). In Fig. 5 we plot the period T of the slow-time oscillations as a function of amplitude a for different frequencies of the oscillatory Poiseuille flow. The range of existence of the slow-time oscillatory solutions increases slightly with increasing flow frequency.

From the experimental point of view a more realistic situation is that of planar boundary conditions for the director [$\theta(z = \pm 1/2) = 0$, $\phi(z = \pm 1/2) = 0$]. A similar bifurcation scenario has been observed when a magnetic field $h = 0.5$ was added in the flow plane at an angle $\theta_m = \pi/4$ with respect to the x axis. Then, at low amplitudes of oscillatory Poiseuille flow, one has in-plane director oscillations which do not exceed the $\pm \theta_{fl}$ limit. With increasing flow amplitude a limit cycle corresponding to the slow director oscillations appears as in the previous cases through a homoclinic bifurcation. Further increase of the amplitude a leads to a reduction and disappearance of the limit cycle. The critical flow amplitude a for the limit cycle instability depends on the value of magnetic field h and remains very sensitive to the anisotropy of the elastic constants.

VI. CONCLUSION

For the first time (to our knowledge) a slow time-periodic director motion has been found theoretically. The oscillations

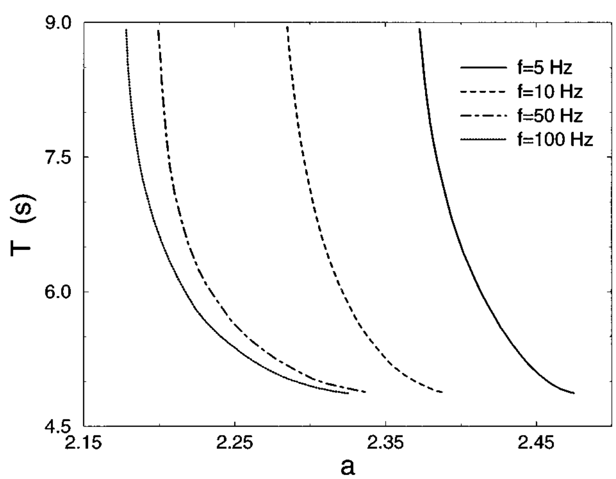

FIG. 5. Dependence of the period of slow-time oscillations T on the amplitude of Poiseuille flow for different frequencies.

appear with increasing amplitude of low-frequency Poiseuille flow through a homoclinic bifurcation and disappear through a Hopf bifurcation. The effect depends strongly on the anisotropy of elastic constants and in fact disappears in the one-constant approximation.

For low-frequency oscillatory Couette flow with its uniform shear rate no homogeneous instabilities are found. Increasing the flow frequency leads to deviations from uniformity and, therefore, the appearance of a time-averaged torque acting on the director [20,21]. This may lead to the experimentally observed slow director oscillations in a way similar to the one found above. Although the resulting rotation of the in-plane director component is confined to at most a half plane (there are two or four symmetry-equivalent states) it can give the impression of full 2π rotations, as reported in [18,19], because the optical detection system involving birefrengence is insensitive to rotations by multiples of $\pi/2$. Work on high-frequency shear flow is in progress.

To test our predictions directly experiments involving plane oscillatory Poiseuille flow are desirable. We suggest doing this with planar director alignment and a magnetic field applied in the shear plane at an angle of about 45° with respect to the x axis.

ACKNOWLEDGMENTS

Financial support by the Deutsche Forschungsgemeinschaft (Sonderforschungsbereich 213, Bayreuth) and Russian Foundation for Basic Research (Grant No. 94-02-05528a) are gratefully acknowledged. A.K. is grateful to the Alexander von Humboldt-Stiftung for financial support and wishes to acknowledge the hospitality of the University of Bayreuth.

APPENDIX A: COEFFICIENTS IN NEMATODYNAMIC EQUATIONS

$$a_1(\theta,\phi)=\cos^2\theta+k_2\sin^2\theta+(k_3-k_2)\sin^2\theta\,\cos^2\phi, \tag{A1}$$

$$a_2(\theta,\phi)=[k_2-1+(k_3-k_2)\cos^2\phi]\sin\theta\,\cos\theta,$$

$$a_3(\theta,\phi)=(k_2-1)\sin\theta\,\cos\theta\,\tan\phi,$$

$$a_4(\theta,\phi)=(2k_2-k_3-1))\sin\theta\,\cos\theta,$$

$$a_5(\theta,\phi)=-2[\cos^2\theta+k_2\sin^2\theta+2(k_3-k_2)\sin^2\theta\,\cos^2\phi]\tan\phi,$$

$$a_6(\theta,\phi,\hat{\mathbf{m}})=\pi^2(\cos\theta\,\cos\phi m_x+\sin\phi m_y+\sin\theta\,\cos\phi m_z)\times(-\sin\theta m_x+\cos\theta m_z)\sec\phi,$$

$$b_1(\theta,\phi)=(k_2-1)\sin\theta\,\cos\theta\,\sin\phi\,\cos\phi,$$

$$b_2(\theta,\phi)=[\sin^2\theta+k_2\cos^2\theta+2(k_3-k_2)\sin^2\theta\,\cos^2\phi]\times\sin\phi\,\cos\phi,$$

$$b_3(\theta,\phi)=\sin^2\theta+k_2\cos^2\theta+(k_3-1)\sin^2\theta\,\cos^2\phi,$$

$$b_4(\theta,\phi)=-(k_3-1)\sin^2\theta\,\sin\phi\,\cos\phi,$$

$$b_5(\theta,\phi)=-2\,\sin\theta\,\cos\theta(k_2-k_3\cos^2\phi-\sin^2\phi),$$

$$b_6(\theta,\phi,\hat{\mathbf{m}})=\pi^2(\cos\theta\,\cos\phi m_x+\sin\phi m_y+\sin\theta\,\cos\phi m_z)\times(-\cos\theta\,\sin\phi m_x+\cos\phi m_y-\sin\theta\,\sin\phi m_z),$$

$$2(-\alpha_2)c_1(\theta,\phi)=\alpha_4+(\alpha_5-\alpha_2)\sin^2\theta\,\cos^2\phi,$$

$$2(-\alpha_2)c_2(\theta,\phi)=\alpha_3+\alpha_6+2\alpha_1\sin^2\theta\,\cos^2\phi,$$

and $k_i=K_{ii}/K_{11}$.

APPENDIX B: MATERIAL PARAMETERS

The numerical computations are carried out for the following MBBA (4-methoxybenzylidene-4′-n-butylaniline) material parameters at 25 °C [24,25].

Viscosity coefficients are in units of 10^{-3} N s/m^2 :

$$\alpha_1=-18.1,\quad \alpha_2=-110.4,\quad \alpha_3=-1.1,\quad \alpha_4=82.6,$$

$$\alpha_5=77.9,\quad \alpha_6=-33.6.$$

Elasticity coefficients are in units of 10^{-12} N :

$$K_{11}=6.66,\quad K_{22}=4.2,\quad K_{33}=8.61,$$

and mass density $\rho=10^3$ kg/m^3. We used the layer thickness $d=20$ μm.

[1] F. M. Leslie, Adv. Liq. Cryst. **4**, 1 (1979).
[2] F. M. Leslie, in *Theory and Applications of Liquid Crystals*, edited by J. L. Ericksen and D. Kinderlehrer (Springer-Verlag, New York, 1987).
[3] J. T. Gleeson, P. Palffy-Muhoray, and W. van Saarloos, Phys. Rev. A **44**, 2588 (1991).
[4] C.-R. Sheu and R.-P. Pan, Mol. Cryst. Liq. Cryst. **265**, 257 (1995).
[5] E. Dubois-Violette and P. Manneville, in *Pattern Formation in Liquid Crystals*, edited by A. Buka and L. Kramer (Springer-Verlag, New York, 1995).
[6] I. Zuniga and F. M. Leslie, Liq. Cryst. **5**, 725 (1989).
[7] I. Zuniga and F. M. Leslie, Europhys. Lett. **9**, 689 (1989).
[8] I. Zuniga and F. M. Leslie, J. Non-Newt. Fluid Mech. **33**, 123 (1989).
[9] M. G. Clark, F. C. Saunders, I. A. Shanks, and F. M. Leslie, Mol. Cryst. Liq. Cryst. **70**, 195 (1981).
[10] F. Scudieri, Appl. Phys. Lett. **29**, 398 (1976).
[11] G. N. Belova and E. N. Remizova, Akust. Zh. **31**, 289 (1985) [Sov. Phys. Acoust. **31**, 171 (1985)].
[12] S. J. Hogan, T. Mullin, and P. Woodford, Proc. R. Soc. London Ser. A **441**, 559 (1993).
[13] E. N. Kozhevnikov, Zh. Eksp. Teor. Fiz. **91**, 1346 (1986) [Sov. Phys. JETP **64**, 793 (1986)].

[14] P. Pieranski and E. Guyon, Phys. Rev. Lett. **39**, 1281 (1977).
[15] E. Guazzelli and E. Guyon, J. Phys. (Paris) **43**, 985 (1982).
[16] E. Dubois-Violette and F. Rothen, J. Phys. (Paris) **39**, 1039 (1978).
[17] J. Sadik, F. Rothen, and W. Bestgen, J. Phys. (Paris) **42**, 915 (1981).
[18] A. N. Chuvyrov, Zh. Eksp. Teor. Fiz. **82**, 761 (1982) [Sov. Phys. JETP **55**, 451 (1982)].
[19] A. N. Chuvyrov, O. A. Scaldin, and V. A. Delev, Mol. Cryst. Liq. Cryst. **215**, 187 (1992).
[20] A. P. Krekhov, L. Kramer, A. Buka, and A. N. Chuvyrov, J. Phys. II (France) **3**, 1387 (1993).
[21] A. P. Krekhov and L. Kramer, J. Phys. II (France) **4**, 677 (1994).
[22] P. G. de Gennes and J. Prost, *The Physics of Liquid Crystals* (Clarendon Press, Oxford, 1993).
[23] O. Parodi, J. Phys. (Paris) **31**, 581 (1970).
[24] W. H. de Jeu, W. A. P. Classen, and A. M. J. Spruijt, Mol. Cryst. Liq. Cryst. **37**, 269 (1976).
[25] H. Kneppe, F. Schneider, and N. K. Sharma, J. Chem. Phys. **77**, 3203 (1982).