

## Convective instabilities in cholesteric and smectic-*A* liquid crystals

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The convective instabilities occurring in cholesteric and smectic-*A* liquid crystals are investigated by linear stability analysis. We find oscillatory instabilities not predicted previously and we present a thorough examination of the stationary instabilities which have been discussed previously in a simplified version by Dubois-Violette and Parsons. The differences in the convective instabilities in cholesterics and smectics *A* due to the differences in the underlying hydrodynamic equations and to the different orders of magnitude of the corresponding transport coefficients and static susceptibilities are considered in some detail. We discuss the mechanisms of the instabilities which are different from those of nematics and we obtain the threshold conditions for cholesterics and smectics *A* for the critical temperature difference and for the frequency by energy-balance considerations. The influence of a temperature-dependent pitch is investigated in a separate section.

### I. INTRODUCTION

The study of convective instabilities, generated by external temperature gradients in liquid crystals is interesting both theoretically and experimentally. First convective instabilities are "non-equilibrium phase transitions" from a state of higher to a state of lower symmetry. Since liquid crystals already have a reduced symmetry compared to simple fluids a further reduction of the symmetry by convective structures has to be expected and will lead to new effects. Secondly the experimental investigation of thermal instabilities in liquid crystals can yield information on various hydrodynamic parameters like viscosities and static susceptibilities. Although the experimental<sup>1-11</sup> and theoretical<sup>9,12-17</sup> effort during the last years was mainly addressed to the study of hydrodynamic instabilities in nematics, the treatment of convective instabilities<sup>18-22</sup> in cholesterics and smectics *A* comes now into experimental reach.

In nematics the existence of a stationary and of an oscillatory convective instability is established both theoretically<sup>14,15</sup> and experimentally.<sup>9,11</sup> In this paper we report theoretically on two analogous instabilities in smectics *A* and cholesterics. Our results contain new predictions, e.g., an intrinsically oscillatory instability, and cover and amend previously given discussions on the stationary instability.<sup>18,19</sup> In addition, we investigate the different behavior of smectics *A* and cholesterics, which is based on different orders of magnitude of some relevant phenomenological parameters as well as on differences in the basic hydrodynamic equations. Although both types of instabilities are analogous

to those in nematics, their main features and the underlying mechanisms are quite different from those in nematics, because of the existence of an elastic energy in smectics *A* and cholesterics, not present in nematics.

We think of horizontal layers ( $z = \text{const}$ ) of smectic-*A* and cholesteric liquid crystals between an upper ( $z = d$ ) and a lower ( $z = 0$ ) boundary. The direction normal to the layers is denoted by  $\hat{p}$ . For smectics *A*,  $\hat{p}$  is identical with the averaged molecule axis  $\hat{n}$ , while for cholesterics,  $\hat{p}$  is the pitch axis of the helical geometry. A temperature gradient is applied across the layers. For a certain threshold value of this temperature gradient a convective motion sets in.

In Sec. II the basic nonlinear hydrodynamic equations for smectics *A* and for cholesterics are introduced and the differences between them are discussed. In Sec. III we solve these equations for the steady heat-conduction state. A linear-stability analysis of this heat-conduction state is performed in Sec. IV under the approximation that the direct temperature dependence of the layer fluctuations can be neglected ("temperature-independent pitch" assumption). The threshold condition for both, the stationary and oscillatory instability, are then given in Sec. V A for smectics *A*; for cholesterics this Sec. V is only valid as a first approximation. In Secs. V B and V C we discuss the mechanisms leading to the instabilities and regain the threshold conditions by energy-balance considerations. A discussion of numerical values and a comparison between the two types of instabilities is given in Sec. V D. The additional new features due to the correct hydrodynamic equations for cholesterics are discussed in Sec. VI. In Sec. VII we lift the assumption of a temperature-independent pitch and discuss

this general case for both types of instabilities for smectics  $A$  and for cholesterics. A summary of our results is contained in Sec. VIII. In two Appendices we derive the influence of a fluctuating

preferred direction  $\hat{p}$  (Appendix A) and discuss the inclusion of a destabilizing magnetic field in the case of smectics  $A$  (Appendix B). In the main text we always allow for a stabilizing magnetic field.

## II. HYDRODYNAMIC EQUATIONS

For the investigation of convective instabilities in smectics  $A$  and cholesterics we use nonlinear hydrodynamic equations whose nonlinear reversible parts were recently derived by the authors<sup>23</sup> and whose nonlinear irreversible parts are a straightforward generalization of previous linear ones.<sup>24-28</sup> For the momentum density  $\vec{g}$ , the density  $\rho$ , the entropy density  $\sigma$ , and the order parameter (layer displacement along  $\hat{p}$ )  $R$ , we have the equations of motion

$$\begin{aligned} \frac{\partial}{\partial t} R + v_k \nabla_k R - \hat{p}_k v_k - \frac{A}{2q_0} \hat{p}_k (\vec{\nabla} \times \vec{v})_k + f_{kj} R \nabla_k v_j + g_{kj} q_0 \nabla_k v_j &= \zeta \nabla_j \phi_j + \xi \hat{p}_j \nabla_j T, \\ \frac{\partial}{\partial t} \rho + \nabla_j \rho v_j &= 0, \\ \frac{\partial}{\partial t} \sigma + \nabla_j \sigma v_j &= \hat{p}_j \nabla_j (\xi \nabla_m \phi_m) + \nabla_j (\kappa_{ij} C_p T_0^{-1} \nabla_i T), \\ \frac{\partial}{\partial t} g_i + \nabla_j g_i v_j + \nabla_i p + \rho g_E \delta_{i\alpha} - \hat{p}_i \nabla_j \phi_j + \nabla_j (\phi_j \nabla_i R) - \frac{A}{2q_0} \hat{p}_k \epsilon_{ijk} \nabla_j \nabla_m \phi_m - \nabla_j (f_{ji} R \nabla_m \phi_m + g_{ji} q_0 \nabla_m \phi_m) &= \nabla_j (v_{ijkl} \nabla_i v_k). \end{aligned} \quad (2.1)$$

Here  $p$  denotes the pressure,  $g_E$  the acceleration due to gravity,  $q_0$  the wave vector associated with the equilibrium pitch of the helices in cholesterics,  $\vec{v}$  the velocity, and  $T$  the temperature. The tensors  $f_{ij}$ ,  $g_{ij}$ ,  $\kappa_{ij}$  contained in (2.1) are of the axial form  $t_{ij} = t_{\parallel} \hat{p}_i \hat{p}_j + t_{\perp} (\delta_{ij} - \hat{p}_i \hat{p}_j)$ ; their anisotropic parts are defined by  $t_{\parallel} \equiv t_{\parallel} - t_{\perp}$ ; for the viscosity tensor  $v_{ijkl}$  cf. Forster<sup>29,30</sup>. The dissipative transport parameters are  $\zeta$  (order-parameter friction),  $\xi$ ,  $\kappa_{\parallel}$ ,  $\kappa_{\perp}$  (heat conduction), and five  $v_i$  (viscosity). For  $\xi$  there is the constraint  $\xi^2 < \zeta \kappa_{\parallel} C_p T_0^{-1}$  due to the positivity of entropy production. The reactive ones are  $f_{\parallel}$ ,  $f_{\perp}$ ,  $g_{\parallel}$ ,  $g_{\perp}$ , and  $A$ .

The hydrodynamics of smectics  $A$  and cholesterics differ from each other with respect to terms containing  $q_0$  (which is not present in smectics  $A$ ), i.e.,  $A = 0 = g_{\perp} = g_{\parallel}$  for smectics  $A$ . For cholesterics the  $A$  terms describe the fact that a rotation of the helices about  $\hat{p}$  is equivalent to a translation of the layers along  $\hat{p}$ , therefore  $A = 1$ , which was already obtained by Lubensky.<sup>25</sup> The  $g$  terms have no simple geometric explanation and  $g_{\perp}$ ,  $g_{\parallel}$  are phenomenological parameters. This term is quite analogous to the  $\lambda$  term in the director equation for nematics.<sup>29,30</sup>

The equations of motion are completed by equations of state. As independent variables we use  $p$  and  $T$ . The equations of state read (cf. Ref. 23)

$$\begin{aligned} v_i &= \frac{1}{\rho} g_i \\ \delta \rho &= \chi_E \delta p - \alpha \delta T, \\ \delta \sigma &= C_p T_0^{-1} \delta T - C_v \gamma_2 T_0^{-1} \hat{p}_j \nabla_j R, \\ \phi_j &= \hat{p}_j \hat{p}_k (\chi_{\parallel} - C_v T_0^{-1} \gamma_2) \nabla_k R + \gamma_2 C_p T_0^{-1} \hat{p}_j \delta T \\ &\quad \pm \chi_a H^2 (\delta_{jk} - \hat{p}_j \hat{p}_k) \nabla_k R, \end{aligned} \quad (2.2)$$

where we have neglected the pressure dependence of  $\delta \sigma$  and  $\phi_j$  ( $\gamma_1 \equiv 0$ ) and nonlinearities in  $\nabla R$ , which will play no role in the following. The isothermal compressibility  $\chi_E$  and the thermal expansion coefficient  $\alpha$  are related to the susceptibilities introduced in Ref. 23 by  $\alpha = \sigma_0 / \rho_0 \lambda$  and  $\chi_E = 1 / \rho_0 \lambda$  with  $\lambda = (\partial^2 \epsilon / \partial \rho^2)_{\sigma}$ . The susceptibility  $\gamma_2$  is defined by  $\gamma_2 \equiv (\partial^2 \epsilon / \partial \sigma \partial \hat{p}_i \nabla_i R)_{\rho}$  and  $\chi_{\parallel} \equiv [(\partial^2 \epsilon / (\partial \hat{p}_i \nabla_i R)^2)]_{\sigma, \rho}$ . Thermodynamic stability requires  $\chi_{\parallel} > C_v T_0^{-1} \gamma_2^2$ . The last term in (2.2) describes the influence of an external magnetic field  $\vec{H}$  applied parallel to  $\hat{p}$ . The upper (lower) sign refers to smectics  $A$  (cholesterics). The magnetic field is stabilizing for  $\chi_a > 0$  in smectics  $A$  and for  $\chi_a < 0$  in cholesterics. The influence of a magnetic field perpendicular to  $\hat{p}$  is discussed in Appendix B. Equations (2.1) and (2.2) are only valid for  $H \neq 0$ ; for  $H = 0$  the transverse fluctuations of the preferred axis are to be considered. This is done in Appendix A. In cholesterics, saturation effects are important for very strong magnetic fields ( $H$

$> 20$  (kG) and  $\chi_a H^2$  in (2.2) has to be replaced<sup>26</sup> by  $\chi_a H^2 [1 + \chi_a H^2 q_0^{-2} (K_1 + K_3)^{-1}]^{-1}$ . In smectics *A* such saturation effects are negligible.

### III. HEAT-CONDUCTION REGIME

If a constant temperature gradient across the layer is applied to a simple fluid, there exists a steady state without convection, where heat conduction is the only dissipative process. This heat-conduction regime is stable up to a critical temperature gradient. In smectics *A* and cholesterics an analogous regime exists, whose stability will be analyzed in the following sections. However, this steady state contains not only heat conduction but also order-parameter friction as a dissipative process. The latter is induced by the dynamical coupling of  $T$  with  $\hat{p}_i \nabla_i R$  characterized by the dissipative transport parameter  $\xi$ . Explicitly, by solving Eqs. (2.1) and (2.2) for  $\partial/\partial t = 0$  and  $\vec{v} = 0$  and applied thermal gradient we obtain for the new heat-conduction regime

$$\begin{aligned} \vec{g}^{\text{HC}} &= 0, \\ T^{\text{HC}}(z) &= T_0 + \theta \frac{z}{d}, \\ R^{\text{HC}}(z) &= -\frac{1}{2} z^2 \frac{\theta}{d} \eta \bar{\chi}_{\parallel}^{-1}, \\ p^{\text{HC}}(z) &\approx p_0 (1 - \chi_E g_E z) - z \left( \rho_0 g_E + \xi \zeta^{-1} \frac{\theta}{d} \right) \left( 1 - \frac{1}{2} z g_E \chi_E \right) \\ &\quad + \frac{1}{2} z^2 \left( \alpha g_E \frac{\theta}{d} + \frac{\theta^2}{d^2} \xi \zeta^{-1} \eta \bar{\chi}_{\parallel}^{-1} (f_{\parallel} - 2) \right), \end{aligned} \quad (3.1)$$

and for the thermodynamic conjugates

$$\begin{aligned} \rho^{\text{HC}}(z) &= \rho_0 - \alpha z \frac{\theta}{d} + (p^{\text{HC}} - p_0) \chi_E, \\ \sigma^{\text{HC}}(z) &= \sigma_0 + C_p T_0^{-1} z \frac{\theta}{d} + C_v \gamma_2 T_0^{-1} z \frac{\theta}{d} \eta \bar{\chi}_{\parallel}^{-1}, \\ \phi_z^{\text{HC}}(z) &= -\xi \zeta^{-1} z \frac{\theta}{d}, \end{aligned} \quad (3.2)$$

with the abbreviations

$$\eta = \xi \zeta^{-1} + \gamma_2 C_p T_0^{-1} \quad \text{and} \quad \bar{\chi}_{\parallel} = \chi_{\parallel} - C_v T_0^{-1} \gamma_2^2.$$

The expression for  $p^{\text{HC}}(z)$  is an expansion in  $g_E \chi_E z$  up to linear order, since  $g_E \chi_E z \ll 1$  for  $z \leq d_0 \approx 1$  cm. Note, that  $\xi$ ,  $\gamma_2$ , and therefore  $\eta$  can have either sign, while  $\bar{\chi}_{\parallel}$  is positive. There is no difference between smectics *A* and cholesterics in this regime. The result  $R^{\text{HC}} \sim z^2 \theta$  means, that the layer distances (or pitch) are no longer constant as in the true equilibrium state, but depend on  $z$  and on the external temperature gradient. This is caused by a sum of two effects, a static and a dynamical coupling of  $T$  and  $R$ . The dynamical coupling, characterized by  $\xi$  was al-

ready mentioned above. The static coupling is described by the susceptibility  $\gamma_2$ . Parts of this static coupling were already discussed by Parsons.<sup>19</sup> However, this temperature dependence of the layer distances (pitch) is assumed to be very weak. Therefore we will neglect this temperature dependence in the next three sections; its influence on the stability of the heat-conduction regime will be discussed in Sec. VII. By neglecting the temperature dependence of  $R$  (i.e.,  $\xi = 0 = \gamma_2$ ) the expressions (3.1) and (3.2) of the heat-conduction regime of smectics *A* and cholesterics are identical to that of simple fluids (and  $R = 0$ ). In the following we analyze the stability of the heat-conduction regime within linear-stability analysis.

### IV. LINEARIZED EQUATIONS

In this section we analyze the stability of the heat-conduction states (3.1) and (3.2) with temperature-independent layer distances ( $\xi = 0 = \gamma_2$ ) with respect to infinitesimal disturbances. Small deviations  $T(\vec{r}, t) - T^{\text{HC}} \equiv T'(\vec{r}, t)$ ,  $p(\vec{r}, t) - p^{\text{HC}} \equiv p'(\vec{r}, t)$ ,  $R(\vec{r}, t)$ , and  $\vec{g}(\vec{r}, t)$  from the heat-conduction state evolve in time according to the hydrodynamic equations (2.1) and (2.2).

We linearize these equations for  $T'(\vec{r}, t)$ ,  $p'(\vec{r}, t)$ ,  $R(\vec{r}, t)$ , and  $\vec{g}(\vec{r}, t)$  and simplify it by the Boussinesq approximation, which contains in our case the same assumptions as in the case of simple liquids, namely,

$$\begin{aligned} \text{(i)} \quad & \text{div } \vec{v} = 0, \\ \text{(ii)} \quad & \chi_E \delta p \ll \alpha \delta T, \\ \text{(iii)} \quad & \rho^{\text{HC}}(z) \approx \rho_0, \text{ except for the buoyancy} \end{aligned} \quad (4.1)$$

force, (iv) susceptibilities and transport parameters = const. These approximations are justified for the same reasons as in simple liquids. However, in order to meet the last condition of (4.1) it is necessary to work sufficiently far away from phase transitions.

We are now left with a set of differential equations<sup>21</sup> for  $T'$ ,  $p'$ ,  $R$ , and  $u, v, w$  (the Cartesian components of  $\vec{v}$ ). Because of the uniaxial symmetry of our problem we can assume  $\partial/\partial y = 0$ , without loss of generality. The heat-conduction state is stable as long as the disturbances are decaying, i.e., for  $R, T, \vec{v} \sim \exp(i\omega t - \sigma t)$  the condition  $\sigma = 0$  denotes the onset of instability ( $\omega$  real). Therefore we are looking for solutions with the time dependence  $\sim e^{i\omega t}$ . With respect to the space dependence we Fourier transform the set of differential equations. Boundary conditions are given below. To this end we obtain a homogeneous system of algebraic equations (dropping the prime

on  $T'$  and  $p'$ ):

$$\begin{aligned}
 i\omega R - w + \zeta \hat{\chi}^2 R &= A(2q_0)^{-1} i k_{\perp} v - q_0 g_a i k_{\parallel} w, \\
 i\omega v + \hat{\mu}_y^2 v &= A(2q_0 \rho_0)^{-1} i k_{\perp} \hat{\chi}^2 R, \\
 i\omega w + i k_{\parallel} \rho_0^{-1} p - \alpha g_E \rho_0^{-1} T + \rho_0^{-1} \hat{\chi}^2 R + \hat{\mu}_s^2 w & \quad (4.2) \\
 &= q_0 \rho_0^{-1} g_{\parallel} i k_{\parallel} \hat{\chi}^2 R, \\
 -p k^2 - i k_{\parallel} \alpha g_E T + i k_{\parallel} \hat{\chi}^2 R + i k_{\parallel} \hat{\mu}_s^2 w &= q_0 \hat{g}^2 \hat{\chi}^2 R, \\
 i\omega T + \frac{\theta}{d} w + \hat{\kappa}^2 T - \frac{\theta}{d} \kappa_a k_{\perp}^2 R &= 0,
 \end{aligned}$$

with

$$\hat{\chi}^2 = \chi_{\parallel} k_{\parallel}^2 \pm \chi_a H^2 k_{\perp}^2 + \bar{K} k_{\perp}^4,$$

$$\rho_0 \hat{\mu}_y^2 = \nu_3 k_{\parallel}^2 + \nu_2 k_{\perp}^2,$$

$$\rho_0 \hat{\mu}_s^2 = (2\nu_1 - \nu_3) k_{\parallel}^2 + \nu_3 k_{\perp}^2,$$

$$\rho_0 \hat{\mu}_s^2 = 2(\nu_1 - \nu_3) k_{\parallel}^2 + 2(\nu_3 - \nu_2) k_{\perp}^2,$$

$$\hat{g}^2 = g_{\parallel} k_{\parallel}^2 + g_{\perp} k_{\perp}^2,$$

$$\hat{\kappa}^2 = \kappa_{\parallel} k_{\parallel}^2 + \kappa_{\perp} k_{\perp}^2,$$

where  $k_{\parallel}, k_{\perp}$  are the components of the wave vector parallel and perpendicular to  $\hat{p}$ , respectively. In the expression for  $\hat{\chi}^2$  we have inserted the contribution  $\bar{K} k_{\perp}^4$  due to the transverse fluctuations of the preferred axis. In Appendix A the derivation of this term is given. It is important for the case  $H \rightarrow 0$ . The  $\pm \chi_a H^2 k_{\perp}^2$  term is positive for a stabilizing field ( $\chi_a > 0$  for smectics,  $\chi_a < 0$  for cholesterics, usually). The solutions of Eqs. (4.2) will be discussed in the following two sections.

## V. INSTABILITIES IN SMECTICS A (FIRST APPROXIMATION FOR CHOLESTERIC)

### A. Threshold conditions

In this section we will put  $A = 0 = g_{\parallel} = g_{\perp}$ , i.e., the right-hand side of Eqs. (4.2) equal to zero. The equations are then suitable for smectics A. In Sec. VI the actual differences between smectics A and cholesterics which show up in the parameters  $A, g_{\parallel}, g_{\perp}$ , are considered. The following discussion can, therefore, be applied to cholesterics only as a first approximation.

Equations (4.2) constitute an eigenvalue problem, whose associate linear operator is non-Hermitian. Thus the eigenvalue  $i\omega$  may become imaginary and we have generally to expect solutions with  $\omega \neq 0$ . In fact, there are two solutions of (4.2), one with  $\omega = 0$ , denoting the onset of a stationary convective instability (STI) and one with  $\omega \neq 0$  denoting the onset of an oscillatory convective instability

(OSI). Explicitly we find<sup>21, 22</sup>

$$\omega = 0,$$

at

$$\theta^{\text{STI}}(k) = \frac{\rho_0 d}{\alpha g_E} \frac{\hat{\chi}^2 \hat{\kappa}^2}{k_{\perp}^2} \frac{\zeta \hat{\mu}^4 + k_{\perp}^2 \rho_0^{-1}}{\kappa_a k_{\perp}^2 - \zeta \hat{\chi}^2}, \quad (5.1)$$

and

$$\begin{aligned}
 \omega^2(k) &= (\kappa_{\parallel} k_{\parallel}^2 + \hat{\mu}^4)^{-1} [\zeta \hat{\chi}^2 (\hat{\mu}^4 + \hat{\kappa}^2 k^2) (\kappa_a k_{\perp}^2 - \zeta \hat{\chi}^2) \\
 &\quad + \kappa_a k_{\perp}^2 \hat{\mu}^4 \hat{\kappa}^2 + (\kappa_{\parallel} k_{\parallel}^2 - \zeta \hat{\chi}^2) k_{\perp}^2 \hat{\chi}^2 \rho_0^{-1}] \quad (5.2)
 \end{aligned}$$

at

$$\begin{aligned}
 \theta^{\text{OSI}}(k) &= - \frac{\rho_0 d}{\alpha g_E} \frac{\mu^4 + \zeta \chi^2 k^2}{\hat{\mu}^4 + \kappa_{\parallel} k_{\parallel}^4} \\
 &\quad \times [k_{\perp}^{-2} (\hat{\mu}^4 + \hat{\kappa}^2 k^2) (\zeta \hat{\chi}^2 + \hat{\kappa}^2) + \rho_0^{-1} \hat{\chi}^2]
 \end{aligned}$$

with

$$\hat{\mu}^4 \rho_0 = \nu_3 (k_{\parallel}^2 - k_{\perp}^2)^2 + 2(\nu_1 + \nu_2) k_{\parallel}^2 k_{\perp}^2,$$

$$\hat{\chi}^2 = \chi_{\parallel} k_{\parallel}^2 \pm \chi_a H^2 k_{\perp}^2 + \bar{K} k_{\perp}^4,$$

$$\hat{\kappa}^2 = \kappa_{\parallel} k_{\parallel}^2 + \kappa_{\perp} k_{\perp}^2.$$

These formulas can be simplified by the assumption  $\hat{\mu}^4 k^{-2} \gg \hat{\kappa}^2 \gg \zeta \hat{\chi}^2$ , which means that relaxation due to viscosity is much faster than thermal conductivity, which is, on the other hand, faster than order-parameter relaxation. These inequalities are fulfilled in most smectics A and cholesterics. We then find

$$\omega = 0,$$

$$\theta^{\text{STI}}(k) = \frac{d}{\alpha g_E} \frac{\hat{\chi}^2 \hat{\kappa}^2}{\kappa_a k_{\perp}^2 - \zeta \hat{\chi}^2}, \quad (5.3)$$

and

$$\omega^2(k) = \kappa_a k_{\perp}^2 k^2 + \frac{\kappa_{\parallel} k_{\parallel}^2 \hat{\chi}^2 \rho_0^{-1}}{\hat{\mu}^4 k^{-2}}, \quad (5.4)$$

$$\theta^{\text{OSI}}(k) = - \frac{\rho_0 d}{\alpha g_E} \left( \frac{\hat{\mu}^4 \hat{\kappa}^2}{k_{\perp}^2} + \rho_0^{-1} \hat{\chi}^2 \right).$$

In order to obtain the true threshold values  $\omega_c = \omega(k_c)$  and  $\theta_c = \theta(k_c)$  we have to specify the critical wave vector  $k_c$ . At the top and bottom of the layer we take, for the flow, so called free-free boundary conditions for simplicity, and for  $T, R$  "natural" boundary conditions, i.e.,

$$T = R = w = \frac{\partial^2 w}{\partial z^2} = \frac{\partial u}{\partial z} = \frac{\partial v}{\partial z} = 0 \quad (5.5)$$

at  $z = 0, d$ . In that case,  $k_{\parallel}^c = \pi/d$  is immediately found, as in simple fluids. For more realistic boundary conditions  $k_{\parallel}^c$  can be obtained numer-

ically. The principal features of the instability are not affected by our choice of boundary conditions. For the horizontal planes we do not impose special boundary conditions. Thus,  $k_1^c$  is determined by minimizing  $\theta(k)$  with respect to  $k_1$  (or  $k_1^2$ ). For the simplified formulas (5.3) and (5.4) we obtain  $k_1^c = \sqrt{B} \pi/d$  with (STI)

$$2B^3 + \left( \frac{\kappa_{\parallel}}{\kappa_{\perp}} \pm \frac{\chi_a H^2 d^2}{\bar{K} \pi^2} \right) B^2 + \frac{\chi_a d^2}{\bar{K} \pi^2} \frac{\xi \chi_{\parallel} + \kappa_a \kappa_{\parallel} / \kappa_{\perp}}{\pm \xi \chi_a H^2 - \kappa_a} = 0 \quad (5.6)$$

and (OSI)

$$2 \left( 1 + \frac{\bar{K}}{\nu_3 \kappa_{\perp}} \right) B^3 + \left( \frac{\kappa_{\parallel}}{\kappa_{\perp}} + 2 \frac{\nu_1 + \nu_2 - \nu_3}{\nu_3} \pm \frac{d^2}{\pi^2} \frac{\chi_a H^2}{\nu_3 \kappa_{\perp}} \right) B^2 - \frac{\kappa_{\parallel}}{\kappa_{\perp}} = 0,$$

respectively. For the exact formulas (5.1) and (5.2) one gets an algebraic equation of fifth and seventh order for  $B$ , which has to be solved numerically. In the following subsections we will discuss the mechanism leading to these instabilities.

#### B. Stationary convective instability

This instability has already been predicted by Dubois-Violette<sup>18</sup> and Parsons.<sup>19</sup> Although derived for cholesterics, their results agree with ours for smectics A. For cholesterics they are valid only approximately (cf. Sec. VI). Since Dubois-Violette<sup>18</sup> and Parsons<sup>19</sup> have discussed this instability extensively, we will only give a short comprehension of the main features in order to be able to compare afterwards with OSI.

As can easily be seen from the denominator of  $\theta_c$  in (5.1) there are two mechanism involved which act together for  $\kappa_a < 0$  and against each other for  $\kappa_a > 0$ . The first ( $\zeta = 0$ ,  $\kappa_a \neq 0$ ) is based on a coupling of  $R$  with  $T$  because of the stationary balance of heat conduction with heat focusing due to the anisotropy of  $\kappa_{ij}$ , i.e.,  $\hat{\kappa}^2 T = (\theta_c/d) \kappa_a k_1^2 R$ . The back coupling is achieved since the buoyancy force is balanced by the elastic force due to the layer structure, i.e.,  $\alpha_{g_E} T = \hat{\chi}^2 R$ . This closed circuit amplifies itself and becomes unstable for  $\theta_c \kappa_a > 0$ . There is no convection involved ( $w = 0$ ). This mechanism is sketched in Fig. 1, shown by the dotted lines.

The second mechanism ( $\zeta \neq 0$ ,  $\kappa_a = 0$ ) consists of a coupling between  $w$  and  $T$ :  $\hat{\kappa}^2 T = -(\theta_c/d) w$  (heat conduction = convective heat transport), between  $T$  and  $R$ :  $\alpha_{g_E} T = \hat{\chi}^2 R$  [buoyancy force = elastic force neglecting a viscous force as in (5.3)] and between  $R$  and  $w$ :  $w = \hat{\chi}^2 R$  (permeation effect). This reaction cycle gets unstable for  $\theta_c < 0$ . This mechanism is sketched in Fig. 1 shown by the full lines. Of course, in nature both effects are

not separable and the instability sets in for  $\theta_c (\kappa_a k_1^2 - \zeta \hat{\chi}^2) > 0$ , i.e., if  $(\kappa_a k_1^2 - \zeta \hat{\chi}^2) > 0$  for  $\theta_c > 0$  (by heating from above) and if  $(\kappa_a k_1^2 - \zeta \hat{\chi}^2) < 0$  for  $\theta_c < 0$  (by heating from below). We will give numerical values for  $k_1^c$ ,  $\theta_c^{\text{STI}}$  and a discussion of their  $\kappa_a$  and  $H$  dependence in connection and comparison with the OSI in subsection V D.

The eigenfunctions belonging to the eigenvalue  $\omega = 0$  and according to our boundary conditions (5.5) are

$$\begin{aligned} w &= R_0 \zeta \hat{\chi}^2 \sin k_{\parallel}^c z \cos k_{\perp}^c x, \\ v &= 0, \\ u &= -R_0 \zeta \hat{\chi}^2 k_{\parallel}^c (k_{\perp}^c)^{-1} \cos k_{\parallel}^c z \sin k_{\perp}^c x, \\ R &= R_0 \sin k_{\parallel}^c z \cos k_{\perp}^c x, \\ T &= R_0 \frac{\theta_c}{d \hat{\kappa}^2} (\kappa_a k_1^2 - \zeta \hat{\chi}^2) \sin k_{\parallel}^c z \cos k_{\perp}^c x, \end{aligned} \quad (5.7)$$

where  $R_0$  is the constant amplitude. Equations (5.7) describe two-dimensional convection rolls accompanied by temperature and layer-structure undulations. The velocities are small ( $\sim \zeta \hat{\chi}^2$ ) and  $w \neq 0$  is solely due to the permeation effect. Therefore it was argued<sup>18</sup> that the above described instability is hardly observable. However, it should be stressed here that the linear-stability analysis can only describe the onset of the instability, but is not able to predict what happens above the threshold. Especially, Eqs. (5.7) only describe against what kind of fluctuations the heat-conduction state becomes unstable at  $\theta_c$ , but do not necessarily yield the spatial pattern above. By the quite stiff layer structure of smectics A, it is possible that above  $\theta_c$  layer textures quite dif-

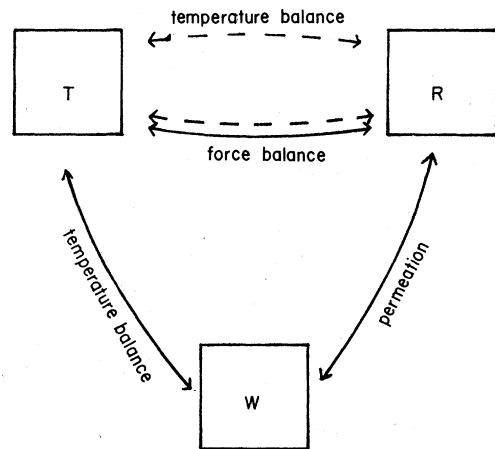


FIG. 1. Mechanisms leading to the stationary instability. The dotted (full) lines refer to couplings obtained for  $\zeta = 0$  ( $\kappa_a \neq 0$ ) [cf. the left-hand side of Eqs. (4.2) with  $\omega = 0$ ].

ferent from Fig. 2, may occur.

We may now look at the instability from a different point of view. The onset of a convective instability sets in when, for a minimum external thermodynamic force, the rate of energy fed into the system equals the rate of energy dissipated by the system.<sup>31</sup> The rate of energy transferred from the gravitational field to the fluid due to the buoyancy force is

$$\begin{aligned}\dot{E}_{in} &= -g_E \int d^3\vec{r} \delta\rho(\vec{r})w(\vec{r}) \\ &= \alpha g_E \int d^3\vec{r} T(\vec{r})w(\vec{r}).\end{aligned}$$

Kinetic energy is dissipated at the rate

$$\dot{E}_{kin}^{dis} = \int d^3\vec{r} \vec{v} \cdot \dot{\vec{g}}^{dis} = -\rho_0 \int d^3\vec{r} v_{ijk} [\nabla_j v_i(\vec{r})][\nabla_i v_k(\vec{r})]$$

and elastic energy at a rate

$$\begin{aligned}\dot{E}_{e1}^{dis} &= \int d^3\vec{r} \phi_i \nabla_i \dot{R}^{dis} \\ &= -\zeta \int d^3\vec{r} [(\chi_{11} \nabla_{11}^2 \pm \chi_a H^2 \nabla_1^2 - \bar{K} \nabla_1^4)R(\vec{r})]^2.\end{aligned}$$

Inserting the eigenfunction (5.7) one obtains ( $\epsilon = E/\nu$ )

$$\begin{aligned}\dot{\epsilon}_{in} &= \frac{\alpha g_E}{4\pi^2} T(k)w(k) = \frac{R_0^2}{4\pi^2} \alpha g_E \zeta \hat{\chi}^2 \frac{\theta_c \kappa_a k_c^2 - \zeta \hat{\chi}^2}{\hat{\kappa}^2}, \\ \dot{\epsilon}_{kin}^{dis} &= -\frac{\rho_0}{4\pi^2} \frac{\hat{\mu}^4}{k_1^2} w^2(k) = -\frac{R_0^2}{4\pi^2} \frac{\rho_0 \hat{\mu}^4}{k_1^2} (\zeta \hat{\chi}^2)^2, \\ \dot{\epsilon}_{e1}^{dis} &= -\frac{1}{4\pi^2} \zeta [\hat{\chi}^2 R(k)]^2 = -\frac{R_0^2}{4\pi^2} \zeta (\hat{\chi}^2)^2.\end{aligned}\quad (5.8)$$

The result (5.1) is regained by the threshold condition  $\dot{\epsilon}_{in} + \dot{\epsilon}_{kin}^{dis} + \dot{\epsilon}_{e1}^{dis} = 0$ . The approximation contained in (5.3) consists, therefore, in neglecting  $\dot{\epsilon}_{kin}^{dis}$  with respect to  $\dot{\epsilon}_{e1}^{dis}$ .

### C. Oscillatory convective instability

The mechanism leading to the instability described in the preceding section is not present in simple fluids, since  $\kappa_a$  and  $\hat{\chi}^2$  are zero there. In simple fluids the convective instability (Bénard

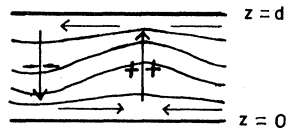


FIG. 2. The spatial pattern of the stationary instability at the critical point;  $\rightarrow$  denotes convective velocities, + and - denote hotter and colder regions, respectively, and  $\sim$  denotes the layer structure.

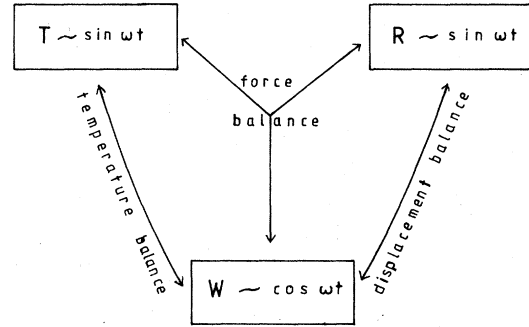


FIG. 3. Simplified mechanism leading to the oscillatory instability under the assumptions mentioned in the main text.

instability) is the result of a coupling between  $T$  and  $w$ , which leads at the threshold to the balance of heat conduction with convective heat transport and of the buoyancy force with the viscous force. Of course, an analogous mechanism is present in smectics  $A$ . However, there are additional couplings between  $T$  and  $R$  and  $w$  and  $R$  [cf. (4.2)], which cause the overstability of this instability. In fact, for  $\kappa_a = 0$  and  $\hat{\chi}^2 = 0$ , Eq. (5.2) reduced to  $\omega = 0$  and  $\theta^{OSI} = \theta^{Ben}$ , the usual threshold condition for the Bénard instability in simple fluids. In order to study the mechanism, we will simplify the problem for the moment by the assumption  $\hat{\mu}^4 k^{-2} \gg \hat{\kappa}^2 \gg \zeta \hat{\chi}^2$  already mentioned above. In addition, there is  $(\hat{\chi}^2 \rho_0^{-1})^{1/2} \gg \hat{\kappa}^2 \gg \zeta \hat{\chi}^2$  by the stiffness of the layer structure (cf. Sec. VD). Therefore, both  $\omega^2$  and  $\theta^{OSI}$  are dominated by the  $\hat{\chi}^2 \rho_0^{-1}$  terms and  $\omega_c \gg \hat{\kappa}^2$ . Therefore, the mechanism described here, is quite different from that leading to the oscillatory instability in nematics [where  $\omega_c \ll \hat{\kappa}^2$  (Refs. 14 and 15)].

Under the above assumptions a fluctuating velocity  $w = w_0 \cos \omega t$  generates [cf. Eq. (4.2)] fluctuations  $R = (w_0/\omega) \sin \omega t$  (i.e., no permeation, since  $\dot{R} = w$ ) and  $T = -(w_0/\omega)(\theta/d) \sin \omega t$  (by the convective heat transportation), cf. Fig. 3. However, by the elastic and buoyancy forces these fluctuations produce a velocity fluctuation  $w = (w_0/\omega^2)(\alpha g_E d^{-1} \rho_0^{-1} \theta + \rho_0^{-1} \hat{\chi}^2) \cos \omega t$ . The system becomes unstable ( $\omega$  imaginary), if  $\alpha g_E \theta/d \rho_0 < -\rho_0^{-1} \hat{\chi}^2$ . Thus,  $\theta_c = -d \hat{\chi}^2 / \alpha g_E$  which is equal to (5.4) under the approximations currently used. The mechanism, therefore, can be described as follows: Oscillations of the layer structure are rigidly accompanied by oscillations of the vertical velocity (no permeation)  $90^\circ$  out of phase. These vertical velocity oscillations transport heat periodically up and down and generate vertical temperature oscillations. By the driving external temperature gradient these temperature oscil-

lations produce a gravitational force directed up and down periodically. If  $\theta < 0$  (heating from below) this "buoyancy" force is directed opposite to the elastic force of the layer oscillations. If  $|\theta|$  exceeds a certain threshold  $|\theta_c|$ , this buoyancy force overcomes the elastic force and the system becomes unstable. This behavior at threshold is sketched in Fig. 4. It shows a two-dimensional convection pattern oscillating as a whole. A quarter period later oscillations of the temperature and of layer deformations occur. Therefore, the situation is different from that in nematics,<sup>14,15</sup> where  $w$  and  $T$  are in phase and the order-parameter fluctuations are out of phase. The reason again, is that in nematics  $\omega_c < \hat{\kappa}^2$ , while in smectics  $A$  or cholesterics  $\omega_c \gg \hat{\kappa}^2$ . This happens, because in the latter case the elastic energy is dominating, which in nematics, is absent or only very weakly produced by a stabilizing magnetic field applied.

Without the approximations used above, the behavior is much more complicated. As eigenfunctions to  $\omega = \omega_c$  of Eq. (5.4) according to the boundary conditions (5.5) we find

$$\begin{aligned} w &= R_0 f(\vec{r}) (\zeta \hat{\chi}^2 \sin \omega_c t + \omega_c \cos \omega_c t), \\ v &= 0, \\ u &= -\frac{k_{\parallel}^c}{k_{\perp}^c} R_0 g(\vec{r}) (\zeta \hat{\chi}^2 \sin \omega_c t + \omega_c \cos \omega_c t), \\ R &= R_0 f(\vec{r}) \sin \omega_c t, \\ T &= -R_0 \frac{\theta_c}{d} f(\vec{r}) \left( \frac{\omega_c^2 - \hat{\kappa}^2 (\kappa_a k_{\perp}^c - \zeta \hat{\chi}^2)}{\omega_c^2 + \hat{\kappa}^2 \hat{\kappa}^2} \sin \omega_c t \right. \\ &\quad \left. + \frac{\kappa_{\parallel} k_{\perp}^c - \zeta \hat{\chi}^2}{\omega_c^2 + \hat{\kappa}^2 \hat{\kappa}^2} \cos \omega_c t \right), \end{aligned} \quad (5.9)$$

with  $f(r) = \sin(k_{\parallel}^c z) \cos(k_{\perp}^c x)$  and  $g(r) = \cos(k_{\parallel}^c z) \times \sin(k_{\perp}^c x)$ , Eqs. (5.9) describe two-dimensional convection rolls (the same spatial pattern as for the stationary instability), which oscillate with frequency  $\omega_c$ . Thereby  $w$ ,  $R$ , and  $T$  are not in phase with each other. For  $\hat{\mu}^4 k^{-2} \gg \hat{\kappa}^2 \gg \zeta \hat{\chi}^2$  we obtain

$$\begin{aligned} w &= R_0 f(\vec{r}) \omega_c \cos(\omega_c t - \omega_c^{-1} \zeta \hat{\chi}^2), \\ R &= R_0 f(\vec{r}) \sin \omega_c t, \\ T &= -\frac{\theta_c}{d} R_0 f(\vec{r}) \sin \left( \omega_c t + \frac{\kappa_{\parallel} k_{\perp}^c - \zeta \hat{\chi}^2}{\omega_c} \right). \end{aligned} \quad (5.10)$$

Neglecting the small phase lags one arrives at the situation described in Fig. 4. Again, we want to point out here, that above  $\theta_c$  the actual spatial pattern of the convection may be more complicated than described by Eqs. (5.9).

We now investigate the oscillatory instability with the help of the balance of different energy

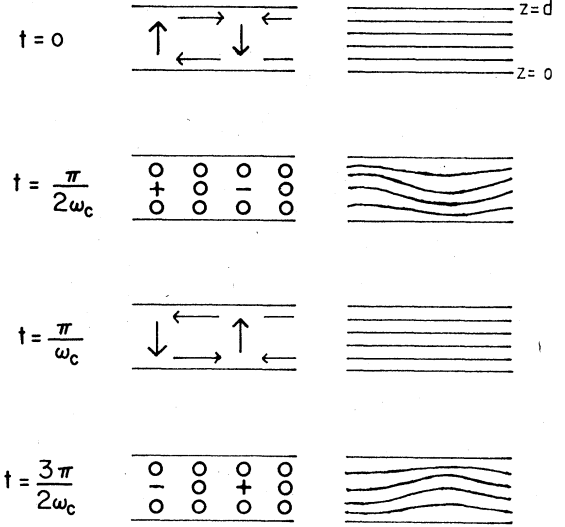


FIG. 4. The time-space pattern of the oscillatory instability at threshold;  $\rightarrow$  denotes convective velocities,  $+$  and  $-$  denotes hotter and colder regions, respectively, and  $\approx$  denotes the layer structure.

changes. As in the case of the stationary instability, we can calculate the rate of dissipation of kinetic and elastic energy  $\dot{\epsilon}_{\text{kin}}^{\text{dis}}$ ,  $\dot{\epsilon}_{\text{el}}^{\text{dis}}$  and the rate of energy  $\dot{\epsilon}_{\text{in}}$  fed into the system by the gravitational field. Using the eigenfunctions (5.9)—instead of (5.7)—we obtain

$$\begin{aligned} \dot{\epsilon}_{\text{kin}}^{\text{dis}} &= -\frac{R_0^2}{4\pi^2} \frac{\rho_0 \hat{\mu}^4}{k_{\perp}^c} (\zeta \hat{\chi}^2 \sin \omega_c t + \omega_c \cos \omega_c t)^2, \\ \dot{\epsilon}_{\text{el}}^{\text{dis}} &= -\frac{R_0^2}{4\pi^2} \zeta \hat{\chi}^2 \chi^2 \sin^2 \omega_c t, \\ \dot{\epsilon}_{\text{in}} &= -\frac{R_0^2}{4\pi^2} \frac{\theta_c}{d} \alpha g_E (\omega_c^2 + \hat{\kappa}^2 \hat{\kappa}^2)^{-1} \\ &\quad \times (\zeta \hat{\chi}^2 \{\omega_c^2 - \hat{\kappa}^2 [\kappa_a (k_{\perp}^c)^2 - \zeta \hat{\chi}^2]\} \sin^2 \omega_c t \\ &\quad + \omega_c^2 [\kappa_{\parallel} (k_{\perp}^c)^2 - \zeta \hat{\chi}^2] \cos^2 \omega_c t \\ &\quad + \{\omega_c^2 - \hat{\kappa}^2 [\kappa_a (k_{\perp}^c)^2 - \zeta \hat{\chi}^2] \\ &\quad + \zeta \hat{\chi}^2 (\kappa_{\parallel} k_{\perp}^c - \zeta \hat{\chi}^2)\} \omega_c \sin \omega_c t \cos \omega_c t). \end{aligned} \quad (5.11)$$

However, by the nonstationary nature of the motions here, the kinetic and elastic energy may not only be changed by dissipation, but also by their explicit time dependence. The condition for the onset of an oscillatory instability, therefore, reads<sup>31</sup>

$$\dot{\epsilon}_{\text{kin}}^{\text{dis}} + \dot{\epsilon}_{\text{el}}^{\text{dis}} + \dot{\epsilon}_{\text{in}} = \frac{\partial}{\partial t} (\epsilon_{\text{kin}} + \epsilon_{\text{el}}), \quad (5.12)$$

with

$$\epsilon_{\text{kin}} = \frac{1}{2V} \int d^3\vec{r} \rho(\vec{r}, t) \vec{v}^2(\vec{r}, t),$$

$$\epsilon_{\text{el}} = \frac{1}{2V} \int d^3\vec{r} \chi_{ij} [\nabla_i R(\vec{r}, t)] [\nabla_j R(\vec{r}, t)].$$

Inserting Eqs. (5.9) one obtains

$$\begin{aligned} \frac{\partial}{\partial t} \epsilon_{\text{kin}} &= \frac{1}{V} \int d^3\vec{r} \rho (\dot{u}\dot{u} + \dot{w}\dot{w}) \\ &= -\frac{R_0^2}{4\pi^2} \rho_0 \omega_c \frac{k_{c2}^2}{k_{c1}^2} [(\omega_c^2 - \xi^2 \hat{\chi}^2 \chi^2) \sin \omega_c t \cos \omega_c t \\ &\quad + \omega_c \xi \hat{\chi}^2 (\sin^2 \omega_c t - \cos^2 \omega_c t)], \end{aligned} \quad (5.13)$$

$$\frac{\partial}{\partial t} \epsilon_{\text{el}} = \frac{R_0^2}{4\pi^2} \omega_c \hat{\chi}^2 \sin \omega_c t \cos \omega_c t.$$

The threshold condition (5.12) must be fulfilled at any time, i.e., in a synchronous manner for the oscillatory time dependence here. Matching the  $\cos^2$ ,  $\sin^2$ , and  $\text{sincos}$  terms obtained by putting Eqs. (5.13) and (5.11) into (5.12), there are two independent conditions left, which are identical with the threshold conditions (5.2). As can be seen from (5.11) there is, for any cycle, a net gain of energy ( $\int_0^{2\pi/\omega_c} \dot{\epsilon}_{\text{in}} dt > 0$ ) for the system, if  $\theta < 0$  (heating from below). The instability sets in, if  $\dot{\epsilon}_{\text{in}}$  is great enough, i.e., for  $\theta = \theta_c$ .

#### D. Comparison between stationary and oscillatory instability

We will discuss here, under which conditions the stationary or the oscillatory instability occurs. Of course, by heating from above ( $\theta_c > 0$ ) only the stationary instability can occur, namely, if  $\kappa_a (k_{c1}^c)^2 - \xi \hat{\chi}^2 > 0$ . By heating from below ( $\theta_c < 0$ ), that instability appears, whose threshold temperature gradient is reached first, i.e., whose  $|\theta_c|$  is smaller. Although the values of  $\theta_c$  depend on various phenomenological parameters, it is possible to give a general discussion. The only parameter which varies over a broad range and which is crucial for the value of  $\theta_c^{\text{STI}}$  is  $\kappa_a$ . The second quantity, which can be influenced considerably, is the strength of the external magnetic field.

In the following we will discuss the  $\kappa_a$  and  $H$  dependence of the threshold gradients  $\theta_c$  and of the frequency  $\omega_c$  for smectics  $A$  and cholesterics. Although for the latter the correct threshold conditions are given in Sec. VI, the following considerations are a first approximation for cholesterics; the necessary additional discussion is

given in Sec. VI.

The numerical results are obtained under the assumption  $d \approx 1$  cm, pitch  $2\pi q_0^{-1} \approx 50$   $\mu\text{m}$  for cholesterics, and interlayer distance 500  $\text{\AA}$  for smectics  $A$ . For transport parameters we use the values of (4-methoxybenzylidene-4'-butylaniline) (MBBA).<sup>18</sup> Since the layers in smectics  $A$  are stiffer than those in cholesterics, the elastic energy  $\sim \chi_{\parallel} k_{\parallel}^2$  is much greater in smectics  $A$  than in cholesterics ( $\chi_{\parallel} \sim q_0^2$ ). Therefore, the influence of a magnetic field ( $\pm \chi_a H^2 k_{\perp}^2$ ) is compared to  $\chi_{\parallel} k_{\parallel}^2$  less important for smectics  $A$ .

We first discuss the stationary instability. The adjacent critical value of the transverse wave vector  $k_{\perp}^c = \sqrt{B} \pi/d$  is given by the first equation of (5.6). Its value depends on the magnetic-field strength. One can roughly distinguish three domains (cf. Fig. 5) for stabilizing magnetic fields

$$\begin{aligned} B &= \left( \frac{d^2}{\pi^2} \frac{\kappa_{\parallel} \chi_{\parallel}}{2\kappa_{\perp} K} \right)^{1/3}, \text{ for small fields,} \\ B &= \left( \frac{\kappa_{\parallel} \chi_{\parallel}}{\kappa_{\perp} |\chi_a| H^2} \right)^{1/2}, \text{ for intermediate fields,} \\ B &= \left[ \frac{\kappa_{\parallel} \chi_{\parallel}}{\kappa_{\perp} |\chi_a| H^2} \left( 1 + \frac{|\chi_a| H^2}{K q_0^2} \right) \right]^{1/2}, \text{ for strong fields.} \end{aligned} \quad (5.14)$$

For cholesterics we have in the low-field regime  $(k_{\perp}^c)^2 \approx 30 \pi^2/d^2$  and in the intermediate regime ( $H \geq 1$  kG)  $(k_{\perp}^c)^2 \approx 2 \times 10^4 (\pi^2/Hd^2)$  G. The saturation regime ("strong fields") is reached for  $H \geq 30$  kG and  $(k_{\perp}^c)^2 \approx \pi^2/d^2$ .

The expression (5.14) for  $B$  in intermediate fields is in agreement with Parsons,<sup>19</sup> that for small fields differ with respect to the exponent  $\frac{1}{3}$  from Dubois-Violette's expression<sup>18</sup> (exponent  $\frac{1}{2}$ ); the reason is that Dubois-Violette maximized the denominator of  $\theta_c$  instead of minimizing the whole quotient. For the critical temperature difference  $\theta_c^{\text{STI}}$  (5.3) the magnetic-field dependence is rather weak.

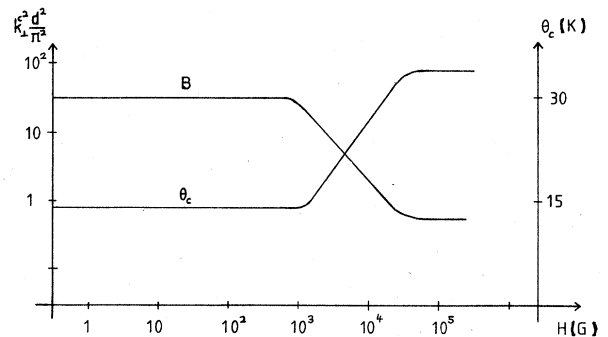


FIG. 5. The transverse wave vector  $k_{\perp}^c$  and the critical temperature gradient  $\theta_c$  for the stationary instability in cholesterics MBBA as a function of an external magnetic field  $H$ .



The magnetic part of the elastic energy ( $\sim \chi_a H^2$ ) is greater than the nonmagnetic part ( $\sim \chi_{||}$ ) only for fields  $> 5$  kG; in that range  $(k_1^c)^2$  drops  $\sim H^{-1}$  and  $\chi_a H^2 (k_1^c)^2 \sim H$  increases slowly with increasing magnetic field. For cholesterics MBBA  $\theta_c^{STI} \approx 16$  K constant up to roughly 1 kG, then slowly increasing up to 35 K for 20 kG (Fig. 5). For smectics A,  $\theta_c^{STI}$  is too high to be observable in any stabilizing magnetic field. In order to obtain experimentally relevant values for  $\theta_c^{STI}$ , one has to use a destabilizing magnetic field, which reduces the elastic energy. For fields in the range of 10 kG, the elastic energy  $\hat{\chi}^2$  goes to zero and  $\theta^{STI}$ , which is proportional to  $\hat{\chi}^2$ , can be made as small as necessary for the experimental application (cf. Appendix B).

We will now turn to the oscillatory instability. The transverse wave vector  $k_1^c$  is given by the second equation of (5.6). For low fields,  $B \approx \frac{1}{2}$  and  $k_1^{c2} \approx \pi^2/2d^2$ , as in the Bénard instability in simple fluids. For magnetic fields  $H \geq 10^3$  G for cholesterics

$$B = \left( \frac{\pi^2}{d^2} \frac{\kappa_{||} \nu_3}{|\chi_a| H^2} \right)^{1/2} \quad (5.15)$$

and  $(k_1^c)^2 \approx \frac{1}{2} \times 10^3 (\pi^2/Hd^2)$  G. This behavior is sketched in Fig. 6. Therefore,  $k_1^c$  for the oscillatory instability is much smaller than  $k_1^c$  for the stationary instability. This has the consequence that for the oscillatory instability  $|\chi_a| H^2 (k_1^c)^2$  is—for all magnetic fields available—smaller than  $\chi_{||} (k_{||}^c)^2$ . Therefore  $\theta_c^{OSI}$  [Eq. (5.4)] is roughly independent of a magnetic field. For MBBA one finds  $\theta_c^{OSI} = -\chi_{||} \alpha^{-1} g_E^{-1} \pi^2/d^2 \approx -5$  K, the absolute value of which is smaller than  $\theta^{STI}$ . For smectics A a large destabilizing magnetic field is necessary to lower  $|\theta_c^{OSI}|$  to experimentally useful values (cf. Appendix B).

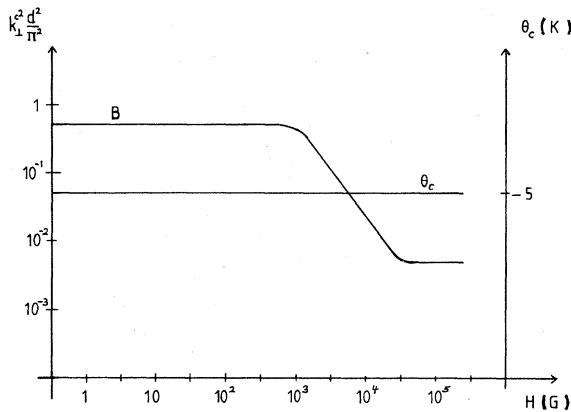


FIG. 6. The transverse wave vector  $k_1^c$  and the critical temperature gradient  $\theta_c$  for the oscillatory instability in cholesterics MBBA as a function of an external magnetic field  $H$ .

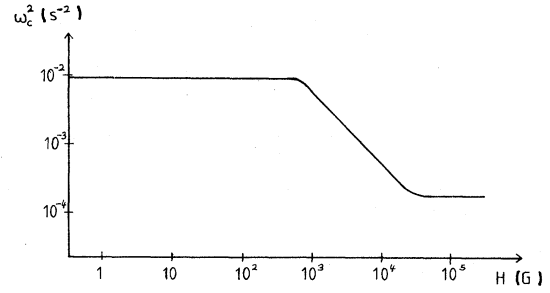


FIG. 7. The critical frequency  $\omega_c$  for the oscillatory instability in cholesterics MBBA as a function of an external magnetic field  $H$ .

The frequency belonging to the oscillatory instability  $\omega_c$  [Eq. (5.4)] is magnetic-field dependent since  $\omega_c^2 \sim k_1^2$ . Again, two regimes are to be distinguished (Fig. 7) [cf. (5.15)]:

$$\omega_c^2 \approx \frac{k_{||} \chi_{||}}{2\rho_0} \left( \frac{\pi}{d} \right)^4 \approx 10^{-2} Hz^2, \quad \text{for } H \leq 10^3 \text{ G}, \quad (5.16)$$

$$\omega_c^2 \approx \frac{\kappa_{||} \chi_{||}}{\rho_0 H} \left( \frac{k_{||} \nu_3}{|\chi_a|} \right)^{1/2} \left( \frac{\pi}{d} \right)^5 \approx \frac{10}{H} Hz^2 \text{ G}, \quad \text{for } H > 10^3 \text{ G},$$

We are now able to discuss what instability will actually occur by heating from below (for cholesterics). Equations (5.3) and (5.4) show that  $|\theta_c^{STI}|/|\theta_c^{OSI}| = (\hat{\chi}_{STI}^2/\hat{\chi}_{OSI}^2)(\kappa_{||}(k^c)^2 - \kappa_a(k_1^c)^2)/(\xi\hat{\chi} - \kappa_a(k_1^c)^2)|_{STI} = Q(\zeta\hat{\chi}^2 > \kappa_a(k_1^c)^2$  by heating from below and  $\kappa_{||}k^2 > \kappa_a k_1^2$  in any case). Taking into account the magnetic-field dependence of  $\hat{\chi}^2$  and  $(k_1^c)^2$  discussed above, it is easily checked that  $Q > 1$  (for any magnetic field); only in the unrealistic limit  $\kappa_a \rightarrow -\infty$  ( $k_1 \rightarrow +\infty$ )  $Q \rightarrow 1$ . Therefore, in cholesterics the oscillatory instability always occurs by heating from below. A stability diagram for cholesterics is given in Fig. 8.

It should be kept in mind that the differences

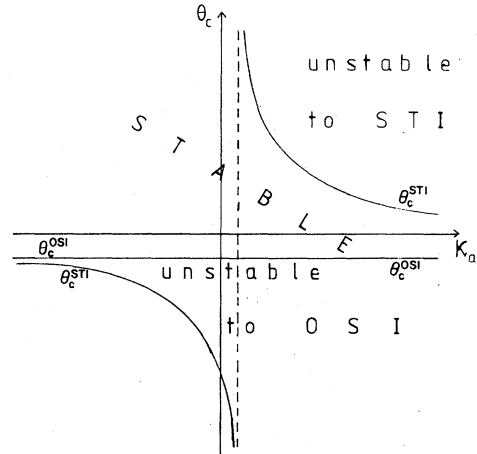


FIG. 8. The stability diagram for cholesterics.

between smectics *A* and cholesterics discussed in this section are only due to differences in the magnitude of parameters, e.g., the pitch (or interlayer distance). Therefore, for smecticlike systems with interlayer distance in the range  $10^{-3}$  cm (like the pitch in cholesterics) both instabilities will become observable without using a destabilizing magnetic field. The differences between smectics *A* and cholesterics based on their different hydrodynamic equations will be discussed in the following section.

## VI. CONVECTIVE INSTABILITIES IN CHOLESTERIC

In Sec. V we have discussed the convective instabilities in cholesterics on the basis of hydrodynamic equations strictly valid only for smectics *A*. As already discussed (Sec. II), there are, however, more complicated hydrodynamic equations for cholesterics than for smectics *A*, due to the lack of mirror planes and the possibility of a distinction between left- and right-handed helices in cholesterics.<sup>28</sup> These additional hydrodynamics terms do not contribute to the mode spectrum  $\omega(k)$  in linear order of  $k$ , but are important if external thermodynamic forces are applied. We will now investigate how the results of Sec. V concerning cholesterics are changed, if these additional contributions to the hydrodynamic equations, which are represented by the right-hand sides of Eqs. (4.2), are taken into account. There are two sorts of such new terms, one with the parameter *A*, and one with parameters  $g_{\parallel}$ ,  $g_{\perp}$ ; actually, *A* = 1 was introduced only to characterize these terms.

Let us first concentrate on that convective instability, which has already been found by Dubois-Violette<sup>18</sup> and Parsons<sup>19</sup> to be stationary, and which is discussed in Sec. V B. The *A* terms, which provide a coupling of the velocity component  $v$  and the layer displacement  $R$  [cf. (4.3)], can easily be taken into account, by replacing  $\zeta \rightarrow \zeta + k_1^2(4q_0^2\rho_0\hat{\mu}_y^2)^{-1}$ . Therefore, the critical temperature  $\theta_c$  of Eq. (5.1) is changed into

$$\theta_c^{\text{STI}} = \frac{\rho_0 d}{\alpha g_E} \frac{\hat{\chi}^2 \hat{\kappa}^2}{k_1^2} \frac{\zeta \hat{\mu}^4 + k_1^2 \rho_0^{-1} + \hat{\mu}^4 k_1^2 (4q_0^2 \rho_0 \hat{\mu}_y^2)^{-1}}{\kappa_a k_1^2 - \hat{\chi}^2 - \hat{\chi}^2 k_1^2 (4q_0^2 \rho_0 \hat{\mu}_y^2)^{-1}}, \quad (6.1)$$

which reduces for  $\hat{\mu}^4 k^{-2}$ ,  $\hat{\mu}_y^2 \gg \hat{\kappa}^2$ ,  $\zeta \hat{\chi}^2$  to

$$\theta_c^{\text{STI}} = \frac{d}{\alpha g_E} \frac{\hat{\chi}^2 \hat{\kappa}^2}{\kappa_a k_1^2 - \hat{\chi}^2 [\zeta + k_1^2 (4q_0^2 \rho_0 \hat{\mu}_y^2)^{-1}]}. \quad (6.2)$$

The value of the critical wave vector  $k_1^c$  remains that which has already been discussed in Sec. V D. Therefore  $(k_1^c)^2 \ll q_0^2$ , and the new contribution in the denominator is roughly of the same order of magnitude as  $\zeta \hat{\chi}^2$ . In the stability diagram (Fig. 8) the curve for  $\theta_c^{\text{STI}}$  and the dotted line have to be shifted to the right at an amount  $\hat{\chi}^2(4q_0^2\rho_0\hat{\mu}_y^2)^{-1}$ .

However, quite more important than this change in the value of  $\theta_c$  is the fact, that by this new term the second transverse velocity component is no longer zero [as in (5.7)], but given by

$$v(\vec{r}) = (2q_0\rho_0\hat{\mu}_y^2)^{-1} \frac{\partial}{\partial x} R(\vec{r}). \quad (6.3)$$

The other eigenfunctions are given by (5.7), if, again  $\zeta$  is replaced by  $\zeta + k_1^2(4q_0^2\rho_0\hat{\mu}_y^2)^{-1}$ . Thus, the convection pattern is now manifestly three dimensional, and two-dimensional rolls cannot occur. This can easily be understood by the physical meaning of these new terms. They describe the connection between a translation along  $\hat{p}$  (=  $\hat{e}_y$ ) (described by  $R$ ) with a rotation about  $\hat{p}$  (described by  $\nabla_y u - \nabla_x v$ ) of the helices. Thus, both transverse velocity components are involved prohibiting any two-dimensional flow pattern. It is obvious in Eq. (6.3), that  $v(\vec{r})$  is maximal or minimal, where  $R(\vec{r})$  and therefore  $w(\vec{r})$  [and  $u(\vec{r})$ ] are zero and vice versa.

In order to include the  $g$  terms we have to redo the linear stability analysis of Sec. V A. The resulting formulas are very complicated and we will not write the general expressions. However, although we do not know the numerical value of  $g_a = g_{\parallel} - g_{\perp}$ , we can assume that  $g_a q_0 k_{\parallel} < 1$ , since otherwise the gradient expansion which underlies the hydrodynamic equation, would break down. Therefore, we expand our results in the small quantity  $g_a q_0 k_{\parallel}$ . In that case the threshold-gradient temperature  $\theta_c$  is changed from its value (6.1) or (6.3) only by an additional contribution  $\sim (g_a q_0 k_{\parallel})^2$  which can be neglected. The most striking new effect, however, is the occurrence of a critical frequency  $\omega_c \neq 0$  with (in linear order of  $g_a q_0 k_{\parallel}$ )

$$\omega_c(k) = -q_0 g_a k_{\parallel} \kappa_a k_1^2 \hat{\mu}_y^2 \left( \frac{\alpha g_E \theta_c}{d \rho_0} \right) k_1^2 \left[ \left( \frac{\alpha g_E \theta_c}{d \rho_0} \right) k_1^2 (\hat{\mu}_y^2 + \zeta \hat{\chi}^2 - \kappa_a k_1^2) + (\hat{\kappa}^2 + \hat{\mu}_y^2) (\zeta \hat{\chi}^2 \hat{\mu}^4 + k_1^2 \rho_0^{-1} \hat{\chi}^2) + \hat{\kappa}^2 \hat{\mu}_y^2 (\hat{\mu}^4 + k_1^2 \zeta \hat{\chi}^2) + \left( \frac{k_1^2 \hat{\chi}^2}{4q_0^2 \rho_0} \right) (\hat{\mu}^4 + \hat{\kappa}^2 k_1^2) \right]^{-1}, \quad (6.4)$$

where  $\theta_c$  is given by (6.1). For  $\hat{\mu}^4 k^{-2}$ ,  $\hat{\mu}_y^2 \gg \hat{\kappa}^2$ ,  $\zeta \hat{\chi}^2$ ,  $\omega_c(k)$  simplifies to

$$\dot{\omega}_c(k^c) = -q_0 g_a k_{\parallel}^c \hat{\kappa}^2 \frac{\kappa_a (k_{\perp}^c)^2}{\kappa_{\parallel} k_a^2}. \quad (6.5)$$

Therefore, the “stationary” instability obtained by Refs 18 and 19 and discussed in Sec. V B is *not* stationary. We stress here that this nonstationary behavior is obtained, although we have neglected (as in Secs. IV and V) the temperature dependence of the pitch. The mechanism involved has, thus, nothing to do with a nonuniform pitch in the heat-conduction state (cf. Sec. VII). The overstability occurs, since via the  $g$  terms there is an additional coupling between  $R$  and  $w$  (besides that already present without  $g$ ), which prohibits the stationary balance between the energy dissipated and flown in required for a stationary threshold. Although there is no simple geometrical explanation possible for the  $g$  terms (like in the case of nematics for the  $\lambda$  terms<sup>29,30</sup>), it can easily be seen from Eq. (4.2) that  $R$  and  $w$  can no longer have the same  $z$  dependence, since  $R$  couples to  $w$  and  $\nabla_z w$ . Therefore, the dissipation of elastic energy ( $\sim R^2$ ) and kinetic energy ( $\sim w^2$ ) can match the condition for the onset of a stationary instability only for distinct values of  $z$ , but not everywhere. If there are regions ( $z > z_0$ ), where the dissipated energy is smaller than that flowing in, and other regions ( $z < z_0$ ), where the contrary holds, there will be a transport of energy between these two regions. At the true threshold this energy transfer is undamped and oscillatory. The characteristic frequency connected with this vertical transport is given by Eq. (6.4). It vanishes for  $g_a \rightarrow 0$  and  $\theta_c \rightarrow 0$  for obvious reasons. We cannot give numerical values for  $\omega_c$ , since we do not know  $g_a$ . As an upper limit ( $|g_a q_0 k_{\parallel}| \leq 1$ ) for  $|\omega_c|$  we can take  $10^{-3}$  Hz ( $t_c \approx 2$  h), but probably  $|\omega_c|$  is smaller.

The eigenfunctions belonging to Eq. (6.4) are now too complicated to give analytic expressions; especially, by the different  $z$  dependence of  $R$  and  $w$  (caused by the  $g$  terms) there is no single mode [ $\sim \sin \pi_z/d$ ] behavior possible, even for free-free boundary conditions. Other possible  $k_{\parallel}$  values ( $k_{\parallel}^n$ ) and even solutions ( $\cos k_{\parallel}^n z$ ) have to be included by numerical means, a procedure which is required for realistic boundary conditions anyway and which is described for the case of simple fluids in Chandrasekhar's book.<sup>31</sup>

We will now discuss the influence of the specific cholesteric terms in the hydrodynamic equations on the oscillatory instability described in Sec. V C. The numerical values of the critical temperature  $\theta_c$  and the critical frequency  $\omega_c$  are only little changed (<1%) by the  $A$  and  $g$  terms of (4.2). We refrain, therefore, from writing the new complicated expressions and the expressions (5.2) and (5.4) for  $\theta_c$  and  $\omega_c$  are still suitable. The main effects

of these terms specific for cholesterics are, again, that no two-dimensional flow pattern and no simple mode pattern (with respect to  $z$ ) can occur. The oscillatory behavior induced by the  $g_a$  terms is however, completely eclipsed by the intrinsic oscillatory nature of this instability. Although in cholesterics both instabilities are oscillatory, they are clearly distinguishable. The first (“stationary”) instability occurs by heating from above and its characteristic frequency is lower than  $10^{-3}$  Hz (and vanishes for  $g_a \rightarrow 0$ ), while the second (oscillatory) one occurs by heating from below with a frequency in the range of  $10^{-1}$  Hz.

## VII. INFLUENCE OF THE TEMPERATURE DEPENDENCE OF THE PITCH

In Sec. III we have found that in the steady heat-conduction regime the distances between the layers differ from their constant value in the true equilibrium state and depend on  $z$  and on the temperature gradient applied. This “temperature dependence of the pitch”, which in Eq. (3.1) is described by  $R^{\text{HC}} = -\frac{1}{2} z^2 \theta d^{-1} \tilde{\chi}_{\parallel}^{-1} \eta$  with  $\eta = \xi \zeta^{-1} + \gamma_2 C_p T_0^{-1}$ , is due to a static (via the susceptibility  $\gamma_2$ ) and a dynamic (via the dissipative transport parameter  $\xi$ ) cross coupling between  $R$  and  $T$ . [Note, however, that the pitch of the true equilibrium state ( $\theta = 0$ ), namely,  $2\pi q_0^{-1}$  is held constant like all other material parameters (Boussinesq approximation).] In Secs. IV–VI we have neglected these cross couplings ( $\gamma_2 = 0 = \xi$ ). We will now discuss the influence of  $\gamma_2$  and  $\xi$  on the stability of the heat-conduction regime in smectics  $A$  and cholesterics. Since  $\gamma_2$  and  $\xi$  are small parameters (which was the reason we neglected them completely in Secs. IV–VI), we only take into account contributions linear in  $\gamma_2$  and  $\xi$ . The linear stability analysis is then performed as in Sec. IV.

The Boussinesq approximation contains the same assumptions as usual [cf. (4.1)] and, in addition, the neglect of terms proportional to  $f_a \eta$ . {The reversible transport parameters  $f_{\parallel}$ ,  $f_{\perp}$ , which are absent in linear hydrodynamics [cf. (2.1)], are assumed to be small, at least  $f_{\parallel}, f_{\perp} < 1$ . We also neglect terms  $\sim \eta$  times contributions due to nonlinear elasticity (cf. Ref. 23).} The system of algebraic equations for the linear deviations from the heat-conduction state then reads

$$\begin{aligned} i \omega R + \dots &= \dots + i k_{\parallel} \xi \eta T, \\ i \omega v + \dots &= \dots, \\ i \omega w + \dots &= \dots + i k_{\parallel} (\xi \zeta^{-1} + \eta) \frac{\theta}{d} R, \\ -k^2 p + \dots &= \dots - k_{\parallel}^2 (\xi \zeta^{-1} + \eta) \frac{\theta}{d} R - 2k_{\perp}^2 \xi \zeta^{-1} \frac{\theta}{d} R, \\ i \omega T + \dots &= \dots - i k_{\parallel} \xi C_p T_0^{-1} \hat{\chi}^2 R, \end{aligned} \quad (7.1)$$

where the ellipses denote all terms already present for  $\xi=0=\gamma_2$  and which can be found on the left- and right-hand side of Eq. (4.2). This system of equations allows, again, for two solutions, i.e., two instabilities which correspond to the stationary and the oscillatory instability discussed already in Secs. V and VI without  $\gamma_2$  and  $\xi$ . However, with respect to the latter, the differences in  $\omega_c$  and  $\theta_c$  caused by the  $\xi$  and  $\gamma_2$  terms are so small that we do not give explicit new formulas. Equations (5.2) or (5.4) are, therefore, very good approximations for the case  $\xi \neq 0$ ,  $\gamma_2 \neq 0$ . However, there are no single mode ( $\sin \pi z/d$ ) solutions possible and  $k_{||} \neq \pi/d$ ; this already happened for cholesterics by including the  $g$  terms (Sec. VI).

We therefore concentrate on that instability which is stationary in smectics (Secs. V A, and V B) and becomes oscillatory in cholesterics due to the  $g$  terms (Sec. VI). Parsons already discussed the influence of a nonconstant pitch in the heat-conducting state for the stationary instability in cholesterics (using the simplified equations strictly valid only for smectics A). However, he neglected the dynamical effect (i.e., put  $\xi=0$ ) and took into account the static coupling between  $R$  and  $T$  only partially, i.e., the term  $\phi_x \sim \gamma_2 \delta T$  [cf. (2.2)], while he omitted the thermodynamic counter term  $\sigma \sim \gamma_2 \partial R / \partial z$ . Taking into account the static and dynamic effect, we obtain for the threshold condition in linear order,

$$\theta_c^{\text{STI}} = \theta_c^{\text{STI}}(\gamma_2 = 0 = \xi) + O(2)$$

and

$$\begin{aligned} \omega_c = k_{||} \hat{\mu}_y^2 k_{\perp}^2 N^{-1} & \left[ \gamma_2 C_p T_0^{-1} \frac{\theta_c}{d \rho_0} (\zeta \hat{\chi}^2 + \zeta \rho_0 \kappa_a \hat{\mu}^4 + \hat{\kappa}^2) \right. \\ & \left. + \xi \frac{\theta_c}{d} \left( \kappa_a \hat{\mu}^4 + \rho_0^{-1} \hat{\chi}^2 - \hat{\chi}^2 \frac{\alpha g_E d T_0}{C_p \theta_c \rho_0} \right) \right] \\ & + \omega_c(g_a) A' + O(2) \end{aligned} \quad (7.2)$$

with

$$\begin{aligned} N = \frac{\alpha g_E \theta_c}{d \rho_0} k_{\perp}^2 & \left[ \hat{\rho}_y^2 + A' (\zeta \hat{\chi}^2 - \kappa_a k_{\perp}^2) \right] + \hat{\kappa}^2 \hat{\mu}_y^2 (\hat{\mu}^4 + k^2 \zeta \hat{\chi}^2) \\ & + (A' \hat{\kappa}^2 + \hat{\mu}_y^2) (\zeta \hat{\chi}^2 \hat{\mu}^4 + k_{\perp}^2 \rho_0^{-1} \hat{\chi}^2) + A' \frac{k_{\perp}^2 \hat{\chi}^2}{4 q_0^2 \rho_0} (\hat{\mu}^4 + k^2 \hat{\kappa}^2). \end{aligned}$$

The critical temperature gradient  $\theta_c^{\text{STI}}(\gamma_2 = 0 = \xi)$  is given for smectics A by (5.1) and for cholesterics by (6.1), since  $\theta_c$  is changed only in quadratic order of  $\gamma_2$  and  $\xi$ . The instability is now oscillatory even for smectics A. In the formula (7.2) for the critical frequency,  $A'$  is zero or one for smectics A and cholesterics, respectively. The term  $A' \omega_c(g_a)$  denotes the frequency due to the  $g$  term in cholesterics and is given by (6.4). The neglected terms  $O(2)$  are proportional to  $\gamma_2^2$ ,  $\gamma_2 \xi$ ,  $\xi^2$ ,  $f_a \gamma_2$ ,  $f_a \xi$ ,  $g_a^2$ ,

$g_a \gamma_2$ , and  $g_a \xi$ . For  $\hat{\mu}^4 k^{-2}$ ,  $\hat{\mu}_y^2 \gg \hat{\kappa}^2$ ,  $\zeta \hat{\chi}^2$ , Eq. (7.2) reduces to

$$\omega_c \approx \frac{k_{||}}{\alpha g_E} (\gamma_2 C_p T_0^{-1} \hat{\kappa}^2 + \xi \hat{\chi}^2) - A' g_a g_0 k_{||} \hat{\kappa}^2 \frac{\kappa_a k_{\perp}^2}{k_{||} k^2}. \quad (7.3)$$

Note that neither the sign of  $\gamma_2$  and  $\xi$  nor the sign of  $g_a$  is fixed. Therefore, for cholesterics, the new frequency contributions due to  $\gamma_2$  and  $\xi$  have to be compared with that due to  $g_a$  which was discussed in Sec. VI. There are no exact data on  $\gamma_2$  or  $\xi$  available. By very rough estimates,<sup>18</sup> one is lead to  $\omega_c(\gamma_2, \xi) \lesssim 10^{-2} - 10^{-3}$  Hz or smaller. For cholesterics this frequency may have the same order of magnitude as  $\omega_c(g_a)$ . In any case, this frequency seems to be considerably smaller than the frequency of the oscillatory instability obtained by heating from below.

The mechanism leading to the oscillatory behavior was already described by Parsons<sup>19</sup> as far as it is related to the nonconstant pitch in the heat-conduction regime. But only parts of  $\omega_c$  (7.2) stem from  $R^{\text{HC}} \sim \eta \theta/d$ , namely,  $\omega_c \sim \eta(\theta_c/d) \times (\zeta \hat{\chi}^2 + \zeta \rho_0 \kappa_a \hat{\mu}^4)$ . There are, however, further contributions  $\omega_c \sim \gamma_2 \kappa^2 \theta_c/d$  and  $\omega_c \sim -\xi \hat{\chi}^2$ , which are present even if  $R^{\text{HC}} = 0$ ,  $\eta = 0$  or  $\gamma_2 C_p \xi = -T_0 \xi$ . The nonstationary behavior due to these terms arises—like in the case of the  $g$  terms (cf. Sec. VI)—from a different  $z$  dependence of kinetic and elastic energy due to simultaneous couplings of  $R$  and  $\partial R / \partial z$  to  $w$  and vice versa. A single mode behavior is impossible with these couplings (even for smectics A) and analytic formulas for the eigenfunctions cannot be given (even for smectics A).

## VIII. CONCLUSIONS

As it has become obvious recently it is now possible to produce monodomain cholesteric and smectic A liquid crystals of approximately 1-cm thickness. Therefore, the study of hydrodynamic instabilities of these types of liquid crystals comes now into experimental reach. In the present paper we have investigated the convective instabilities of smectic A and cholesteric liquid crystals, theoretically.

For smectics A we have found a stationary and an oscillatory instability which have not been discussed previously. When typical values for the interlayer distance are inserted into the expressions for the critical temperature difference and for the critical frequency we find that both instabilities are out of experimental reach for any stabilizing external magnetic field because of the very large elastic energy of smectic liquid crystals. To overcome this difficulty one has to apply a

large destabilizing magnetic field of order 10 kG in order to weaken the stiffness of the smectic layers, i.e., one must approach the Helfrich-Hurault instability. If one is interested in the investigation of smecticlike systems with large interlayer distance (comparable to the pitch of cholesteric liquid crystals) it will be possible to observe both types of instabilities in experiments.

The same is true for smectic-C liquid crystals. For these it is straightforward but somewhat tedious to carry out the analogous considerations as for smectics A. Because the corresponding expressions for the critical temperature differences and for the critical frequency look rather unwieldy we have refrained from writing them explicitly. Concerning the cholesteric liquid crystals it seems very important to note that there exists a stationary instability only in the simplest approximation. In this case we obtain results generalizing those of Dubois-Violette and Parsons. Furthermore we predict the occurrence of an intrinsically oscillatory instability which was not considered before. When compared with nematics where both types of instabilities have already been observed experimentally, it seems important to note that the mechanisms of the instabilities for cholesterics are completely different from those of nematics, due to the different orders of magnitude of the elastic energy and due to the fact that we have to face a broken translational symmetry in cholesterics compared to a broken orientational symmetry in nematics.

The threshold conditions for both the oscillatory and the stationary instability in cholesterics (and smectics) are reobtained by energy-balance considerations, i.e., if the energy fed into the system by the external force is compensated for by the dissipated energy. If the planar texture is heated from below we always find (for all  $\kappa_a$ ) the oscillatory instability, contrary to the case of nematics, where the type of instability (oscillatory or stationary) depends on the magnitude and sign

of  $\kappa_a$ . When heating from above the stationary instability is clearly only present for  $\kappa_a > \xi \chi^2 k_1^{-2}$ . The results have been supplemented by a discussion of the dependence of the transversal critical vector, the critical temperature difference, and the critical frequency on the magnitude of a stabilizing external magnetic field. Contrary to the case of nematics the critical frequency does not vanish for large magnetic fields but reaches a finite value. If the  $g_a$  terms and the Lubensky term are included in the description, there exists no true two-dimensional flow pattern and a three-dimensional structure occurs. (Of course its beyond the scope of a linearized theory to make predictions for the flow patterns occurring well above the critical temperature difference.) In addition, the stationary instability changes into an instability with a probably small but finite frequency  $\omega_c$  proportional to  $g_a$ . This means that no true stationary instability occurs in cholesteric liquid crystals (contrary to the case of nematics).

The inclusion of a temperature-dependent pitch leads to a heat-conduction state which depends on the coordinate along which the temperature gradient is applied. This dependence is due to a dynamic and a static coupling between the displacement vector and the temperature. It seems worthwhile to mention that these additional cross-coupling coefficients are typical for systems with broken translational symmetry (smectics A and cholesterics) and do not occur for systems with broken orientational symmetries like nematics.

These additional couplings lead to further contributions to the critical temperature differences and the critical frequency for cholesterics and change the stationary instability of smectics to an oscillatory one. To summarize, it seems to be very interesting to test the presented predictions experimentally and to get further information on the unknown phenomenological parameters involved.

#### APPENDIX A: INCLUSION OF FLUCTUATIONS OF THE PREFERRED AXIS

As long as one regards the preferred axis  $\hat{p}$  as completely fixed, only longitudinal gradients of  $R$  occur. Fluctuations of the axis  $\hat{p}$  show up in transverse gradients of  $R$  (i.e.,  $\delta \hat{p}_i \approx -\nabla_i^t R$ ). Since  $\epsilon \sim (\nabla_i \delta \hat{p}_i)^2$  (homogeneous rotations of  $\hat{p}$  do not change the energy<sup>23</sup>), these transverse gradients of  $R$  are of higher order in the gradient expansion. In Secs. II and III we neglected these terms in order to shorten the formulas. In Sec. IV we introduced the effect of fluctuating  $\hat{p}$  in an *ad hoc* manner by writing  $\hat{\chi}^2 = \dots \bar{K} k_1^4$  in (4.2). We will here justify the inclusion of the  $\bar{K} k_1^4$  term.

The basic hydrodynamic equations (2.1) have to be amended by the following expressions<sup>23</sup>:

$$\begin{aligned} \frac{\partial}{\partial t} g_i + \dots + \nabla_j \left[ \delta_{ij} \nabla_k \psi_{kj} + \psi_{jk} \nabla_k \nabla_i R + \psi_{kj} \nabla_k \nabla_i R + \left( \frac{1}{2q_0} \epsilon_{ijz} + g_{ij} q_0 + f_{ij} R \right) \nabla_k \nabla_m \psi_{km} \right] = \dots, \\ \frac{\partial}{\partial t} R + \dots = \dots - \xi \nabla_k \nabla_m \psi_{km}, \end{aligned} \quad (\text{A1})$$

where the ellipses denote the expressions already given in (2.1). The thermodynamic force  $\psi_{ij}$  is given by

$$\psi_{ij} = (\bar{K}\delta_{ij}^{\text{tr}}\delta_{K_m}^{\text{tr}} + K'\delta_{ik}^{\text{tr}}\hat{p}_j\hat{p}_m)\nabla_K\nabla_m R. \quad (\text{A2})$$

Expressed by Franck elastic constants  $\bar{K} = \frac{1}{3}(K_2 + 3K_3)$  and  $K' = \frac{1}{2}K_1$  for cholesterics<sup>25</sup> and  $\bar{K} = K_1$ ,  $K' = K_3$  for smectics *A*. Note that  $\chi_{11} = q_0^2 K_2$  in cholesterics.

The heat-conduction state (3.1) is unaltered and in addition to (3.2) there is

$$\psi_{K_m}^{\text{HC}}(z) = 0. \quad (\text{A3})$$

For the linear deviations from the heat-conduction state one obtains in addition to the expressions (...) already present:

$$\begin{aligned} \rho_0 \dot{w} + \dots + K^{\text{op}}R - \eta\chi_{11}^{-1}\frac{\partial}{\partial z}K^{\text{op}}R + q_0 g_{11}\frac{\partial}{\partial z}K^{\text{op}}R &= \dots, \\ \rho_0 \dot{v} + \dots - \frac{1}{2q_0}\frac{\partial}{\partial z}K^{\text{op}}R &= \dots, \\ \rho_0 \dot{u} + \dots + q_0 g_{11}\frac{\partial}{\partial z}K^{\text{op}}R &= \dots, \\ \dot{R} + \dots &= \dots - \zeta K^{\text{op}}R, \end{aligned} \quad (\text{A4})$$

with

$$K^{\text{op}} = \bar{K}\frac{\partial^4}{\partial x^4} + K'\frac{\partial^4}{\partial x^2\partial z^2}.$$

After Fourier transformation, all these terms can be incorporated in Eq. (4.2), where  $\eta$  is put equal to zero by writing

$$\hat{\chi}^2 = \hat{\chi}^2(\bar{K} = 0 = K') + \bar{K}k_{\perp}^4 + K'k_{\perp}^2k_{\parallel}^2. \quad (\text{A5})$$

Since  $\hat{\chi}^2(\bar{K} = 0 = K')$  already contains  $\chi_{11}k_{\parallel}^2$ , we neglect  $K'k_{\perp}^2k_{\parallel}^2$  in favor of  $\chi_{11}k_{\parallel}^2$ , because  $q_0^2 \gg k_{\perp}^2$ . Thus, the inclusion of the term  $\bar{K}k_{\perp}^4$  in  $\hat{\chi}^2$  in Sec. IV (and as a consequence in Secs. V and VI) is justified. For  $\eta \neq 0$  (used in Sec. VII) there is an additional term,  $\eta(\theta/d)\chi_{11}^{-1}ik_{\perp}(\bar{K}k_{\perp}^2 + K'k_{\parallel}^2)$  in the equation for  $\dot{w}$ , which is not covered by the replacement (A5). However, this term can be neglected against the term  $ik_{\parallel}\eta(\theta/d)R$ , already present [cf. (7.1)], since  $k_{\perp}^4 \ll k_{\parallel}q_0^2$ .

#### APPENDIX B: APPLICATION OF A TRANSVERSE MAGNETIC FIELD

In the main text we allow for a static homogeneous external magnetic field parallel to the preferred axis  $\hat{p}$ . This magnetic field changes the elastic energy of the layers. In our formulas this effect is shown up in  $\hat{\chi}^2 = \hat{\chi}^2(H=0) \pm \chi_a H^2 k_{\perp}^2$  (plus or minus for smectics *A* or cholesterics, respectively). Usually, such a field is stabilizing the equilibrium layer structure since  $\chi_a > 0$

( $\chi_a < 0$ ) for smectics *A* (cholesterics) in most cases.

In this Appendix we will deal with a transverse magnetic field  $\vec{H} \perp \hat{p}$ . For cholesterics such a field drives the system towards the nematic phase transition.<sup>28</sup> By the use of Boussinesq's approximation we have to avoid the vicinity of any phase transition. Thus, we will discuss the case  $\vec{H} \perp \hat{p}$  for smectics *A* only. For smectics *A*, on the other hand, a destabilizing magnetic field (of course, it destabilizes the equilibrium layer structure of plane layers not the smectic phase itself) was required, in order to reach the convective instability (cf. Sec. VD). For the usual case  $\chi_a > 0$ , which we will assume in the following, a field  $\vec{H} \perp \hat{p}^0$  acts as a destabilizer.

The transverse field introduces a second preferred direction perpendicular to  $\hat{p} (= \hat{e}_z)$ , i.e.,  $\vec{H}/|\vec{H}| = \hat{e}_x$ . Thus, we can no longer put  $\partial/\partial y = 0$  (or  $k_y = 0$  (or  $k_y = 0$ ) by symmetry. We have to repeat the linear stability analysis of Secs. II–V with  $k_y \neq 0$  and with the magnetic-field dependence of  $\phi_j$  different from that of Eq. (2.2):

$$\phi_j = \phi_j(H=0) - \chi_a H^2 \frac{\partial R}{\partial z}. \quad (\text{B1})$$

The heat-conduction state is unchanged and given by (3.3). The algebraic system of equations for linear deviations from the heat-conduction state is as given by Eq. (4.2) ( $A = 0 = g_{\perp} = g_{\parallel}$  for smectics *A*), except for the following changes:

$$\begin{aligned} i\omega v + \hat{\mu}_y^2 v + ik_y \frac{p}{\rho_0} + (\mu_3 - \mu_2)k_y k_x w &= 0, \\ \hat{\chi}^2 &= \chi_{11}k_{\perp}^2 - \chi_a H^2 k_x^2 + \bar{K}(k_x^2 + k_y^2), \\ k_{\parallel}^2 &= k_x^2, \\ k_{\perp}^2 &= k_x^2 + k_y^2. \end{aligned} \quad (\text{B2})$$

We have neglected thereby, the possible splitting of the transverse components of the material parameters, e.g.,  $\kappa_{\perp}$  into  $x$  and  $y$  components, the difference between which, e.g.,  $\kappa_x - \kappa_y$  would be proportional to  $H^2$ .

The threshold conditions for the two instabilities are the same as given by (5.1) and (5.2) if one replaces there  $\hat{\chi}^2$ ,  $k_{\parallel}^2$ ,  $k_{\perp}^2$  by their new expressions (B2). The main difference to the case of a stabilizing magnetic field now shows up in the magnetic-field dependence of the critical transverse wave vectors. For fields  $H < 10^3$  and  $10^2$  G for the stationary and oscillatory instability, respectively, there is no field dependence at all. Thus,  $k_x^c$  and  $k_y^c$  are equivalent by symmetry and without loss of generality  $k_y^c = 0$ ,  $k_x^c = k_{\perp}^c$  with (for STI),  $(k_{\perp}^c)^2 = (\chi_{11}\kappa_{\perp}d^2/2\kappa_{\perp}\pi^2\bar{K})^{1/3}\pi^2/d^2 \approx 7 \times 10^3 \times (\pi^2/d^2)$  [cf. (5.14)] and (for OSI),  $(k_{\perp}^c)^2 \approx \frac{1}{2}\pi^2/d^2$ .

If  $H$  cannot be neglected in the formulas (5.1) and (5.2) and (B2), i.e., if  $H > 10^3$  G for STI and  $> 10^2$  G for OSI, the transverse wave vectors  $k_x$  and  $k_y$  are clearly distinguishable. The minimum of  $\theta$  is obtained for  $k_y^c = 0$  and

$$(k_x^c)^2 = \frac{\chi_a H^2}{2\bar{K}} \quad (\text{STI}) \quad (\text{B3})$$

and

$$(k_x^c)^2 = \frac{\chi_a H^2}{2(\nu_3 \kappa_1 + \bar{K})} \quad (\text{OSI}).$$

Therefore,  $(k_x^c)^2$  increases, if the destabilizing field increases. This is easily understood, since the elastic energy is lowered in that case, which reflects in undulations of the layers on a shorter length scale. For  $(k_y^c)^2 = 0$  the two-dimensional convection rolls (cf. Secs. VB and VC) are still

eigenfunctions at the threshold.

By the strong magnetic-field dependence of  $(k_x^c)^2$  the numerical values of the critical temperature gradients  $\theta_c^{\text{STI}}$  and  $\theta_c^{\text{OSI}}$  will also depend heavily on  $H$ . For small  $H$ ,  $|\theta_c^{\text{STI}}|$  and  $|\theta_c^{\text{OSI}}|$  are outside any practical range. Only for magnetic fields, for which  $\chi_{11} \pi^2/d^2 + \bar{K} (k_x^c)^4 \approx \chi_a H^2 (k_x^c)^2$ , will there be small enough temperature gradients  $\theta_c$ . That regime is obtained for  $H \approx 10$  kG. If  $H$  is chosen so that  $\hat{\chi}^2 = 0$  exactly, then  $\theta_c^{\text{STI}} = 0$  and no longer describes a convective instability, but the well-known Helfrich-Hurault transition<sup>28,32,33</sup> (since  $T = 0$ ,  $w = v = u = 0, R \neq 0$ ). For the oscillatory instability  $\hat{\chi}^2 \approx 0$  ( $H \approx 10$  kG) means that

$$\theta_c^{\text{OSI}} = -\frac{\pi^2}{\alpha_{gE}} \frac{\chi_a H^2}{2d} \left( \frac{\kappa_{11}}{2\kappa_1} + \frac{\nu_1 + \nu_2 - \nu_3}{\nu_3} \right) \approx -4 \text{ K}.$$

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