Thermoconvective effect in a nematic liquid crystal

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We consider the two-dimensional problem of the onset of a convective instability in a temperature-gradient field (the thermoconvective effect) in a nematic liquid crystal (NLC) whose director **n** is inclined to the surface. At an arbitrary orientation of the vector **n**, a relation is obtained for the threshold temperature gradient of the stationary $\beta_c(\mathbf{q},\varphi)$ and oscillatory $\beta_o(\mathbf{q},\varphi)$ convection, where **q** is the wave vector of the modulated structure and φ is the inclination angle of the director to the NLC boundary. It is shown that the $\beta_c(\mathbf{q},\omega)$ spectrum has singular discontinuities that separate the regions $\beta_c>0$ and $\beta_c<0$, and a classification of these discontinuities is presented. A numerical calculation for MBBA has shown that a limiting angle $\varphi_0\approx 75^\circ$ exists such that stationary convection is produced at a minimum gradient $|\beta_c|$ by heating from below when the director orientation is $0^\circ\leqslant\varphi<\varphi_0$ and by heating from above at $\varphi_0<\varphi\leqslant 90^\circ$.

INTRODUCTION

The hydrodynamic instability observed in nematic liquid crystals (NLC) when a cell with the liquid crystal is placed in a temperature-gradient field was described with the aid of an anisotropic mechanism based on the Helfrich-Carr approach. The theoretical investigations dealt mainly with planarly and homeotropically oriented NLC, owing to the relative simplicity of the initial models. It was noted even in the early papers[1–3] that an important role is played by the boundary conditions on the director orientation, whereby the boundaries of the NLC are not equivalent. An expression was obtained in Ref. 3 for the spectrum of the threshold gradient $\beta(\mathbf{q})$ at $\varphi=0^\circ$ and 90°; this expression had a simple pole $\mathbf{q}\neq 0$, a fact not discussed further. The existence of positive and negative branches of $\beta(\mathbf{q})$ becomes particularly interesting if account is taken of the oscillatory instability connected with the development of inverse bifurcation.[4–6] A transition from planar (p) to homeotropic (h) orientation is accompanied by a change in the pole position from $\mathbf{q}_p$ to $\mathbf{q}_h\sim 10\mathbf{q}_p$; the absolute minima of the threshold of gradients β_p and β_h become comparable in magnitude. These last circumstances lead us to expect in the intermediate region of the director-orientation angles a more complete picture of the thermal convection, including several simple or degenerate poles $\mathbf{q}_i\neq 0$ in the $\beta(\mathbf{q})$ spectrum.

The purpose of the present paper is to find the spectrum of the threshold temprature gradient of the stationary $\beta_c(\mathbf{q},\varphi)$ and the oscillatory $\beta_o(\mathbf{q},\varphi)$ convection in NLC with oblique orientation of the director at the NLC boundaries.

1. FORMULATION OF PROBLEM

We consider in this paper thermal convection in NLC within the framework of the Boussinesque approximation,[7] neglecting terms of second and higher orders of smallness relative to the variations of the temperature and hydrodynamic variables. In addition, we confine ourselves to the single-constant approximation of the Frank elasticity theory, which preserves the main features of the hydrodynamics of an obliquely oriented NLC. The two-dimensional model of thermal convection turns out in this case to be complete enough[3] for the investigation of the Benard-Rayleigh problem in NLC, in contrast to the one-dimensional model.[2–5].

Consider an NLC with a direction $\mathbf{n}_0$, making an angle φ with the boundary of the NLC layer. The nonstationary flow in the NLC is described by a system of four differential equations for four unknown functions: V_x and V_z, the components of the velocity vector **V** of the points of the medium; the angle $\theta(x,z)$ of deflection of the director **n** from its unperturbed position n_0; the deviation $T(x,z)$ of the temperature in the NLC layer from a homogeneous distribution β_z, where $\beta=\Delta T/L$ is the temperature gradient and L is the thickness of the NLC layer.

The continuity equation for an incompressible liquid is

$$\hat{p}_x V_x+\hat{p}_z V_z=0. \tag{1.1}$$

The heat-conduction equation is

$$\rho C\beta V_z+[\varkappa_\perp(\hat{p}_x^2+\hat{p}_z^2)+\varkappa_a(\hat{p}_x\cos\varphi+\hat{p}_z\sin\varphi)^2]T =\varkappa_a\beta(\hat{p}_x\cos 2\varphi+\hat{p}_z\sin 2\varphi)\theta+\rho C\hat{p}_t T. \tag{1.2}$$

The Navier-Stokes equation in an anisotropic NLC is

$$\rho\hat{p}_t(\hat{p}_x V_z-\hat{p}_z V_x)-\hat{p}_t\left[\frac{-\gamma_1+\gamma_2\cos 2\varphi}{2}\hat{p}_x^2+\gamma_2\sin 2\varphi\hat{p}_x\hat{p}_z-\frac{\gamma_1+\gamma_2\cos 2\varphi}{2}\hat{p}_z^2\right]\theta=(B_3\hat{p}_x^3+B_4\hat{p}_x\hat{p}_z^2-B_5\hat{p}_x^2\hat{p}_z-B_6\hat{p}_z^3)V_z+\rho g\alpha\hat{p}_x T+(B_7\hat{p}_x^3+B_8\hat{p}_x\hat{p}_z^2-B_1\hat{p}_x^2\hat{p}_z-B_2\hat{p}_z^3)V_x. \tag{1.3}$$

The equation of motion of the director, neglecting the small specific moment of inertia J of the liquid ($J\ll\rho L^2$) is:

$$K(\hat{p}_x^2+\hat{p}_z^2)\theta=\gamma_1\hat{p}_t\theta+\frac{-\gamma_1+\gamma_2\cos 2\varphi}{2}\times\hat{p}_x V_z+\gamma_2\sin 2\varphi\hat{p}_z V_z+\frac{\gamma_1+\gamma_2\cos 2\varphi}{2}\hat{p}_z V_x \tag{1.4}$$

The following notation is used in (1.1)–(1.4): $\hat{p}_t, \hat{p}_x, \hat{p}_z$ are the differential operators $\hat{p}_t = \partial/\partial t$, $\hat{p}_x = \partial/\partial x$, $\hat{p}_z = \partial/\partial z$, algebraic actions on which being understood in symbolic sense, e.g., $\hat{p}_x^2 = \partial^2/\partial x^2$ etc.; ρ is the NLC density; g is the free-fall acceleration; α is the coefficient of thermal expansion of the NLC; $\varkappa_a = \varkappa_\parallel - \varkappa_\perp$, where $\varkappa_\parallel$ and $\varkappa_\perp$ are the principal values of the diagonalized NLC thermal-conduction tensor, C is the isobaric specific heat, K is the Frank elastic constant in the single-constant theory, γ_i and B_i are linear combinations of the Leslie viscosity coefficients α_i, defined as follows:

$$\begin{gathered}
\gamma_1=\alpha_3-\alpha_2, \quad \gamma_2=\alpha_6-\alpha_5=\alpha_3+\alpha_2,\\
B_1=\alpha_1 n_x^2(n_x^2-2n_z^2)+{}^1/_2[\alpha_4+n_x^2(\alpha_3+\alpha_6+2\alpha_5)-n_z^2(\alpha_2+\alpha_5)],\\
B_2=\alpha_1 n_x^2 n_z^2+{}^1/_2[\alpha_4+n_x^2(\alpha_3+\alpha_6)+n_z^2(\alpha_5-\alpha_2)],\\
B_3=\alpha_1 n_x^2 n_z^2+{}^1/_2[\alpha_4+n_z^2(\alpha_3+\alpha_6)+n_x^2(\alpha_5-\alpha_2)],\\
B_4=\alpha_1 n_z^2(n_z^2-2n_x^2)+{}^1/_2[\alpha_4+n_z^2(\alpha_3+\alpha_6+2\alpha_5)-n_x^2(\alpha_2+\alpha_5)],\\
B_5=n_x n_z[\alpha_1 n_x^2+{}^1/_2(\alpha_2+\alpha_3+\alpha_5+\alpha_6)],\\
B_6=n_x n_z[\alpha_1(n_z^2-2n_x^2)+{}^1/_2(\alpha_2+\alpha_3-3\alpha_5+\alpha_6)],\\
B_7=n_x n_z[\alpha_1(n_x^2-2n_z^2)+{}^1/_2(\alpha_2+\alpha_3-3\alpha_5+\alpha_6)],\\
B_8=n_x n_z[\alpha_1 n_z^2+{}^1/_2(\alpha_2+\alpha_3+\alpha_5+\alpha_6)],\\
n_x=\cos\varphi, \quad n_z=\sin\varphi.
\end{gathered} \tag{1.5}$$

The coefficients B_i are a generalization of the known Miesowicz viscosity coefficients[8] to include the case of an obliquely oriented NLC.

For the solution to be unique, Eqs. (1.1)–(1.4) must be supplemented by boundary conditions for the functions T, θ, V_x, V_z on the NLC boundary:

$$T=0, \quad \theta=0, \quad V_x=V_z=0 \quad \text{at} \quad z=\pm L/2. \tag{1.6}$$

2. THRESHOLD GRADIENT OF THERMOCONVECTIVE EFFECT

Solution of the system (1.1)–(1.4) with the aid of Fourier transformation of the functions T, θ, V_x, V_z leads to a system of four homogeneous linear algebraic equations, from which we can obtain a characteristic third-degree equation for the perturbation decremtnt.

For convective instability to occur it is necssary that at least one of the solutions λ_i of the characteristic equations have Re $\lambda \geqslant 0$. With the aid of the Routh-Hurwitz theorem we can obtain an expression for the threshold temperature gradient of the stationary convection (Im $\lambda = 0$).

$$\beta_c(\mathbf{q},\varphi)=\frac{m^4}{\rho g\alpha}\frac{K(q_x^2+q_z^2)\Gamma_1\Gamma_2}{K(q_x^2+q_z^2)\rho C q_x^2+{}^1/_2 q_x\Gamma_3\Gamma_4} \tag{2.1}$$

and of the oscillatory convection (Im $\lambda = \omega \neq 0$)

$$\beta_o(\mathbf{q},\varphi)=\frac{m^4}{\rho g\alpha\gamma_1}\frac{M(P+Q)+EP}{q_x^2 M-q_x S}; \tag{2.2}$$

the threshold frequency of the oscillatory convection being

$$\omega=(D_1/D_2)^{1/2}. \tag{2.3}$$

In these equations q_x and q_z are the components of the wave vector $\mathbf{q}$, m is the number of the Fourier representation,

$$\begin{gathered}
\Gamma_1=B_2 q_z^4-(B_6+B_8)q_z^3 q_x+(B_1+B_4)q_z^2 q_x^2\\
-(B_5+B_7)q_z q_x^3+B_3 q_x^4,\\
\Gamma_2=\varkappa_\perp(q_x^2+q_z^2)+\varkappa_a(q_x\cos\varphi+q_z\sin\varphi)^2,\\
\Gamma_3=(\gamma_1+\gamma_2\cos 2\varphi)q_z^2-2\gamma_2 q_x q_z\sin 2\varphi+(\gamma_1-\gamma_2\cos 2\varphi)q_x^2,\\
\Gamma_4=\varkappa_a(q_x\cos 2\varphi+q_z\sin 2\varphi),\\
D_1=\gamma_1[m^4\Gamma_1\Gamma_2-\rho^2 C\alpha g\beta q_x^2]\\
+Km^4\rho(q_x^2+q_z^2)[C\Gamma_1+(q_x^2+q_z^2)\Gamma_2]-{}^1/_4 m^4\Gamma_2\Gamma_3^2,\\
D_2=\gamma_1\rho^2 C(q_x^2+q_z^2),\\
M=\gamma_1[\Gamma_1+\rho(q_x^2+q_z^2)\Gamma_2]-{}^1/_4\Gamma_3^2,\\
E=K\rho(q_x^2+q_z^2)^2, \quad Q=\gamma_1\Gamma_1\Gamma_2,\\
P=K(q_x^2+q_z^2)[\Gamma_1+\rho(q_x^2+q_z^2)\Gamma_2]-{}^1/_4\Gamma_2\Gamma_3^2,\\
S={}^1/_2\rho(q_x^2+q_z^2)\Gamma_3\Gamma_4.
\end{gathered}$$

To stabilize the initial orientation of the director, a magnetic field **H** is sometimes applied parallel to **n**.[2,5] This increases the threshold in accord with the law $\beta \sim [1+(H/H_0)^2]$, where H_0 is a field that coincides with the threshold field in the Freedericksz effect in a magnetic field. This dependence can be easily obtained also in the case of an obliquely oriented NLC, by introducing in Eq. (1.4) a term $\mu_a H^2\theta$ connected with the variation of the free energy of the NLC in a magnetic field, where μ_a is the anisotropy of the NLC magnetic susceptibility.

The wave-vector component q_z is usually close to π/L. It can be found more accurately by numeric methods[3] from the solution of the system (1.6).

It is therefore easy to ascertain that (2.1) and (2.2) are the conditions for the critical Rayleigh numbers $R = (\rho g\alpha\beta/\nu\chi)L^4$ (Ref. 7), where $\nu = \alpha_4/\rho$ and $\chi = \varkappa/\rho C$ are respectively the coefficients of the kinematic viscosity and of the thermal diffusivity of the NLC.

3. SINGULARITIES OF THE THRESHOLD GRADIENT OF STATIONARY CONVECTION

We introduce the variable $y = q_x/q_z$ and transform in (2.1) to $\beta_c(y,\varphi)$:

$$q_z^4\beta_c(\mathbf{q},\varphi)=\beta_c(y,\varphi)=F(y,\varphi)/U(y,\varphi), \tag{3.1}$$

where $F(y,\varphi)$ $U(y,\varphi)$ are polynomials of 8-th and 4-th degree in y, respectively.

A feature of the $\beta_c(y,\varphi)$ dependence that distinguishes it from the analogous one for an incompressible liquid[7] is the presence of nonzero poles y_i, which are due to simultaneous anisotropy of the thermal conductivity and of the shear viscosity in the NLC. This leads to the presence in the $\beta_c(y,\varphi)$ spectrum of singular discontinuities that separate the regions with $\beta_c > 0$ and $\beta_c < 0$ and, as a consequence, to the existence of stationary thermal convection in the NLC layer when heated both from above and from below.

We turn now to the distribution of the positive zeros of the polynomial $U(y,\varphi)$, which are the poles of $\beta_c(y,\varphi)$. To this end we use the Descartes theorem[9] on the number of sign reversals in the sequence of coefficients of the polynomial $U(y,\varphi)$.

Analaysis shows that there exist four regions of the director-orientation angle, and the positive zeros of the polynomial U are located in them in the following manner:

1. $0° \leqslant \varphi < \arctan(\alpha_3/\alpha_2)^{1/2} \equiv \varphi_{32}$ ($\varphi_{32} \approx 7°$ for **MBBA**).

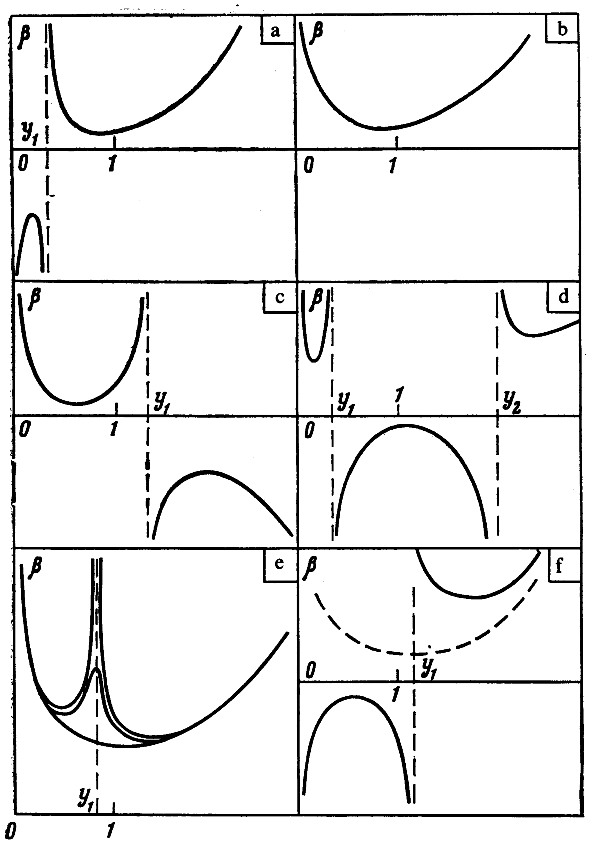

FIG. 1. Plots of $\beta(y)$ for convective instability in NLC under various conditions of limiting orientation of the director (β_c—continuous curve, β_o—dashed).

There is one nondegenerate zero $y_1 \ll 1$ (Fig. 1a).

2. $\varphi_{32} \leqslant \varphi \leqslant 45^\circ$ [1].
   There are no zeros (Fig. 1b).
3. $45^\circ < \varphi \leqslant \arctan(\alpha_2/\alpha_3)^{1/2} \equiv \varphi_{23}$ ($\varphi_{23} \approx 83^\circ$ for MBBA).
   There is one nondegenerate zero $y_1 \gtrsim 1$ (Fig. 1c).

4a. $\varphi_{23} < \varphi < 90^\circ$, $\chi_a > 0$.
   There are two nondegenerate zeros $y_1 \ll 1$, $y_2 \gg 1$ (Fig. 1d).

4b. $\varphi_{23} < \varphi < 90^\circ$, $\chi_a < 0$.
   The same as in case 3.

It is known[10] that in the temperature interval in which the liquid-crystal phase exists the Leslie coefficient α_3 can become small enough and can even vanish. This possibility leads to the appearance of a dimensionless parameter $\varepsilon = |K/\alpha_3\chi_a|$ in the problem. The number N of positive zeros of the polynomial U is listed in Tables I and II, for p- and h-orientation of the director, as a function of the magnitude of ε and of the sign of the anisotropy of the thermal diffusivity χ_a.

TABLE I.

p- orientation

N	$\varepsilon<1$	$\varepsilon>1$
$\chi_a>0$	1	0
$\chi_a<0$	1	1

TABLE II.

h- orientation

N	$\varepsilon<1$	$\varepsilon>1$
$\chi_a>0$	1	1
$\chi_a<0$	1	0

Special attention must be paid to the rare[2)] but physically not impossible condition.

$$\alpha_3=0,\quad \chi_a<0, \tag{3.2}$$

which presents an example of the appearance in the $\beta_c(y)$ spectrum of a doubly degenerate pole at an orientation φ_0 close to planar ($0<\varphi_0<[K/5\alpha_2\chi_a]^{1/2}$). With further increase of the orientation angle ($\varphi_0<\varphi<[K/5\alpha_2\chi_a]^{1/2}$ there appears in the $\beta_c(y)$ a characteristic potential barrier (Fig. 1e) that becomes rapidly smoothed out and imparts to $\beta_c(y)$ the known form (Fig. 1b).

4. OSCILLATORY CONVECTION

It is known[7] that in the classical Benard-Rayleigh problem the vibrational perturbations of the hydrodynamic and thermal functions do not cause convective instability in a layer of isotropic incompressible liquid. In an NLC, on the contrary, oscillatory convective instability can develop because of the stabilizing orientation-relaxation stabilizing mechanism that is typical of liquid crystals and has a characteristic relaxation time $\tau_0 = (\gamma_1/K)(L/\pi)^2$. A physical explanation of the oscillatory convect was proposed in Ref. 4, using a comparison of τ_0 with the time $\tau_t = \chi^{-1}(L/\pi)^2$ of the destabilizing thermal relaxation. The condition for the development of oscillatory convection is of the form

$$\tau_0/\tau_t=\gamma_1\chi/K>1, \tag{4.1}$$

and the characteristic frequency of the convection is

$$\omega\approx\tau_t^{-1}=(\pi/L)^2\chi. \tag{4.2}$$

For MBBA $\tau_0/\tau_t \approx 10^3$ and the expected oscillation frequency for a 5-mm layer is $\omega \approx 0.04$ Hz, as confirmed by experiment.[5]

For the known NLC that satisfy the condition (4.1), the spectrum $\beta_o(y,\varphi)$, in contrast to $\beta_c(y,\varphi)$, has no poles in the region $0^\circ \leqslant \varphi \leqslant 90^\circ$, $0<y$. At any orientation of the director at the boundary, an oscillatory instability will be observed when the NLC layer is heated from below. The instability occurs physically if the corresponding stationary convection threshold $\beta_c > \beta_o$ (Fig. 1f, dashed curve).

5. NUMERICAL CALCULATION

We present in this section the results of a numerical calculation of the threshold temperature gradients $\beta_c(y,\varphi)$ and $\beta_o(y,\varphi)$ for the thoroughly investigated liquid crystal MBBA at $T_0 = 298$ K with the following parameters 3, 8, 12, 13: $\alpha_1 = 6\cdot10^{-3}$ Pa·sec, $\alpha_2 = -77\cdot10^{-3}$ Pa·sec, $\alpha_3 = -1.2\cdot10^{-3}$ Pa·sec, $\alpha_4 = 83\cdot10^{-3}$ Pa·sec, $\alpha_5 = 46\cdot10^{-3}$ Pa·sec, $\alpha_6 = -34\cdot10^{-3}$ Pa·sec; $K = 6.5\cdot10^{-12}$ N; $\chi_\parallel = 1.54\cdot10^{-7}$ m²/sec; $\chi_\perp = 0.93\cdot10^{-7}$ m²/sec; $\rho = 1088$ kg/m³; $\alpha = 4.92\cdot10^{-4}$ K^{-1}.

In (2.1) and (2.2), q_{z2} is assumed equal to π/L. For com-

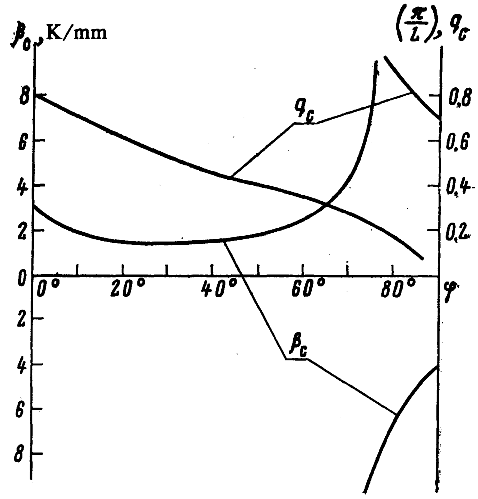

FIG. 2. Dependence of the threshold temperature gradient $\beta_c(\varphi)$ and of the wave vector $q_c(\varphi)$ of the periodic structure on the angle φ in MBBA.

parison with the experimental data,[2,5] we used $L_c = 1$ mm for the calculation of $\beta_c(y, \varphi)$ and $L_0 = 5$ mm for $\beta_o(y, \varphi)$, respectively. The calculation was performed for $m = 1$. As expected, $\beta_c(y, 0°)$ takes, in accord with the results of the preceding sections, the form shown in Fig. 1a with a minimum of the positive branch $\beta_p = 3$ K/mm at $y = 0.8$ and with a maximum of the negative branch -3×10^2 K/mm at $y = 0.08$. For homeotropic orientation (Fig. 1f, solid curve) the maximum of the negative branch $\beta_h = -4$ K/mm occurs at $y = 0.7$, and the minimum of the positive branch $\beta' = 7\times 10^4$ K/mm occurs at $y = 10$. The values presented for β_p and β_h are in good agreement with the known experimental data.[2] At the same time, the anomalously large threshold gradient β' in the sideband explains why it was not observed in experiments[5] on oscillatory instability.

We note that since $\beta \propto L^{-4}$, the negative branch of $\beta(y, 0°)$ could be observed in experiment when the cell thickness L_c was increased to 3 mm, but the resultant modulated stationary structure would then have a period of 3 cm.

The calculation of the oscillatory instability in MBBA shows that only in the vicinity of the homeotropic orientation ($89° < \varphi \leqslant 90°$) does the oscillatory branch play a dominant role ($\beta_o < \beta_c$, Fig. 1f) when the NLC layer is heated from below, the minimum $\beta_o(y, 90°) = 1.5$ K/mm is then observed at $y = 1$. The frequency corresponding to the oscillation threshold is $\omega = 0.05$ Hz. The values cited agree well with the data of Ref. 5, namely $y = 1$, $\beta_o = 1.2$ K/mm, and $\omega = 0.06$ Hz.

Figure 2 shows plots of the absolute extremum of $\beta_c(\varphi)$ at the extremum point y_c and of the corresponding wave vector $q_c = y_c\pi/L$ against the angle φ. In the angle range 20°–40° the $\beta_c(\varphi)$ curve has a weakly pronounced minimum smaller by a factor of two than the threshold gradient in the case of p orientation.

We note that thermal convection with minimum external energy loss ($|\beta_c|$ is a minimum) is produced in MBBA by heating from below for a limiting orientation of the director $0° \leqslant \varphi \lesssim 75°$ and by heating from above at $75° \lesssim \varphi \leqslant 90°$. The proximity of the limiting angle 75° to the homeotropic orientation is due to the large value of the ratio α_2/α_3.

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[1)]The exact value of the angle differs from $\pi/4$ by $|K/\alpha_2\chi_a|$, which amounts in practice to 10^{-2}–10^{-3}.

[2)]The known NLC have $\chi_a > 0$ (Refs. 5, 12), although this is not required by thermodynamic considerations.[10] See Ref. 4 as well as the papers[1,11] on NLC calorimetry, in which the possibility of $\chi_a < 0$ is admitted.

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