Molecular Crystals and Liquid Crystals
Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/gmcl18

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Version of record first published: 24 Sep 2006.

To cite this article: E. D. Belotskii , B. I. Lev & P. M. Tomchuk (1991): Effects of Incommensurability in the Nematic Liquid Crystals with Induced Gyrotropy, Molecular Crystals and Liquid Crystals, 209:1, 179-182

To link to this article: http://dx.doi.org/10.1080/00268949108036192

Mol. Cryst. Liq. Cryst., 1991, Vol. 209, pp. 179–182
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Printed in the United States of America

Effects of Incommensurability in the Nematic Liquid Crystals with Induced Gyrotropy

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(Received September 17, 1990)

In this paper we predict theoretically the existence of two incommensurable periods in the distribution of the director in a nematic liquid crystal, as the gyrotropy induced by chiral admixture.

The difference between proper and induced cholesterics is determined by the mechanics of formation of spiral supra-molecular structure. The distribution of individual chiral molecules in mesophase with orientation ordering, is determined by the dissymmetry and anharmonicity of the interaction forces.

The orientation order in a nematic liquid with induced gyrotropy is locally disturbed by chiral admixture. The large dimensions of the introduced molecules, or their existence in the matrix only as large complexes, leads to the deformation of orientation order at considerable distances. The distance at which the deformation of orientation ordering is manifest depends on the interaction energy of the molecules of a liquid crystal and the admixture. The superposition of local deformations, which was investigated earlier in the single-constant approximation,[1] leads to the appearance of spiral supra-molecular structure. The wave vector of such a structure is $g_0 = 4\pi\beta c$, where c is the concentration of the admixture and β is determined by the interaction between the molecular liquid crystal and the admixture.

When the variation of the elastic constant is taken into account, the superposition of local deformations leads to the distortion of the ideal spiral supra molecular structure, as we demonstrate further on. More specifically, it leads to the appearance of two incommensurable periods in the distribution of the director. This state is energetically advantageous and corresponds to a minimum of the free energy, provided that the initial energy introduced by the admixtures is treated in the single constant approximation.

The theoretical investigation is based on the free energy of a nematic liquid crystal:

$$F_d = \frac{1}{2}\{K_{33}(\mathrm{div}\vec{n})^2 + K_{22}(\vec{n} \cdot rot\vec{n})^2 + K_{33}[\vec{n} \times rot\vec{n}]^2\} \qquad (1)$$

where K_{ii} are Frank's elastic constants.

The director distribution has the form:

$$\vec{n} = \{\cos\theta \cos\varphi, \cos\theta \sin\varphi, \sin\theta\} \tag{2}$$

where $\theta(z)$, $\varphi(z)$ are functions of the coordinate z in the direction of the spiral's axis.

The free energy expressed in terms of the angle variables can be written as follows:

$$F_d = \frac{1}{2}\left\{(K_{11}\cos^2\theta + K_{33}\sin^2\theta)\left(\frac{d\theta}{dz}\right)^2 + (K_{22}\cos^2\theta + K_{33}\sin^2\theta)\cos^2\theta\left(\frac{d\varphi}{dz}\right)^2\right\} \tag{3}$$

We introduce the notation $\alpha \equiv 1 - K_{11}/K_{33}$, $\beta \equiv 1 - K_{22}/K_{33}$ and assume that $\varphi = qz$, where q is the wave vector of the detailed deformed spiral structure, which can be easily obtained from the minimum of the free energy.

The free energy, in this notation has the form:

$$F_d = \frac{K_{33}}{2}\left\{(1 - \alpha\cos^2\theta)\left(\frac{d\theta}{dz}\right)^2 + q^2(1 - \beta\cos^2\theta)\cos^2\theta\right\} \tag{4}$$

and in principle, is similar to the free energy, which is used in investigations of effects of incommensurability.[2,3]

We assume, at first, that all the energy of deformation, which is introduced by the admixtures and induces the spiral structure is completely taken into account in the single constant approximation. This permits us to write that $F_d = K_{33}/2\, q_0^2$. From this it follows immediately that,

$$\left(\frac{d\theta}{dz}\right)^2 = q_0^2 - q^2 + q^2\sin^2\theta \tag{5}$$

When $\alpha = \beta = 0$, $q = q_0$ and $\theta = 0$. When $q \neq q_0$ the solution of this equation is the solution of the nonlineal equation Sin-Gordon:

$$\theta = am\left[-4K\left(\frac{\sqrt{a^2 - 1}}{a}\right) z/l\right] \tag{6}$$

where $am\, f(z)$ is ellipitical Jakobi amplitude, K is the ellipitical integral of first

kind. The new parameter a is related, to the wave vector q by the condition $a^2 = q^2/q^2 - q_0^2$. The solution (6) corresponds to a rotation of the angle θ by an angle divisible by 2π with period:

$$l = \frac{4K\left(\frac{\sqrt{a^2 - 1}}{a}\right)}{q} \tag{7}$$

After substituting Equation (5) into Equation (4) and evaluating the integral $\int_0^L F_d\, dz$, where L is thickness liquid crystal, which can be written in the form $F = L/l \int_0^l F_d\, dz$, we obtain, for the normalized free energy, $f = 2F/LK_{33}$ the expression involving the parameter

$$f = q_0^2 \left\{1 - \frac{\alpha}{3}\left[\frac{a^2 + 1}{a^2 - 1} e\left(\frac{\sqrt{a^2 - 1}}{a}\right) - \frac{1}{a^2}\left(1 + \frac{a^2 + 1}{a^2 - 1}\right)\right]\right.$$
$$\left. + \frac{\beta}{3}\left[\frac{1}{a^2}\left(\frac{4a^2 - 2}{a^2 - 1} + \frac{3a^4 - 5a^2 + 2}{a^2 - 1}\right) - \frac{4a^2 - 2}{a^2 - 1} e\left(\frac{\sqrt{a^2 - 1}}{a}\right)\right]\right\} \tag{8}$$

where $e(\sqrt{a^2 - 1}/a) = E/K$ is determined by elliptical integrals of the second and first kind with argument $\sqrt{a^2 - 1}/a$. We introduce a new variable $m \equiv \sqrt{a^2 - 1}/a$, and rewrite the above formulas in terms of this variable, whereupon:

$$q = \frac{q_0}{m}, \qquad l = \frac{4m\, K(m)}{q_0} \tag{9}$$

and the free energy can be written as follows:

$$f = q_0^2 \left\{1 - \frac{\alpha}{3}\left[\frac{2}{m^2}(e(m) - 1) - (e(m) - 1) + 1\right]\right.$$
$$\left. + \frac{\beta}{3}\left[\frac{2}{m^2}(e(m) - 1) + 2(e(m) - 1) + 1\right]\right\} \tag{10}$$

The equation for the m is determined by minimizing the free energy. The asymptotic behaviour of $e(m)$, namely that $m \to 0$ $e(m) \to 1 - m^2/2$ and $m \to 1$, $e(m) \to 1/2(1 - m^2)$, leads to the following solution for m:

$$m^2 \simeq \frac{2(\beta - \alpha)}{\alpha + 2\beta} \tag{11}$$

The condition $m < 1$ determines the condition when this structure is realized. This condition is very simple, being $\alpha > 0$ or $K_{33} \to K_{11}$, and holds for majority liquid crystals. For $K_{33} \to K_{11}$ $m \to 1$, $q \simeq q_0$ only one period of the ideal spiral

structure is realized. With the asymptotic condition $m \to 1$, the free energy at the minimum, has the $f = q_0^2 (1 - \beta)$ and the initial free energy $F_d = K_{22}/2\, q_0^2$. In other words, equating the initial free energy to this value, we obtain the solution (II) without minimization. This corresponds to the fact that the energy, which is contributed by the admixtures, is determined by the energy of rotation. Because there is a redistribution of energy among other deformations, their superposition leads to distortion of the ideal supra-molecular structure. The solution obtained above corresponds to a periodic distribution of static solutions in the orientation of the director, where retaining the spiral supra-molecular structure only with the new period. The relationship of the period of distribution of orientation solitons to the spiral stip $D = 2\pi/q$ has the form:

$$\frac{l}{P} = \frac{2K(m)}{\pi} \tag{12}$$

and can be evaluated at any value of the elastic constants.

In our opinion, this was first observed experimentally in the investigation of References 4 and 5. A two-level laser generation, with a distributed reverse connection based on nematic liquid crystals with induced gyrotropy was demonstrated in these works.

Even principle of operation of a laser with a distributed reverse connection, generation is possible only with periodic structure. Its Bragg frequency falls into the region of increasing generating admixture. Arbitrary changes of an aperiodic character, such as the spiral stip gradient, distribution in thickness of the spiral axis, the heating of the medium by radiation results only in the thickening of the line of generation. The appearance of two lines at the edges of the region of selective reflection is indicative of the presence of a new modulation of the spiral structure.[4]

There is good agreement between the theoretical and experimental values of l/P. The difference between the approach of Reference 4 and the present one rests in the selection of the starting structure. In Reference 4 the starting point is taken to be the structure of the induced cholesteric. In the present work, the starting point is the nematic, and the approximate treatment of the energy contributed by the admixture is motivated better.

In our opinion, the structure obtained in the present work corresponds to the real situation in nematic liquid crystals with induced gyrotropy. The effects incommensurability must be taken into account in fundamental investigations of such media.

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