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Stability conditions of a twist-bend nematic phase according to Barbero's theory

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The stability conditions of twist-bend nematics given by Barbero *et al.* [Phys. Rev. E **92**, 030501(R) (2015)] lead to doubtful conclusions when the influence of the magnetic field on the N_{TB} structure is analyzed. For this reason, the stability criteria have been redetermined in the present paper. The calculations have revealed that some of the conditions presented by Barbero and his co-workers are incorrect. It has been shown that the parameters b_oK_{33} and η must be positive to induce the formation of a stable twist-bend nematic phase. Furthermore, some additional criteria concerning the value of η have been derived. The phenomenon of the shift of the stability interval in the magnetic field has been analyzed in detail.

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I. INTRODUCTION

In the last years twist-bend nematics (N_{TB}) have become a popular subject of research. This unique liquid-crystalline phase was theoretically predicted in 1973 by Meyer [1] and then experimentally discovered in 2011 by Cestari and co-workers [2]. Twist-bend nematics are characterized by heliconical structure [3–5], in which the director **n** spontaneously precesses around a certain axis **t**, but the orientation of these unit vectors is not perpendicular—they form an acute angle θ, called a heliconical angle or a tilt angle. The spatial periodicity can be described with a parameter p called a pitch which is extremely small, about a few nanometers [6]. The coexistence of twist and bend deformations is necessary to fill the whole space [7]. The molecular arrangement in twist-bend nematics is seemingly similar to the structure of smectics C*, but in the case of the N_{TB} phase there is no positional order and the layers are not distinguishable [8]. Twist-bend nematics are usually formed by achiral flexible molecules (dimers, trimers, and tetramers) which have highly curved shape [9,10].

The characteristic parameters of a twist-bend nematic (such as the heliconical angle and the pitch) can be calculated only if the formula for the free-energy density of a liquid crystal is known. This formula is also essential to recognize the influence of external fields on the structure of the N_{TB} phase. Many papers have been devoted to these issues. The models of elastic properties of twist-bend nematics have been proposed by Dozov [3], Shamid [11], Parsouzi [12], Meyer [8], Virga [13], Barbero [14], Longa [15], Čopič [16], and Vaupotič [17]. These theories have been reviewed in Ref. [18].

II. INFLUENCE OF MAGNETIC FIELD

The motivation for the analysis of stability of the N_{TB} phase presented below is the article written by Zola *et al.* [19]. In this paper the authors apply Barbero's theory to the description of deformations occurring in twist-bend nematics when the magnetic field is directed along the helix axis. According to their considerations, the main field-induced effect is connected with the shift of stability interval towards lower values of parameter η measuring the strength of coupling between the usual nematic director **n** and the helix axis **t**. Moreover, the authors of Ref. [19] present how the quantity $\sin^2\theta$ and the wave number $q = 2\pi/p$ depend on the magnetic-field intensity H:

$$\sin^2\theta = -\frac{b_0K_{33} \mp K_{22}q_0\sqrt{\frac{b_0K_{33}}{\eta+H^2\mu_0\Delta\chi}}}{K_{22}-K_{33}}, \tag{1}$$

$$q = \pm\sqrt{\frac{\eta+H^2\mu_0\Delta\chi}{b_0K_{33}}}, \tag{2}$$

where $b_0 = 1 + 2\nu_4/K_{33}$; $q_0 = \kappa_2/K_{22}$; K_{22} is the twist elastic constant; K_{33} is the bend elastic constant; μ_0 is the vacuum permeability; $\Delta\chi$ is the magnetic susceptibility anisotropy; and ν_4, κ_2 are other phenomenological elastic constants introduced by Barbero in his formula for the free-energy density. The upper signs are chosen when $q_0 > 0$; otherwise, the lower ones are suitable. The sign of q defines the handedness of the material.

Zola *et al.* [19] do not reveal the consequences resulting from Eqs. (1) and (2), so the corresponding analysis is performed in the following, starting from the stability criteria of twist-bend nematics given in Ref. [14]. For $q_0 > 0$ the conditions for the formation of the N_{TB} phase take the following form:

$$\begin{aligned} &K_{22} > K_{33},\\ &K_{22}q_0 > \sqrt{\eta b_oK_{33}},\\ &\eta > 0,\\ &b_oK_{33} > 0, \end{aligned} \tag{3}$$

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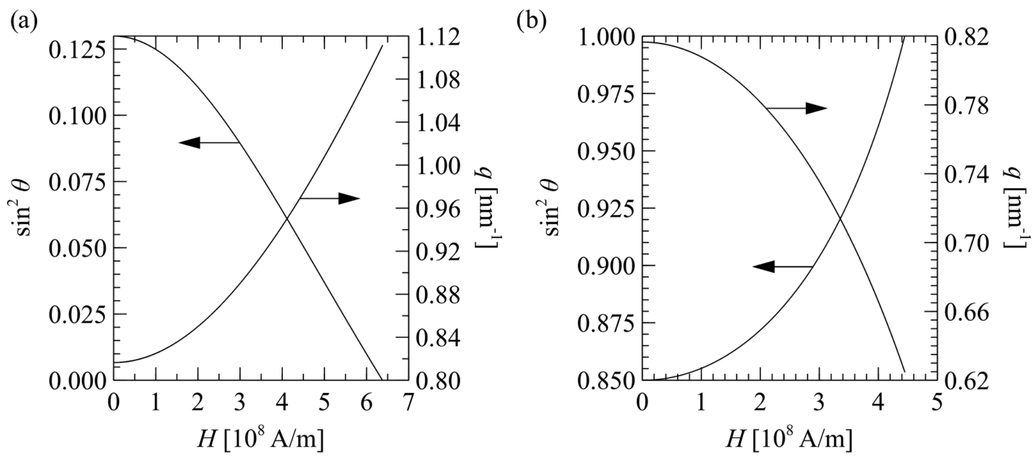

FIG. 1. Sine squared of tilt angle $\sin^2\theta$ and wave number q of twist-bend nematic phase as a function of magnetic field H, calculated from Eqs. (1) and (2). In (a) the material parameters satisfy conditions (3): $K_{22} = 3$ pN, $K_{33} = 0.5$ pN, $\nu_4 = 0.2$ pN ($b_0K_{33} = 0.9$ pN), $\eta = 6 \times 10^5$ N/m^2, $K_{22}q_0 = 0.001$ N/m, $\Delta\chi = 10^{-6}$. In (b) material parameters correspond to conditions (4): $K_{22} = 3$ pN, $K_{33} = 0.5$ pN, $\nu_4 = -0.7$ pN ($b_0K_{33} = -0.9$ pN), $\eta = -6 \times 10^5$ N/m^2, $K_{22}q_0 = 0.001$ N/m, $\Delta\chi = 10^{-6}$.

or

$$\begin{aligned} &K_{22} > K_{33}, \\ &K_{22}q_0 > \sqrt{\eta b_o K_{33}}, \\ &\eta < 0, \\ &b_o K_{33} < 0. \end{aligned} \tag{4}$$

The above inequalities correspond to $\sin^2\theta$ and q given by

$$\sin^2\theta = -\frac{b_0K_{33} - K_{22}q_0\sqrt{\frac{b_0K_{33}}{\eta}}}{K_{22} - K_{33}}, \tag{5}$$

$$q = \sqrt{\frac{\eta}{b_0K_{33}}}. \tag{6}$$

The further analysis is conducted for $\Delta\chi > 0$. When the formulas (1) and (2) are used with parameters satisfying the inequalities (3), then $\sin^2\theta$ decreases and q increases with the field intensity H. These effects are illustrated in Fig. 1(a) plotted for typical values of material parameters characterising a twist-bend nematic phase. In a very strong magnetic field, $\sin^2\theta$ reaches zero and the N_{TB}-N phase transition takes place. Such an effect is commonly described in the literature for an electric field (for example, by Meyer [8] and Pająk [15]). For this reason the analogous phenomenon occurring in a magnetic field could be expected; however, the necessary field strength (10^8 A/m) seems unreachable.

When the parameters satisfy the inequalities (4), the formulas (1) and (2) yield some surprising results, namely $\sin^2\theta$ increases and q decreases when H becomes larger, as shown in Fig. 1(b). For a certain value of the magnetic-field intensity, $\sin^2\theta$ becomes equal to unity. This could indicate that the external field induces a phase transition from the twist-bend nematic to the cholesteric structure. However, none of the papers mentions this phenomenon. For this reason, the derivation of the stability criteria of the N_{TB} phase as well as the expressions for $\sin^2\theta$ and q have been investigated in detail starting from Barbero's theory.

III. STABILITY CONDITIONS OF THE N_{TB} PHASE

According to Barbero's theory, the formula for the free-energy density of a twist-bend nematic can be written as

$$\begin{aligned} f(x,q) = f_1 - \tfrac{1}{2}K_{22}q_0^2 - \tfrac{1}{2}\eta(1-x) + \tfrac{1}{2}K_{22}(q_0 - qx)^2 \\ - \tfrac{1}{2}K_{33}q^2(x^2 - b_0x), \end{aligned} \tag{7}$$

where $x = \sin^2\theta$ and f_1 is the uniform part of the free-energy density of a liquid crystal, independent of the orientation of the director $\mathbf{n}$ and the helix axis $\mathbf{t}$ and therefore insignificant for the further considerations [14]. Equation (7) has been derived under the assumption that $\mathbf{n}$ and $\mathbf{t}$ are expressed by the formulas $\mathbf{n} = \sin\theta\cos qz\hat{\mathbf{x}} + \sin\theta\sin qz\hat{\mathbf{y}} + \cos\theta\hat{\mathbf{z}}$ and $\mathbf{t} = \hat{\mathbf{z}}$, which describe the typical structure of the N_{TB} phase with the fixed helix axis parallel to the certain direction in the space (conventionally the z axis of the coordinate system).

The conditions for the minimum of the free-energy density are, according to the multivariable calculus, as follows:

$$\begin{aligned} &\frac{\partial f}{\partial x} = 0, \\ &\frac{\partial f}{\partial q} = 0, \\ &\frac{\partial^2 f}{\partial x^2} > 0, \\ &\det \mathbf{H} = \begin{vmatrix} \frac{\partial^2 f}{\partial x^2} & \frac{\partial^2 f}{\partial x \partial q} \\ \frac{\partial^2 f}{\partial q \partial x} & \frac{\partial^2 f}{\partial q^2} \end{vmatrix} > 0. \end{aligned} \tag{8}$$

In other words, the first-order partial derivatives must be equal to zero in the minimum and at the same time the Hessian matrix $\mathbf{H}$ should be positive definite. The application of these conditions to Eq. (7) leads to the system of equations and inequalities:

$$\begin{aligned}
\tfrac{1}{2}\eta + K_{22}q(qx - q_0) - \tfrac{1}{2}K_{33}q^2(2x - b_0) = 0,\\
K_{22}x(qx - q_0) - K_{33}q(x^2 - b_0x) = 0,\\
(K_{22} - K_{33})q^2 > 0,\\
q^2(K_{22} - K_{33})[K_{22}x^2 - K_{33}(x^2 - b_0x)]\\
-[K_{22}(2qx - q_0) - K_{33}q(2x - b_0)]^2 > 0.
\end{aligned} \tag{9}$$

As a result of some mathematical operations, the following four nontrivial solutions of (9) can be obtained:

$$\begin{aligned}
x &= \frac{-b_0K_{33} + K_{22}q_0\sqrt{\frac{b_0K_{33}}{\eta}}}{K_{22} - K_{33}},\\
q &= \sqrt{\frac{\eta}{b_0K_{33}}},\\
K_{22} &> K_{33},\\
K_{22}q_0 &> \sqrt{\eta b_oK_{33}},\\
\eta &> 0,\\
b_oK_{33} &> 0,
\end{aligned} \tag{10}$$

$$\begin{aligned}
x &= \frac{-b_0K_{33} - K_{22}q_0\sqrt{\frac{b_0K_{33}}{\eta}}}{K_{22} - K_{33}},\\
q &= -\sqrt{\frac{\eta}{b_0K_{33}}},\\
K_{22} &> K_{33},\\
K_{22}q_0 &< -\sqrt{\eta b_oK_{33}},\\
\eta &> 0,\\
b_oK_{33} &> 0,
\end{aligned} \tag{11}$$

$$\begin{aligned}
x &= \frac{-b_0K_{33} + K_{22}q_0\sqrt{\frac{b_0K_{33}}{\eta}}}{K_{22} - K_{33}},\\
q &= \sqrt{\frac{\eta}{b_0K_{33}}},\\
K_{22} &> K_{33},\\
K_{22}q_0 &< -\sqrt{\eta b_oK_{33}},\\
\eta &< 0,\\
b_oK_{33} &< 0,
\end{aligned} \tag{12}$$

$$\begin{aligned}
x &= \frac{-b_0K_{33} - K_{22}q_0\sqrt{\frac{b_0K_{33}}{\eta}}}{K_{22} - K_{33}},\\
q &= -\sqrt{\frac{\eta}{b_0K_{33}}},\\
K_{22} &> K_{33},\\
K_{22}q_0 &> \sqrt{\eta b_oK_{33}},\\
\eta &< 0,\\
b_oK_{33} &< 0.
\end{aligned} \tag{13}$$

The results (10) and (11) relate to two variants of the N_{TB} structure having opposite handedness—right handed ($q > 0$) and left handed ($q < 0$), respectively. The in-depth analysis of solutions (12) and (13) indicates that x takes negative values, so these cases are nonphysical and they must be rejected. It can be concluded that a stable twist-bend nematic phase is formed only if parameters η and b_oK_{33} are positive, as in (10) and (11). This fact differs from that presented by Barbero *et al.* [14], who predict the stability of the N_{TB} phase also for negative values of η and b_oK_{33}. The detailed calculations indicate that the free-energy density does not reach minimum for the values of x and q given by Eqs. (5) and (6) when the criteria (4) are satisfied. In this situation the determinant of the Hessian matrix remains negative and thus the twist-bend nematic phase is not stable.

The same conclusions can be formulated after the analysis of graphs presenting how the free-energy density f of the N_{TB} phase depends on $\sin^2\theta$ and q (Fig. 2). When $\eta > 0$ and $b_oK_{33} > 0$ [Fig. 2(a)], the values of $\sin^2\theta$ and q calculated from Eqs. (5) and (6) actually correspond to the minimum of f. In the case of negative values of η and b_oK_{33} [Fig. 2(b)], there exists only a saddle point, the achievement of which does not ensure the stability of a twist-bend nematic phase.

It should be noticed that the condition ensuring $x < 1$ is not given in any papers devoted to Barbero's theory and its consequences. For both cases (10) and (11) this criterion takes a simple form:

$$\eta > \frac{b_0K_{33}K_{22}^2q_0^2}{(K_{22} - K_{33} + b_0K_{33})^2}. \tag{14}$$

After the equivalent transformation of the condition $K_{22}q_0 > \sqrt{\eta b_oK_{33}}$ (or $K_{22}q_0 < -\sqrt{\eta b_oK_{33}}$), it can be concluded that the N_{TB} phase appears when the coupling parameter η satisfies the inequality

$$\frac{b_0K_{33}K_{22}^2q_0^2}{(K_{22} - K_{33} + b_0K_{33})^2} < \eta < \frac{K_{22}^2q_0^2}{b_0K_{33}}. \tag{15}$$

It is noteworthy that the upper limit for η guarantees that $x > 0$.

Unfortunately, the incorrect stability conditions have been applied by other scientists in their own research. For example, Lelidis and Kume have analyzed the case $b_oK_{33} < 0$ in their study on the linear electro-optic effect occurring in the N_{TB} phase [20]. This indicates that the knowledge of the stability criteria of a twist-bend nematic is especially important when the deformations in external fields are considered from the theoretical point of view.

It is worth examining which liquid-crystalline phases appear when η takes values from outside the stability interval (15). The cholesteric phase (N^*) becomes stable in the following case:

$$\eta \leqslant \frac{b_0K_{33}K_{22}^2q_0^2}{(K_{22} - K_{33} + b_0K_{33})^2}. \tag{16}$$

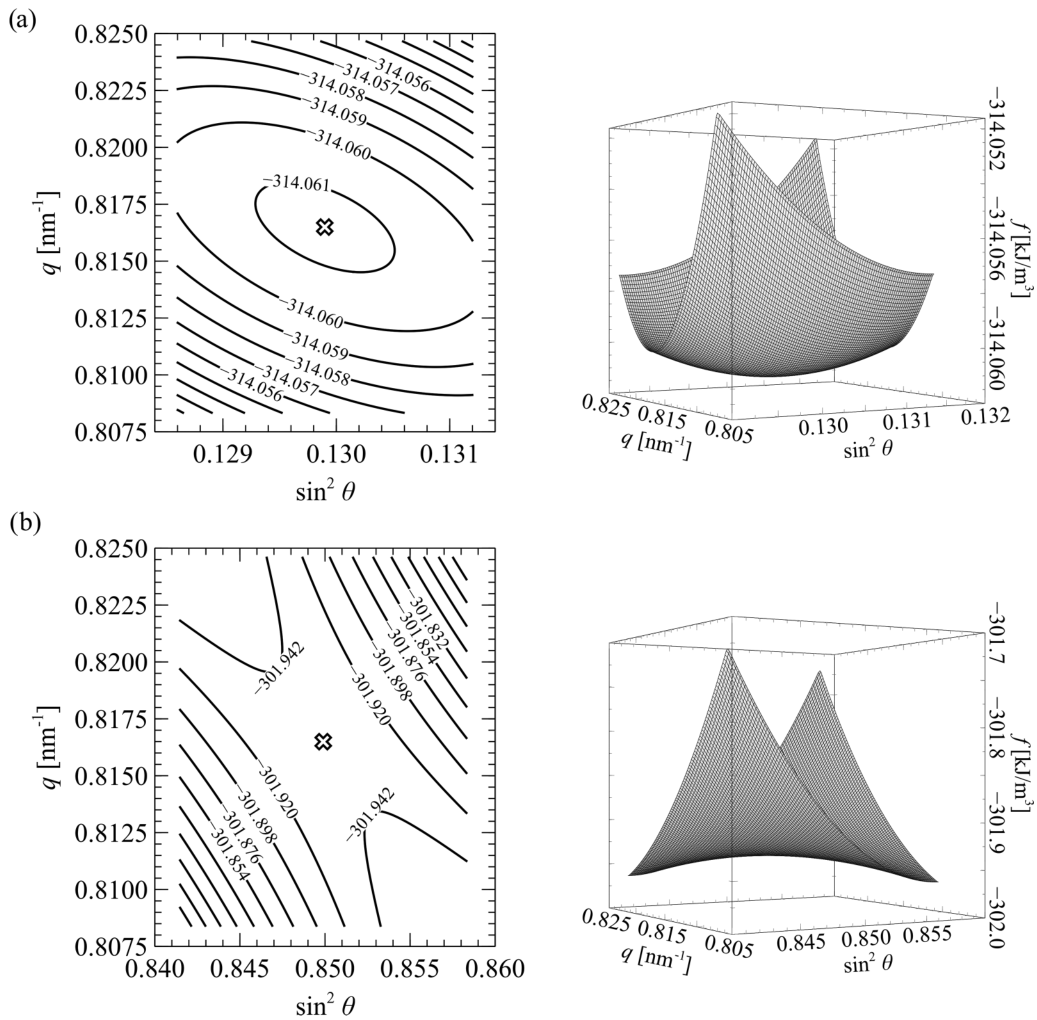

FIG. 2. Free-energy density f of twist-bend nematic as a function of sine squared of tilt angle $\sin^2\theta$ and wave number q. In (a) material parameters satisfy conditions (3): $K_{22} = 3$ pN, $K_{33} = 0.5$ pN, $\nu_4 = 0.2$ pN ($b_0K_{33} = 0.9$ pN), $\eta = 6 \times 10^5$ N/m^2, $K_{22}q_0 = 0.001$ N/m. In (b) material parameters fulfill conditions (4): $K_{22} = 3$ pN, $K_{33} = 0.5$ pN, $\nu_4 = -0.7$ pN ($b_0K_{33} = -0.9$ pN), $\eta = -6 \times 10^5$ N/m^2, $K_{22}q_0 = 0.001$ N/m. Cross signs indicate the values of $\sin^2\theta$ and q calculated from Eqs. (5) and (6), corresponding to minimum (a) and saddle point (b) of free-energy density. Values of f at contour lines are expressed in kJ/m^3.

The uniform nematic structure is formed when

$$\eta \geqslant \frac{K_{22}^2 q_0^2}{b_0 K_{33}}. \tag{17}$$

These conclusions have been formulated after the comparison of the values of the free-energy density f calculated for various liquid-crystalline phases:

$$f_{N_{\mathrm{TB}}} = f_1 + \frac{K_{22}^2 q_0^2 - 2 b_0 K_{33} |K_{22} q_0| \sqrt{\frac{\eta}{b_0 K_{33}}} + \eta (K_{22} - K_{33} + b_0 K_{33})}{2(K_{33} - K_{22})} \tag{18}$$

for twist-bend nematics,

$$f_N = f_1 - \tfrac{1}{2}\eta \tag{19}$$

for nematics, and

$$f_{N^*} = f_1 - \frac{K_{22}^2 q_0^2}{2(K_{22} - K_{33} + b_0 K_{33})} \tag{20}$$

for cholesterics. Figure 3 shows how f depends on η when typical values of material parameters are assumed. The phase which is characterized by the smallest value of the free-energy density is formed in reality. When η is increased, the following sequence of liquid-crystalline structures is revealed: $N^* \rightarrow N_{\mathrm{TB}} \rightarrow N$. Similar results have been obtained in computer simulations based on Barbero's model.

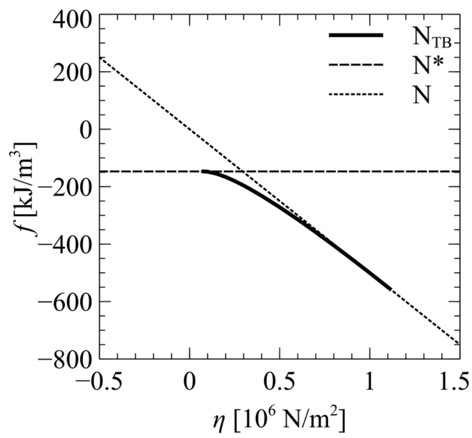

FIG. 3. Free-energy density f of twist-bend nematics (N_{TB}), cholesterics (N^*), and ordinary nematics (N) as a function of coupling parameter η. Material parameters are as follows: $K_{22} = 3$ pN, $K_{33} = 0.5$ pN, $\nu_4 = 0.2$ pN ($b_0K_{33} = 0.9$ pN), $K_{22}q_0 = 0.001$ N/m. It is assumed that uniform part of free-energy density $f_1 = 0$.

When the magnetic field is applied to the liquid crystal, the term due to the magnetic free-energy density, $\mu_0\Delta\chi H^2$, should be added to η in (15):

$$\frac{b_0K_{33}K_{22}^2q_0^2}{(K_{22} - K_{33} + b_0K_{33})^2} < \eta + \mu_0\Delta\chi H^2 < \frac{K_{22}^2q_0^2}{b_0K_{33}}. \tag{21}$$

The inequality (21) can be rewritten in the equivalent form:

$$\frac{b_0K_{33}K_{22}^2q_0^2}{(K_{22}-K_{33} + b_0K_{33})^2}-\mu_0\Delta\chi H^2 < \eta < \frac{K_{22}^2q_0^2}{b_0K_{33}} - \mu_0\Delta\chi H^2. \tag{22}$$

Figure 4 presents how the limiting values η_{lower} and η_{upper} depend on the square of the magnetic-field intensity H^2. As shown in Ref. [19], the magnetic field shifts the stability interval of twist-bend nematics towards lower values of η, but its width remains constant. The analysis of this phenomenon indicates that the magnetic field can induce not only the N_{TB}-N transition, but also the transformation from the cholesteric to the twist-bend nematic structure. Indeed, when the magnetic susceptibility anisotropy $\Delta\chi$ is positive, the magnetic torque tends to align the director parallel to the vector of the field intensity so the tilt angle decreases and thus the N^*-N_{TB} transition occurs while the reverse transition is impossible. Moreover, it should be noticed that in a very strong magnetic field the N_{TB} phase can be stable even when η is negative. This statement is not in contradiction to the above considerations which concern twist-bend nematics in the absence of external fields.

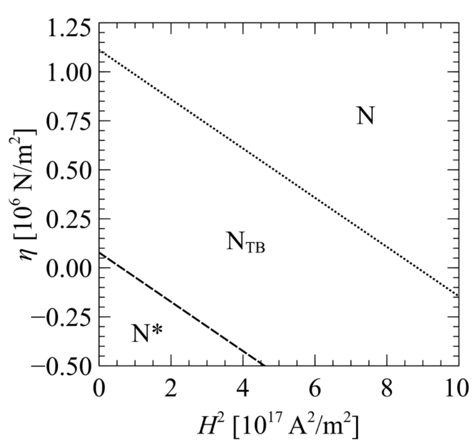

FIG. 4. Lower limit η_{lower} (dashed line) and upper limit η_{upper} (dotted line) of stability interval of twist-bend nematics (N_{TB}) as a function of square of magnetic-field intensity H^2. Above η_{upper} ordinary nematic phase (N) is stable. Below η_{lower} cholesteric structure (N^*) is formed. Material parameters are as follows: $K_{22} = 3$ pN, $K_{33} = 0.5$ pN, $\nu_4 = 0.2$ pN ($b_0K_{33} = 0.9$ pN), $K_{22}q_0 = 0.001$ N/m, $\Delta\chi = 10^{-6}$.

IV. CONCLUSIONS

To sum up, the formulas for the characteristic parameters of the N_{TB} phase and the conditions for its stability are gathered in the final form:

$$x = \frac{-b_0K_{33} \pm K_{22}q_0\sqrt{\frac{b_0K_{33}}{\eta}}}{K_{22} - K_{33}},$$

$$q = \pm\sqrt{\frac{\eta}{b_0K_{33}}},$$

$$K_{22} > K_{33},$$

$$b_oK_{33} > 0,$$

$$\frac{b_0K_{33}K_{22}^2q_0^2}{(K_{22} - K_{33} + b_0K_{33})^2} < \eta < \frac{K_{22}^2q_0^2}{b_0K_{33}}. \tag{23}$$

In particular, the inequalities for the parameter η have been derived. The upper signs should be chosen when $K_{22}q_0 > 0$, the lower ones when $K_{22}q_0 < 0$.

It has been shown that the negative values of η and b_oK_{33} do not lead to the formation of the stable N_{TB} structure in the absence of external fields. The phase transition from twist-bend nematics to cholesterics does not occur in magnetic field. For $\eta > 0$ and $b_oK_{33} > 0$, the N_{TB}-N transition has been theoretically predicted, but the production of the necessary magnetic-field strength seems unfeasible, especially when η is small. The analysis of the shift of the stability interval in the magnetic field reveals that the N^*-N_{TB} transition is also theoretically possible. When the coupling parameter η is smaller than the lower limit of the stability interval, the cholesteric phase is formed. When η exceeds the upper limit of the stability interval, the uniform nematic structure is stable.

Further research could be directed towards the comparison of the stability criteria obtained from various elasticity models and further analysis of the phenomena occurring in external fields.

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