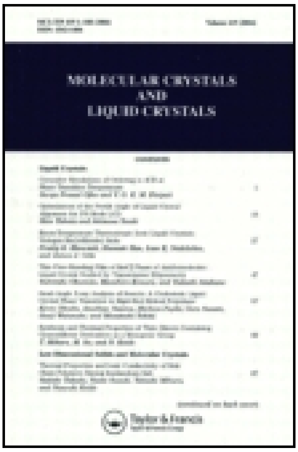

Molecular Crystals and Liquid Crystals
Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/gmcl20

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Version of record first published: 14 Jun 2011

To cite this article: A. Kapanowski (2011): Straley Model of Biaxial Nematics Extended, Molecular Crystals and Liquid Crystals, 540:1, 50-58

To link to this article: http://dx.doi.org/10.1080/15421406.2011.568327

Mol. Cryst. Liq. Cryst., Vol. 540: pp. 50–58, 2011

ISSN: 1542-1406 print/1563-5287 online
DOI: 10.1080/15421406.2011.568327

Straley Model of Biaxial Nematics Extended

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*In 1974 Straley [*Phys. Rev. A *10, 1881 (1974)] presented the mean-field model of interacting particles similar to hard rectangular blocks. The orientation-dependent potential was built from the four basic functions composed from the standard rotation matrix elements with $j = 2$. As there is only one basic function composed from the standard rotation matrix elements with $j = 3$, it is interesting to find out whether it has any influence on the known phase diagrams. We show that the $j = 3$ term can enlarge the biaxial nematic phase range. The article also discusses an extension of the Romano model mimicking shape amphiphilicity [*Phys. Lett. A *333, 110 (2004)]. It is showed that the $j = 3$ interaction term can create the tetrahedral phase with the second order transition to the isotropic phase. What is more, the first order transition from the biaxial nematic phase to the isotropic phase or to the tetrahedral phase is predicted. Finally, the possible presence of the $j = 3$ term in other theories is discussed.*

Keywords Biaxial nematics; symmetry adapted; tetrahedral symmetry

1. Introduction

Statistical theories of uniaxial nematic liquid crystals span between two opposite ideas. The first idea started from the paper by Onsager [1], where the nematic ordering was found by assuming that molecules can be represented as elongated hard cylinders only acting by excluded volume. The second idea is Maier and Saupe's mean-field description of liquid crystals [2], where the most important are anisotropic long-range, attractive interactions. A similar model for biaxial molecules was proposed by Straley in 1974 [3]. In this model the orientation-dependent potential was built from the four basic functions compatible with the D_{2h} symmetry. The phase diagram with four phases was predicted: the isotropic phase I, the prolate uniaxial nematic phase N_{U+}, the oblate uniaxial nematic phase N_{U-}, and the biaxial nematic phase N_B. The predicted phase transitions were first order (N_U – I) or second order (N_B – N_U, N_B – I).

The Straley predictions were considered to be universal for many years because they were confirmed by the excluded volume models and computer simulations also in rod-plate mixtures [4–8]. However, it was realized [9] that the Straley potential could lead to a different phase diagram: it shows the onset of two tricritical points [10,11]. This eventually resulted to be the most general picture as it includes the one with a

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single Landau point as a special case [12]. The existence of the critical point was confirmed by both a Monte Carlo simulations [13] and an experimental study [14]. Similar or even more complex phase diagrams were obtained recently using the Landau-de Gennes theory [15]. In 2004 Romano [16] used a simplified Straley model to describe mesogens containing both rod-like and disc-like parts covalently bonded together. The model has an extreme biaxial mesogenic form and we will analyze its extension.

The Straley model is based on the symmetry-adapted functions built from the standard rotation matrix elements with j = 2. Many authors limit theirs considerations to the j = 2 subspace because it is the smallest subspace that accommodates isotropic, uniaxial, and biaxial phases [17]. But there is the infinite set of functions with greater j that also satisfy the same symmetry conditions [4,18]. There is only one function in the j = 3 family and it is interesting to find out whether it has any influence on the known phase diagrams. On the other hand, we would like to check the presence of the j = 3 term in other theories.

This paper is organized as follows. In Section 2 the symmetry-adapted functions for the description of the biaxial nematic phase are defined. The Straley model of biaxial nematics and its extension is described in Section 3. Section 4 contains an extension of the Romano model mimicking shape amphiphilicity. In Sections 5 and 6 we discuss the presence of the j = 3 term in hard particle fluids and in the statistical theory with Corner interactions, respectively. In Section 7 we summarize the conclusions of our paper.

2. Invariants and Order Parameters

The orientation of a biaxial object can be specified by the three Euler angles $R = (\varphi, \vartheta, \psi)$ or by the three orthonormal vectors $(\vec{l}, \vec{m}, \vec{n})$. The distribution of orientations can be described by a distribution function $f(R)$, where

$$\int dR\, f(R) = \int d\varphi d\vartheta \sin\vartheta d\psi f(\varphi, \vartheta, \psi) = 1.$$

The ensemble average of any function $A(R)$ can be calculated as

$$\langle A \rangle = \int dR f(R) A(R).$$

In the case of the biaxial molecules the distribution function $f(R)$ can be expanded in a complete basis set of real functions $F^{(j)}_{\mu\nu}(R)$ spanning the relevant orientational space. The functions $F^{(j)}_{\mu\nu}(R)$ are defined in Ref. [18] and here we give only basic relations. The coefficients μ and ν are always even, the functions $D^{(j)}_{\mu\nu}(R)$ are standard rotation matrix elements [19]. If j is even, then $0 \le \mu,\ \nu \le j$,

$$F^{(j)}_{00}(R) = D^{(j)}_{00}(R),$$

$$F^{(j)}_{0\nu}(R) = \frac{1}{\sqrt{2}}[D^{(j)}_{0\nu}(R) + D^{(j)}_{0-\nu}(R)],$$

$$F^{(j)}_{\mu 0}(R) = \frac{1}{\sqrt{2}}[D^{(j)}_{\mu 0}(R) + D^{(j)}_{-\mu 0}(R)],$$

$$F^{(j)}_{\mu\nu}(R) = \frac{1}{2}[D^{(j)}_{\mu\nu}(R) + D^{(j)}_{\mu-\nu}(R) + D^{(j)}_{-\mu\nu}(R) + D^{(j)}_{-\mu-\nu}(R)].$$

If j is odd, then $2 \leq \mu,\ \nu \leq j$,

$$F^{(j)}_{\mu\nu}(R) = \frac{1}{2}[D^{(j)}_{\mu\nu}(R) - D^{(j)}_{\mu-\nu}(R) - D^{(j)}_{-\mu\nu}(R) + D^{(j)}_{-\mu-\nu}(R)].$$

Here is a list of the first five $F^{(j)}_{\mu\nu}(R)$ functions:

$$F^{(2)}_{00}(R) = \frac{1}{2}(3\cos^2\vartheta - 1),$$

$$F^{(2)}_{02}(R) = \frac{\sqrt{3}}{2}\sin^2\vartheta\cos(2\psi),$$

$$F^{(2)}_{20}(R) = \frac{\sqrt{3}}{2}\sin^2\vartheta\cos(2\varphi),$$

$$F^{(2)}_{22}(R) = \frac{1}{2}(1 + \cos^2\vartheta)\cos(2\varphi)\cos(2\psi) - \cos\vartheta\sin(2\varphi)\sin(2\psi),$$

$$F^{(3)}_{22}(R) = -\frac{1}{2}\cos\vartheta(3\cos^2\vartheta - 1)\sin(2\varphi)\sin(2\psi) + (2\cos^2\vartheta - 1)\cos(2\varphi)\cos(2\psi).$$

The $\langle F^{(2)}_{00}\rangle$ order parameter is a measure of the alignment of the $\vec{n}$ molecule axis along the Z axis of the reference frame. The $\langle F^{(2)}_{02}\rangle$ order parameter describes the relative distribution of the $\vec{l}$ and the $\vec{m}$ axes along the Z axis. Both $\langle F^{(2)}_{00}\rangle$ and $\langle F^{(2)}_{02}\rangle$ can be nonzero in the uniaxial nematic phase. The $\langle F^{(2)}_{20}\rangle$ order parameter describes the relative distribution of the $\vec{n}$ axis along the X and the Y axes. The $\langle F^{(2)}_{22}\rangle$ order parameter is related to the distribution of the $\vec{l}$ axis along the X axis and the distribution of the $\vec{m}$ axis along the Y axis. We note that the functions with j odd are sometimes missing in the expansions found in the literature [4]. On the other hand, there is a wealth of notations introduced in the past to denote orientational order parameters for biaxial nematics [20].

3. Straley Model

The potential energy of interactions in the Straley model can be written in a general form

$$V(R_1, R_2) = \sum_j \sum_{\mu\nu} v^{(j)}_{\mu\nu} F^{(j)}_{\mu\nu}(R_2^{-1} R_1),$$

where the coefficients $v^{(0)}_{00}$, $v^{(2)}_{00}$, $v^{(2)}_{02} = v^{(2)}_{20}$, and $v^{(2)}_{22}$ are nonzero in the original paper [3]. In the extended Straley model we will add the additional $v^{(3)}_{22}$ term. The state of the system is described by order parameters $\langle F^{(j)}_{\mu\nu}\rangle$. It is assumed that a given particle is subject to a mean field of the form

$$W(R) = \sum_j \sum_{\mu\nu} w^{(j)}_{\mu\nu} F^{(j)}_{\mu\nu}(R).$$

The distribution function is found from the Boltzmann distribution

$$f(R) = \exp[-\beta W(R)]/Z,$$

where Z is determined from the normalization of $f(R)$,

$$Z = \int dR\ \exp[-\beta W(R)].$$

The coefficients of W are determined from the self-consistency condition

$$W(R_1) = \int dR_2 f(R_2) V(R_1, R_2),$$

and the set of equations is

$$w^{(j)}_{\mu\nu} = \sum_{\rho} v^{(j)}_{\rho\nu} \langle F^{(j)}_{\mu\rho} \rangle.$$

In fact, the products $\beta v^{(j)}_{\mu\nu}$ are important because the distribution function of the system depends on $\beta w^{(j)}_{\mu\nu}$. The free energy of an equilibrium phase can be calculated as $F = U - TS$, where the internal energy U and the entropy S of the system are the following

$$U = \frac{N}{2} \int dR_1 dR_2 f(R_1) f(R_2) V(R_1, R_2),$$

$$S = -k_B N \int dR f(R) \ln f(R).$$

Note that T is the temperature, k_B is the Boltzmann constant, and $1/\beta = k_B T$. In his paper, Straley expressed the coefficients $v^{(2)}_{\mu\nu}$ by means of B, W, and L, which stand for breadth, width, and length of the platelets, respectively. He computed the excluded volume of a pair of platelets in some special relative orientations and then he made interpolation.

There is an interesting dual transformation between rod-like and disk-like systems in the model. If we exchange (L, B, W) with (L′, B′, W′) = (W, LW/B, L), then $v'^{(2)}_{\mu\nu} = (LW/B^2) v^{(2)}_{\mu\nu}$. The rod-like system ($B < \sqrt{LW}$) may be transformed to the disk-like system ($\sqrt{LW} < B$). The most biaxial object has $LW = B^2$ (self-dual point) and then there is the direct second order transition from the isotropic to the biaxial nematic phase.

Figure 1 is a phase diagram showing how the temperature at which the phase transition occurs depends on B for fixed L = 9 and W = 1. The tendency for the transition temperature to increase with B is due to the fact that the blocks are physically larger.

Figure 2 is a phase diagram for the extended Straley model, where $v^{(3)}_{22} = v^{(2)}_{22}$ and the dependence of $v^{(2)}_{\mu\nu}$ on (L, B, W) is unchanged. The line of the first order I – N_U transition is almost the same, but the area of the biaxial nematic phase is wider.

We note that the coefficient $v^{(3)}_{22}$ can not be obtained by means of the excluded volume method in the case of the platelets. The $v^{(3)}_{22}$ term is the lowest order term in the excluded volume of tetrahedral molecules [21]. Thus, the extended Straley model with $v^{(3)}_{22} = v^{(2)}_{22}$ describes slightly deformed biaxial molecules.

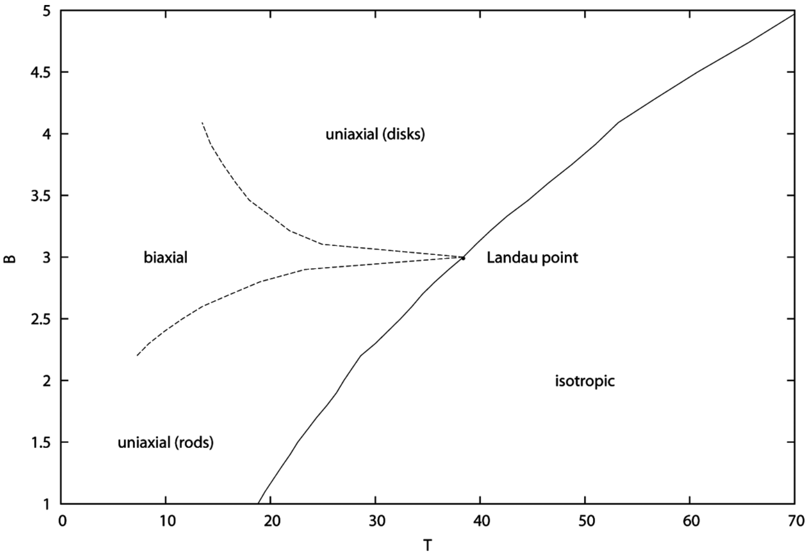

Figure 1. Phase diagram for blocks of varying breadth in the Straley model. The solid line marks the first-order transition and the dashed line represents the second-order transition. For the case L = 9, W = 1, and variable B, the direct second order transition from the isotropic to the biaxial nematic phase takes place for B = 3. The phases for B > 3 are related to the phases B < 3 by the dual transformation. The temperature T is in energy units.

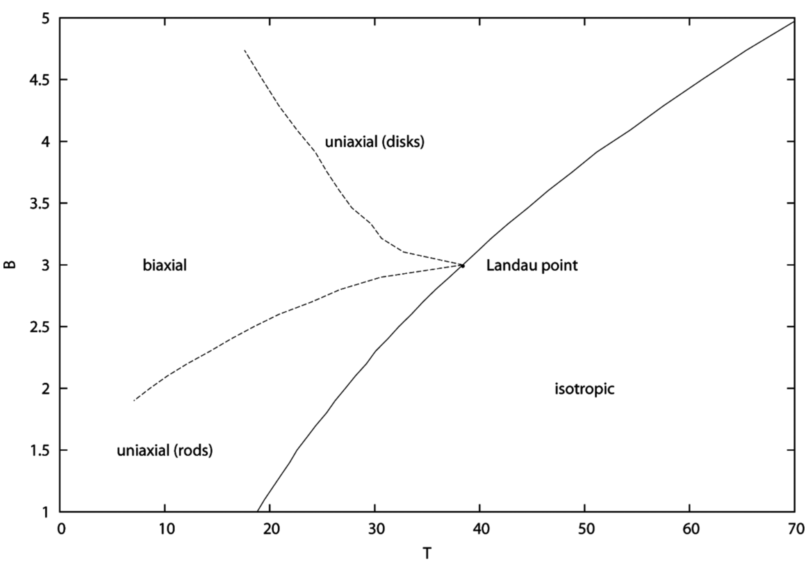

Figure 2. Phase diagram for the extended Straley model, where $v_{22}^{(3)} = v_{22}^{(2)}$ and the dependence of $v_{\mu\nu}^{(2)}$ on (L, B, W) is like in the original Straley model. The solid line marks the first-order transition and the dashed line represents the second-order transition. For the case L = 9, W = 1, and variable B, the line of the first order I – N_U transition is almost the same, but the area of the biaxial nematic phase is wider. The temperature T is in energy units.

4. Romano Model

In 2003 Sonnet, Virga, and Durand [9] proposed a mean-field model to describe the uniaxial and biaxial phases of mesogenic molecules presenting a shape dispersion of their biaxial dielectric susceptibility. The analyzed orientational interaction energy has the form

$$V(R_1, R_2) = v_{00}^{(2)} F_{00}^{(2)}(R_2^{-1} R_1) + v_{22}^{(2)} F_{22}^{(2)}(R_2^{-1} R_1),$$

where $v_{00}^{(2)} < 0$ and $v_{22}^{(2)} < 0$. The same model with $v_{00}^{(2)} = 0$ was used by Romano [16] to describe mesogens with shape amphiphilicity. A mean-field analysis predicted the existence of a direct second-order transition between biaxial and isotropic phases. The predictions were qualitatively confirmed by Monte Carlo simulations.

We would like to extend the Romano potential to the form

$$V(R_1, R_2) = v_{22}^{(2)} F_{22}^{(2)}(R_2^{-1} R_1) + v_{22}^{(3)} F_{22}^{(3)}(R_2^{-1} R_1),$$

where both terms are biaxial (D_{2h} symmetry). A mean-field calculations are similar to the case of the Straley model. A phase diagram for the extended Romano model is plotted in Figure 3.

Three phases are present: isotropic ($(\langle F_{\mu\nu}^{(j)} \rangle = 0)$), biaxial nematic (nonzero $\langle F_{00}^{(2)} \rangle$, $\langle F_{22}^{(2)} \rangle$, $\langle F_{22}^{(3)} \rangle$), and tetrahedral nematic ($(\langle F_{\mu\nu}^{(2)} \rangle = 0)$, nonzero $\langle F_{22}^{(3)} \rangle$). Two critical points C_1 and C_2 are predicted. C_1 is a tricritical point where the transition changes from second-order to first-order. C_2 is a point where three phases coexist in equilibrium: isotropic, biaxial nematic, and tetrahedral nematic.

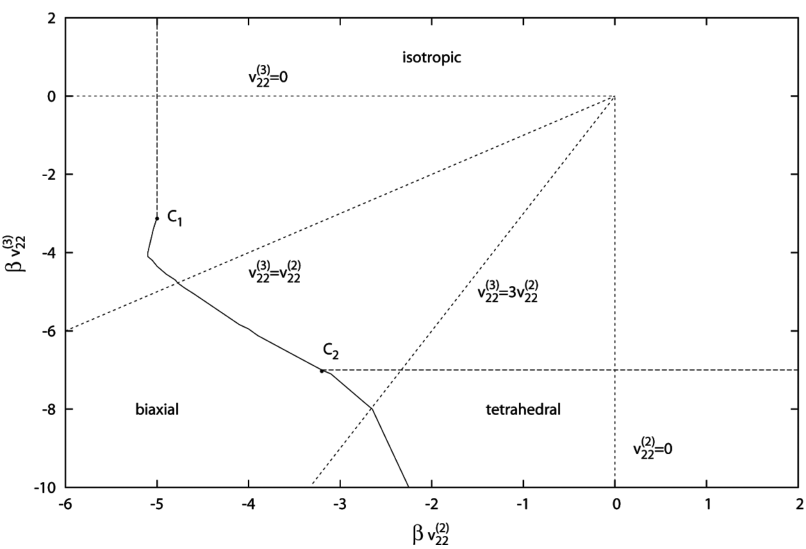

Figure 3. Phase diagram for the extended Romano model. The solid line marks the first-order transition and the dashed line represents the second-order transition. Three phases are present: isotropic, biaxial nematic, and tetrahedral. The C_1 and C_2 denote the tricritical and critical point, respectively. For a given physical system on decreasing the temperature we are moving from the centre (0, 0) to the edge of the figure.

It was shown recently [21] that $F_{22}^{(3)}$ is the lowest order function compatible with the tetrahedral symmetry T_d. Next is the following combination

$$F_{00}^{(4)} + \sqrt{5/7}(F_{04}^{(4)} + F_{40}^{(4)}) + (5/7)F_{44}^{(4)}.$$

Our calculations confirmed that in the tetrahedral phase

$$\langle F_{00}^{(4)} \rangle = \sqrt{7/5}\langle F_{04}^{(4)} \rangle = \sqrt{7/5}\langle F_{40}^{(4)} \rangle = (7/5)\langle F_{44}^{(4)} \rangle.$$

Figure 4 shows the temperature dependence of the order parameters in the case $v_{22}^{(3)} = 3v_{22}^{(2)}$. On decreasing temperature we meet the second-order transition to the tetrahedral phase and the first-order transition to the biaxial nematic phase.

5. Hard Particle Fluids

The (biaxial) spheroplatelet studied by Mulder [22,4] is a natural generalization of the (uniaxial) spherocylinder. It can be described as a rectangular block with dimensions $2a \times b \times c$, caped with quarter spheres of radius a and half-cylinders with radius a and lengths b and c such as to produce a piece-wise smooth convex body. It was the first non-axially symmetric convex body for which the pair-excluded volume $V(R_1, R_2)$ at fixed relative orientations was known in closed form.

The excluded volume $V(R_1, R_2)$, which plays the role of effective interaction, can be expanded in the series of the form

$$V(R_1, R_2) = \sum_j \sum_{\mu\nu} v_{\mu\nu}^{(j)} F_{\mu\nu}^{(j)}(R_2^{-1} R_1).$$

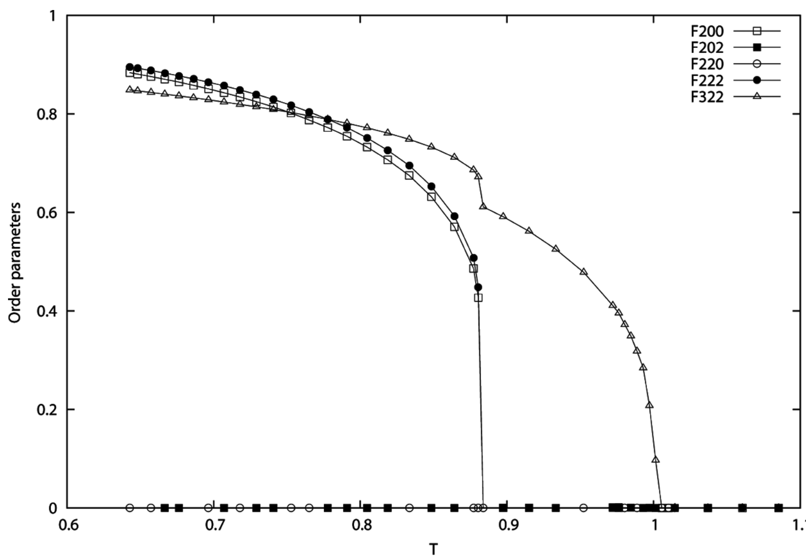

Figure 4. Temperature dependence of the order parameters in the extended Romano model for $v_{22}^{(3)} = 3v_{22}^{(2)}$. On decreasing the temperature T, there is the second-order transition from the isotropic to the tetrahedral phase (nonzero $\langle F_{22}^{(3)} \rangle$ only). Next, there is the first-order transition to the biaxial nematic phase. T is the dimensionless temperature.

It appeared, that the $v_{22}^{(3)}$ term is zero in the presented hard molecules model. In 2005 Mulder [23] computed the excluded volume for an important class of convex bodies, the spherozonotopes. By applying Mulder's results, Rosso and Virga [17] computed the quadrupolar projection ($v_{\mu\nu}^{(2)}$ terms) of the excluded volume between two spherocuboids. We checked that the $v_{22}^{(3)}$ term is not present also for the case of spherocuboids.

6. Statistical Theory with the Corner Potential Energy

The Corner potential energy [24] has the form $\Phi_{12}(u/\sigma)$, where u is the distance between molecules ($\overrightarrow{u} = u\overrightarrow{\Delta}$) and σ depends on the molecule orientation R_1 and R_2, and on the vector $\overrightarrow{\Delta}$. The functional dependence of the potential energy on u/σ can be the square well, the Lennard-Jones m-n, or some other functions. For σ it is possible to write the general expansion proposed by Blum and Torruella [25] which is evidently invariant under rotations and translations. In the case of the achiral biaxial molecules the lowest order terms of the expansion are

$$\sigma = \sigma_0 + \sigma_1\left[\left(\overrightarrow{\Delta}\cdot\overrightarrow{n_1}\right)^2 + \left(\overrightarrow{\Delta}\cdot\overrightarrow{n_2}\right)^2\right] + \sigma_2\left(\overrightarrow{n_1}\cdot\overrightarrow{n_2}\right)^2 + \sigma_3\left[\left(\overrightarrow{\Delta}\cdot\overrightarrow{l_1}\right)^2 + \left(\overrightarrow{\Delta}\cdot\overrightarrow{l_2}\right)^2\right] + \sigma_4\left(\overrightarrow{l_1}\cdot\overrightarrow{l_2}\right)^2 + \sigma_5\left[\left(\overrightarrow{l_1}\cdot\overrightarrow{n_2}\right)^2 + \left(\overrightarrow{l_2}\cdot\overrightarrow{n_1}\right)^2\right].$$

In the statistical theory of biaxial nematics [26] the following kernel is defined

$$K(R_1, R_2) = \int d\overrightarrow{\Delta}(\sigma/\sigma_0)^3 = \sum_j \sum_{\mu\nu} K_{\mu\nu}^{(j)} F_{\mu\nu}^{(j)}(R_2^{-1}R_1).$$

We note that $-\beta v_{\mu\nu}^{(j)}$ from the Mulder model corresponds to $\lambda K_{\mu\nu}^{(j)}$ from the Corner model, where λ depends on the density, σ_0, and on the function Φ_{12}. The nematic phase is present only if $\lambda < 0$. The $K_{22}^{(3)}$ term is present if any of the following expressions is nonzero: σ_5, $\sigma_2\sigma_4$, $\sigma_1\sigma_4$, $\sigma_2\sigma_3$. The nonzero $K_{22}^{(3)}$ term implies the presence of the nonzero $K_{22}^{(2)}$ term but the inverse of it is not true. In typical calculations the $K_{22}^{(3)}$ term is one order of magnitude smaller than $K_{22}^{(2)}$ term.

7. Conclusions

In this paper, we showed that the $F_{22}^{(3)}$ term can increase the uniaxial-biaxial nematic transition in the case of the Straley model. In the case of the Romano model the $j = 3$ term can change the isotropic-biaxial nematic transition type from second-order to first-order. A tetrahedral nematic phase can also appear with the second-order transition to the isotropic phase. According to Lubensky and Radzihovsky [28] thermal fluctuations should drive the transition first-order. On decreasing the temperature, the first-order transition from the tetrahedral nematic to the biaxial nematic phase is predicted.

We checked that the considered $j = 3$ term is not present in the excluded volume potential for spheroplatelets and spherocuboids. It seems that it can be true also for other hard convex biaxial particles. On the other hand, the $j = 3$ term appears naturally in a statistical theory with the Corner potential energy.

The functions $F_{\mu\nu}^{(j)}$ were introduced in the context of the biaxial nematic phase with the D_{2h} symmetry but it was necessary to use only the D_2 symmetry in the calculations [18]. As D_2 is a subgroup of T symmetry group, some $F^{(j)}_{\mu\nu}$ functions (or linear combinations) are compatible with T symmetry. The problem is that D_{2h} is not a subgroup of T_d but the functions $F^{(j)}_{\mu\nu}$ work well in both cases. It seems that implementation of the inversion is not properly understood in the literature [27].

At the end we would like to add that the order parameter $F^{(3)}_{22}$ should be calculated in computer experiments in order to get better phase description, especially when the tetrahedral phase can occur.

Acknowledgment

I would like to acknowledge Dr. Grzegorz Pająk (Jagiellonian University) for useful comments on tetrahedral phases.

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