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Viscosity of Biaxial Nematic Liquid Crystals

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Within the framework of phenomenological approach the expressions for dissipative function D and viscous stress tensor σ' of biaxial nematic liquid crystal (NLC) with arbitrary point symmetry group are constructed. The numbers N_v of independent viscous coefficients for the nematics of 32 crystal's lattice pointgroups and icosahedric groups are found. The condition following from the thermodynamical consideration of the non-negative determination for quadratic matrix Ξ , that was constructed with viscous coefficients, is formulated. For some of the point groups obvious inequalities for viscous coefficients are mentioned.

INTRODUCTION

In the past ten years a biaxial NLC class presenting a mesophase with a biaxial breakdown of full rotational symmetry $O(3)$, typical for the isotropic liquid, was singled out among liquid crystal phases and was actively investigated. These phases were first discovered¹ in lyotropic LC, and after that²—in thermotropic LC. Biaxial nematic phases admit arbitrary point symmetry groups—subgroups of $O(3)$ —including the groups, forbidden in crystal lattice. The symmetry of the majority of really existing biaxial nematics has not been established yet.

The investigations of biaxial nematics were devoted to the development of the continuous elasticity theory,^{3–5} phase transition theory,⁶ LC polarisation,⁴ classification of defects.^{7,8}

The hydrodynamical properties of biaxial NLC were discussed in many papers^{3–5, 9–13}; in great detail were considered the nematics with orthorhombic symmetry,^{3,4,9–13} some papers were also devoted to any other symmetries of NLC—triclinic,^{5,10} monoclinic,⁵ tetragonal,¹¹ hexagonal,¹⁰ cubic¹⁰ and quasiisotropic.¹⁰

The complexity of symmetrical analysis, which is necessary to build the dissipative function, invariant in relation to the acting of operations of some point groups, led to discrepancy in results for the number of viscous coefficients of orthorhombic nematic in paper¹⁰ and papers.^{4,9,11,12}† Besides this the correct form and, respectively, the right number of biaxial NLC viscous coefficients were not for all point groups of middle and high crystallographic systems found^{10,11}; e.g. going into hexagonal system groups C_6 and D_6 gave different numbers of independent com-

† In paper¹⁰ this discrepancy was mentioned.

ponents of 4-th rank tensor ν_{ijkl} and 3-d rank tensor λ_{ijk} (in Liu's¹⁰ description). All these concern groups C_4 and D_4 of tetragonal system and groups T and O of cubic system. No papers known to the author discussed the hydrodynamics of rhombohedral nematics.

Special interest is attached to incompressible biaxial nematics which most probably reflects the real situation. For these NLC the number of independent viscous coefficients were found only for orthorhombic symmetric.^{3,9}

The new biaxial mesophases synthesis^{14,15} testifies to the specific features of hydrodynamical properties in NLC of different symmetries (see also Reference 16). All this make it necessary to extend the earlier obtained results on hydrodynamics of biaxial NLC to a more wider set of symmetry classes—32 crystal lattice point-groups and icosahedric groups.

In recent papers^{17,18} analogical problems were considered by the author during the development of the continuous elasticity and flexoelectricity theories of biaxial nematics with an arbitrary point symmetry group. In these papers a well arranged apparatus¹⁹ of tensor analysis was used, when symmetry requirements were imposed on the tensor. In the present paper this apparatus is also employed.

ENERGY DISSIPATION IN BIAxIAL NEMATIC

The existence of viscosity leads to energy dissipation in the moving liquid. For an unlimited volume of isothermal liquid, which doesn't move at the infinity, the dissipative function D describes the local production of entropy $\delta ds/dt$ and is the quadratic form of independent thermodynamical forces x_i , if the latter are small²⁰

$$\delta T \frac{ds}{dt} = 2D = \sum_{i,j}^m L_{ij} x_i x_j \geq 0, \quad (1)$$

where ds/dt is full derivative of liquid entropy's mass density S with respect to the time t , δ and T —the density and temperature of liquid respectively, m —the number of valid thermodynamical forces. In the absence of influence exerted on the system by an external magnetic field or in the absence of rotation of the system as the whole the kinetic coefficients L_{ij} are satisfied to Onsager's reciprocity relations

$$L_{ij} = L_{ji} \quad (2)$$

The non-negative determination of the quadratic form (I) expressed a thermodynamical inreciprocity of the liquid's motion connected with the existence of internal friction in it. The thermodynamic forces x_i can be scalars (in case of volume viscosity), vectors (in case of heatconductivity) and tensors (in case of shear viscosity).

The thermodynamic flows T_i , adjointed to the thermodynamic forces x_i , are defined by the expression

$$T_i = \frac{\partial D}{\partial x_i} \quad (3)$$

The role of the thermodynamic forces in the case of isotropic viscous liquid is played by components of a second rank symmetrical tensor

$$A_{ij} = \frac{1}{2} \left(\frac{\partial v_i}{\partial x_j} + \frac{\partial v_j}{\partial x_i} \right),$$

where v_i are the cartesian components of the vector of points media velocity.

At the motion of the nematics phase an energy dissipation is caused by the alteration of NLC-molecules symmetries axes orientation's at their local rotation around their own inertial centers. This leads to the additional freedom degrees of the system and consequently leads to additional thermodynamic forces x_i , which are absent in the isotropic liquid. The theory of biaxial nematic is constructed by introducing in either NLC-point the vectors $\vec{n}_1, \vec{n}_2, \vec{n}_3$ triade of point groups G —a subgroup of $O(3)$ —group's. These vectors are interconnected by relations

$$\langle \vec{n}_i, \vec{n}_j \rangle = \xi_{ij}, \quad \frac{d}{dt} \xi_{ij} = 0, \quad (4)$$

where \langle, \rangle is the scalar product's symbol. At the homogeneous rotation of nematic as the whole with the rotational velocity $\vec{\omega} = \frac{1}{2} \text{rot } \vec{v}$ the same rotational velocity $\vec{\omega}$ will have all vectors fields $\vec{n}_i(\vec{r})$, $i = 1, 2, 3$. To describe the inhomogeneous nematic's rotation, three vectors \vec{N}_i could be used

$$\vec{N}_i = \frac{d\vec{n}_i}{dt} - [\vec{\omega} \times \vec{n}_i] = [(\vec{\Omega} - \vec{\omega}) \times \vec{n}_i], \quad (5)$$

satisfying the following relations

$$\langle \vec{N}_i, \vec{n}_j \rangle + \langle \vec{N}_j, \vec{n}_i \rangle = 0. \quad (6)$$

The relationship (Equation 6) could be derived by means of Equations (4), (5). The instantaneous rotational velocity's vector $\vec{\Omega}$ has been introduced into Equation (5) by the definition $d\vec{n}_i/dt = [\vec{\Omega} \times \vec{n}_i]$. The use of 9 components of 3 vectors $\vec{N}_i(N_{ix}, N_{iy}, N_{iz})$ as additional thermodynamic forces is inconvenient owing to 2 reasons:

- i) 6 conditions (Equation 6) reserve only 3 independent components of the vectors \vec{N}_i ;
- ii) by the construction of the true scalar D it would be more convenient to use the scalar values, that remain invariant after acting the operations of $O(3)$ -group.

By the use of the system of non-orthogonal coordinates, that are built on the ground of the directors \vec{n}_i triade, naturally 3 independent thermodynamic forces $\langle \vec{N}_i, \vec{n}_j \rangle$ are arised (f.i., $\langle \vec{N}_1, \vec{n}_2 \rangle, \langle \vec{N}_2, \vec{n}_3 \rangle, \langle \vec{N}_3, \vec{n}_1 \rangle$). Besides that, the tensors A_{ij} components in a new coordinate's system will be transformed in such a manner

$$B_{ij} = \sum_{k,l} c_{ik} A_{kl} c_{jl}, \quad (7)$$

where $c_{ik} = \langle \tilde{n}_i, \tilde{e}_k \rangle$, \tilde{e}_k is one of 3 normed ortes of cartesian coordinates system. Then the expression for the dissipative function D takes the form

$$2D = \sum_{ijkl} \{ \eta_{ijkl} B_{ij} B_{kl} + \mu_{ijkl} B_{ij} \langle \tilde{N}_k, \tilde{n}_l \rangle + \nu_{ijkl} \langle \tilde{N}_i, \tilde{n}_j \rangle \langle \tilde{N}_k, \tilde{n}_l \rangle \}, \quad (8)$$

where the 4-th rank tensors η , μ and ν have a inner Jahn-symbolic symmetry²¹

$$\begin{aligned} [[V^2]^2] \quad \eta_{ijkl} &= \eta_{ijlk} = \eta_{jikl} = \eta_{klij} \\ [V^2]\{V^2\} \quad \mu_{ijkl} &= \mu_{jikl} = -\mu_{ijlk} \\ [\{V^2\}^2] \quad \nu_{ijkl} &= \nu_{klij} = -\nu_{jikl} = -\nu_{ijlk} \end{aligned} \quad (9)$$

Due to the duality's relationship¹⁹ $\{V^2\} \sim \varepsilon V$ the tensor μ is equivalent to the symmetrical with respect to 2 indices pseudotensor of 3-d rank, and tensor ν —to the symmetrical 2-d rank tensor. The Onsager's reciprocity relations for tensors η , μ and ν are satisfied, also it must be mentioned, that the indices “ ij ” (also “ kl ”) are understood as one index.

Considering the expression (Equation 8) for D as a symmetric quadratic form in 9-dimensional euclidian space (6 tensor's B_{ij} components and 3 components of $\langle \tilde{N}_i, \tilde{n}_j \rangle$), the necessary and sufficient conditions²² of the expression's D non-negative determination might be formulated: all main minors (not only the corned minors) of determinant $\det \Xi$ must be non-negative, where Ξ is the quadratic 9×9 matrix. This leads to the inequalities, that tie the tensors η , μ , ν components.

The numbers N_η , N_μ , N_ν of these tensor's independent components will accordingly distinguish one from other in respect to the biaxial nematic's group of symmetry. The symmetry's existence leads to any relationship between the tensor's components. This relationship may be found for every symmetry group G ¹⁹, observing the transformation of components B_{ij} and $\langle \tilde{N}_i, \tilde{n}_j \rangle$ after group's G operations acting. In the Table the numbers N_η , N_μ , N_ν , N_α are presented, where $N_\alpha = N_\eta + N_\mu + N_\nu$ are the independent viscous coefficients for nematic phases number.

The data of Table coincide with the results, obtained for triclinic,^{5,10} monoclinic,⁵ orthorhombic^{3,4,9,11,12} and quasisotropic¹⁰ nematics and also for the nematics of high-symmetrical point groups of tetragonal,¹¹ hexagonal¹⁰ and cubic¹⁰ systems. For the low-symmetrical point groups of these systems and for the rhombohedral point groups our results are new.

Kini⁵ has paid attention to the coincidence of numbers N_α for biaxial nematics of triclinic, monoclinic and orthorhombic symmetries with the numbers N_λ of non-zero components of elastisity tensor λ for nematics with the same symmetries accordingly. The inner symmetry of λ is $[(V^2)^2]$.¹⁷ In reality this fact is of a general

‡ The number of independent components of 3-d rank tensor λ_{ijk} , obtained by Liu,¹⁰ is wrong. This number equals 3, in accordance with papers.^{4,9,11,12}

TABLE

Class of symmetry	$N_{\eta}/N_{\eta i}$	$N_{\mu}/N_{\mu i}$	N_{ν}	$N_{\alpha}/N_{\alpha i}$
Triclinic	21/15	18/15	6	45/36
Monoclinic	13/9	8/7	4	25/20
Orthorhombic	9/6	3	3	15/12
Rhombohedral				
C_3, S_6	7/5	6/5	2	15/12
C_{3v}, D_3, D_{3d}	6/4	2	2	10/8
Tetragonal				
C_4, S_4, C_{4h}	7/5	4/3	2	13/10
$C_{4v}, D_{2d}, D_{4h}, D_4$	6/4	1	2	9/7
Hexagonal				
C_{3h}, C_6, C_{6h}	5/3	4/3	2	11/8
$C_{6v}, D_{3h}, D_{6h}, D_6$	5/3	1	2	8/6
Cubic				
T, T_h	3/2	1	1	5/4
O, T_d, O_h	3/2	0	1	4/3
Icosahedral	2/1	0	1	3/2

nature and is common to biaxial nematics of an arbitrary point group of symmetry: that is a corollary from the trivial tensor-identity

$$\kappa_{ij}\kappa_{kl} = \bar{\kappa}_{ij}\bar{\kappa}_{kl} + \bar{\kappa}_{ij}\bar{\kappa}_{kl} + \bar{\kappa}_{ij}\bar{\kappa}_{kl} + \bar{\kappa}_{ij}\bar{\kappa}_{kl}$$

where $\bar{\kappa}_{ij}$ and $\bar{\kappa}_{ij}$ —symmetrical and antisymmetrical parts of 2-d rank tensor κ_{ij} accordingly.

By using Jahn's symbols the latter identity can be put down as follows:

$$[(V^2)^2] = [[V^2]^2] + [V^2]\{V^2\} + [\{V^2\}^2]$$

which proves our statement about the coincidence of numbers N_{α} and N_{λ} .

The numbers of inequalities binding the viscous coefficients of biaxial nematics of an arbitrary symmetry group coincide with the numbers of inequalities, binding the elasticity coefficients of biaxial nematics, and were presented in paper.¹⁷

Let us write the 16 thermodynamical inequalities for 15 viscous coefficients of orthorhombic nematics

$$\begin{aligned}
 \eta_{iiii} &\geq 0, \eta_{ijij} \geq 0, \eta_{iiii} \cdot \eta_{ijij} \geq \eta_{iiij}^2, \\
 \nu_{ijij} &\geq 0, \eta_{ijij} \cdot \nu_{ijij} \geq \mu_{ijij}^2, \\
 \eta_{1111} \cdot \eta_{2222} \cdot \eta_{3333} &+ 2\eta_{2323} \cdot \eta_{3131} \cdot \eta_{1212} \\
 &\geq \eta_{1111} \cdot \eta_{2323}^2 + \eta_{2222} \cdot \eta_{3131}^2 + \eta_{3333} \cdot \eta_{1212}^2
 \end{aligned} \tag{10}$$

Making the biaxial nematic's symmetry class higher, by means of Equation (10) we can get the inequalities for tetragonal, cubic and icosahedric nematics.

For instance, the expression of dissipative function D for cubic nematics of point groups O , T_d , O_h has the form

$$2D = \alpha_1(B_{11}^2 + B_{22}^2 + B_{33}^2) + 2\alpha_2(B_{11}B_{22} + B_{22}B_{33} + B_{33}B_{11}) + \alpha_3(B_{12}^2 + B_{23}^2 + B_{31}^2) + \alpha_4(\langle \vec{N}_1, \vec{n}_2 \rangle^2 + \langle \vec{N}_2, \vec{n}_3 \rangle^2 + \langle \vec{N}_3, \vec{n}_1 \rangle^2), \quad (11)$$

where α_i are coefficients, which satisfied to the inequalities

$$\alpha_1 \geq 0, \alpha_3 \geq 0, \alpha_4 \geq 0, \alpha_1 \geq |\alpha_2|, \alpha_1^3 + 2\alpha_2^3 \geq 3\alpha_1\alpha_2^2 \quad (12)$$

For the icosahedric groups Y , Y_h it must be taken in Equations (11), (12)

$$\alpha_1 - \alpha_2 = 2\alpha_3$$

Special interest is attached to the incompressible nematic phase, characterised by $\text{div } \vec{v} = 0$. The latter condition can be rewritten by means of Equation (7) in the following form

$$\sum_{ijk} d_{ij} B_{jk} g_{ki} = 0 \quad (13)$$

where matrixes d and g inverse to matrixes $\{c_{ij}\}$ and $\{c_{ji}\}$ accordingly.

The relationship (13) retains only 5 independent components of tensor B_{ij} . This leads to a decrease of the numbers N_η , N_μ , N_α and also to an alteration of the thermodynamical inequalities. Now the matrix's Ξ rank is 8. In the Table the numbers $N_{\eta i}$, $N_{\mu i}$, $N_{\alpha i}$ for incompressible nematic phase also are presented.

For the viscous stress tensor σ_{pq}^l construction let us find its symmetrical σ_{pq}^{ls} and antisymmetrical σ_{pq}^{la} parts, where “ p , q ”—cartesian system's indices and this system is built on the introduced in Equation (7) orthonormal basis \vec{e}_k . The adjoined thermodynamical force for the flow σ_{pq}^{ls} is A_{pq} , and for the flow $\sigma_{pq}^{la} - (\vec{\Omega} - \vec{\omega})_{pq} = \langle (\vec{\Omega} - \vec{\omega}), [\vec{e}_p \times \vec{e}_q] \rangle$. Transferring expression (8) into cartesian coordinates, we find

$$\begin{aligned} \sigma_{pq}^l &= \sigma_{pq}^l + \sigma_{pq}^l \\ \sigma_{pq}^{ls} &= \sum_{ijklzs} c_{1p} c_{1q} c_{kz} c_{ls} [\eta_{ijkl} A_{zs} + \mu_{ijkl} (\vec{\Omega} - \vec{\omega})_{zs}] \\ \sigma_{pq}^{la} &= \sum_{ijklzs} c_{iz} c_{js} c_{kp} c_{lq} [\mu_{ijkl} A_{zs} + \nu_{ijkl} (\vec{\Omega} - \vec{\omega})_{zs}] \end{aligned} \quad (14)$$

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