

Symmetry and topology analysis of the fluctuation parameters in nematic liquid crystals

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The fluctuations of the order parameter in nematic liquid crystals are analyzed on the basis of topology and symmetry considerations. It is shown that in the five-dimensional space of the order parameter of a nematic (represented by symmetric zero-trace second-rank matrices) there can exist only two cases corresponding to two types of orbits in five-dimensional space. One case (the orbit is a sphere S^2) is described by two no-gap and three gap modes of the order-parameter fluctuations. In the limit as $q \rightarrow 0$ (\mathbf{q} is a wave vector) two of the modes with gaps are degenerate. The second case [when the orbit is $SO(3)$] is characterized by three zero-gap modes and two gap fluctuation modes that are degenerate in the limit as $q \rightarrow 0$. Both cases are realized in experiment and correspond to uniaxial and biaxial nematic liquid crystals. In a uniaxial nematic, axial-symmetry conditions lead to a definite connection between the variation of the order-parameter components corresponding to degenerate gap and gapless modes. The dynamic aspects of symmetry and the physical consequences of the study are discussed.

1. INTRODUCTION

The calculation of fluctuations in liquid crystals (see e.g., Refs. 1–4) is based on an investigation of the free energy, determination of the equilibrium structure of the order parameter, and linearization of the corresponding equations of motion near this equilibrium structure. This procedure is as a rule quite cumbersome and is furthermore dependent on a specific model form of the free energy of the system.

This raises the natural question of identifying the fluctuation characteristics that are determined by the order-parameter itself and depend also on the form of the energy (a form based on model representations of the phase transition, of the type of interaction, and others). The present paper is devoted to just this group of questions.

It turns out that it is possible to make for the order-parameter fluctuations of nematic liquid crystals specific predictions without making any assumptions concerning the explicit form of the thermodynamic potential. A substantial role is played only by local symmetry and topology characteristics of the order-parameter space, which is five-dimensional in the general case.

Starting from the dimensionality of this space, it might be possible to obtain five different types of locally stable ground states of nematic liquid crystals, i.e., five types of nematic. It is well known, however, that in nature there are only two nematic liquid crystal types, called uniaxial and biaxial nematics. In both cases the order parameter is a symmetric zero-trace matrix of rank 2, but in the usual case some two arbitrary eigenvalues coincide, so that the matrix is described by only three (and not five) independent parameters. We show below that this simple fact is connected with rather general properties of the order-parameter space.

Similarly, some general characteristics of the order-parameter fluctuations are connected only with the properties of the aforementioned five-dimensional space. Foremost are the characteristics describing mode degeneracy and splitting, the presence or absence of a gap in the dispersion law, the structure of the Poisson brackets between the dynamic variables, etc. To establish these general laws it is not at all necessary to know the explicit form of the thermodynamic

potential, which is specified, for example in the case of biaxial nematic liquid crystals, by a large number of unknown phenomenological coefficients (in the simplest Landau–de Gennes approximation these are six coefficients of the expansion of the homogeneous part of the energy and 15 elastic moduli).

The scheme proposed in the present paper is in fact an analog of the traditional group-theoretical analysis of the lattice-oscillation spectrum in crystals. The only difference is that in solids one deals with discrete symmetry groups of the crystal, and in nematics with continuous symmetry groups acting in the order-parameter space. A similar approach was already used to describe and classify the eigenmodes in the B phase of He^3 (Ref. 5).

2. ORDER-PARAMETER SPACE OF A NEMATIC LIQUID CRYSTAL

It is known^{1–4} that the order parameter in a nematic liquid crystal is a symmetric real zero-trace matrix Q_{ij} . The specific physical meaning of the order parameter in the present paper is immaterial. Any quantity linear in Q_{ij} and therefore having the same transformation properties can serve equally well as the order parameter. The possible values of the matrix Q_{ij} are determined by minimizing the free energy, which can be symmetrically put in the form

$$F = V(Q_{ij}) + \text{Sp}(\nabla_i Q_{mn} t_{il} \nabla_l Q_{nm}). \quad (1)$$

Here V is the vector potential, whose explicit form is of no importance to us, and the second term specifies the so-called alastic energy. The nematic liquid crystals existing in nature are formed from the isotropic phase by a weak first-order transition (i.e., close to continuous). The potential must therefore be invariant to the three-dimensional rotation group (the elastic energy is written in (1) with explicit allowance for this invariance).

It is important that the free-energy minimum is degenerate and the symmetry present is described by the transformation

$$\hat{Q} \rightarrow R\hat{Q}R^{-1},$$

where R is the three-dimensional rotation matrix. In the absence of additional degeneracy, the values of the order parameter for this phase of the liquid crystal are of the form

$$\hat{Q} = R\hat{Q}_0R^{-1}, \quad (2)$$

where \hat{Q}_0 is a certain fixed matrix. Equation (2) makes possible a topological description of the order-parameter space, i.e., of the aggregate of its values that are compatible with the nematic phase.

According to the general theory⁶ the order-parameter space as a manifold will be a factor in the spaces

$$SO(3)/H,$$

where $SO(3)$ is the group of all three-dimensional rotations, H is the group of rotations that conserve \hat{Q}_0 :

$$R\hat{Q}_0R^{-1} = \hat{Q}_0.$$

We are not interested here in discrete symmetries. As to continuous nontrivial subgroups in $SO(3)$, there exists, accurate to conjugation, only one: $SO(2)$. The space of the order parameter can be only a sphere S^2 or $SO(3)$.

For actual calculations it is convenient to have a basis in the space of real symmetric zero-trace matrices

$$\begin{aligned} \hat{\psi}_1 &= \frac{1}{\sqrt{2}} \begin{pmatrix} 1 & 0 & 0 \\ 0 & -1 & 0 \\ 0 & 0 & 0 \end{pmatrix}, & \hat{\psi}_2 &= \frac{1}{\sqrt{2}} \begin{pmatrix} 0 & 1 & 0 \\ 1 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}, \\ \hat{\psi}_3 &= \frac{1}{\sqrt{2}} \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & 1 \\ 0 & 1 & 0 \end{pmatrix}, & & \\ \hat{\psi}_4 &= \frac{1}{\sqrt{2}} \begin{pmatrix} 0 & 0 & 1 \\ 0 & 0 & 0 \\ 1 & 0 & 0 \end{pmatrix}, & \hat{\psi}_5 &= \frac{1}{\sqrt{6}} \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & -2 \end{pmatrix}. \end{aligned} \quad (3)$$

The numerical coefficients in (3) are chosen to ensure the normalization condition:

$$\text{Sp}(\hat{\psi}_n \hat{\psi}_m) = \delta_{nm}.$$

This very basis was used earlier to investigate collective modes in He^3 -B (see Refs. 5 and 7-9).

Let us consider a situation in which the order-parameter space is two-dimensional. Without loss of generality we can confine ourselves to the case $\hat{Q}_0 = \hat{\Psi}_3$. The subgroup H of rotations that conserve $\hat{\Psi}_3$ consists then of rotations

$$D = \begin{pmatrix} \cos \varphi & -\sin \varphi & 0 \\ \sin \varphi & \cos \varphi & 0 \\ 0 & 0 & 1 \end{pmatrix}. \quad (4)$$

The action of the operator D on the basis matrices $\hat{\Psi}_i$ is specified by the equations

$$\begin{aligned} D\hat{\psi}_1D^{-1} &= \cos 2\varphi \cdot \hat{\psi}_1 + \sin 2\varphi \cdot \hat{\psi}_2, \\ D\hat{\psi}_2D^{-1} &= -\sin 2\varphi \cdot \hat{\psi}_1 + \cos 2\varphi \cdot \hat{\psi}_2, \end{aligned} \quad (5)$$

$$D\hat{\psi}_3D^{-1} = \cos \varphi \cdot \hat{\psi}_3 - \sin \varphi \cdot \hat{\psi}_4, \quad (6)$$

$$D\hat{\psi}_4D^{-1} = \sin \varphi \cdot \hat{\psi}_3 + \cos \varphi \cdot \hat{\psi}_4,$$

$$D\hat{\psi}_5D^{-1} = \hat{\psi}_5. \quad (7)$$

It follows thus from Eqs. (5)-(7) that with respect to the transformation D the entire five-dimensional space breaks up into two two-dimensional and one one-dimensional manifolds. Note also the following circumstance. For the operator D corresponding to rotation through a certain fixed angle φ the component of the order parameter in the $\hat{\psi}_3, \hat{\psi}_4$ plane is rotated through the same angle φ , whereas the components in the $\hat{\psi}_1, \hat{\psi}_2$ plane are rotated through double the angle, 2φ .

It is easy to verify by direct calculation that the space tangent to the sphere (which specifies in this case the order-parameter space) stretches at the point $\hat{\psi}_3$ over the matrices $\hat{\psi}_3, \hat{\psi}_4$ and the normal one on $\hat{\psi}_1, \hat{\psi}_2$.

3. NATURAL MODES OF ORDER-PARAMETER FLUCTUATIONS

Everything said above concerning the order-parameter space and some of its properties leads to the following statements concerning the ground state of the system and small deviations (fluctuations) from it.

As established in the preceding section, from the standpoint of the order-parameter symmetry only two different cases are possible (corresponding to two different types of nematic liquid crystals). In the first the order-parameter space is specified by the entire $SO(3)$ group, and in the second by the two-dimensional sphere S^2 .

Consider initially the first case. Let there be a minimum of free energy (1) in the class of solutions belonging to $SO(3)$. Obviously, all the displacements of this maximum can be of two types. In the first these are three-dimensional displacements in a space tangent in the initial point of the space minimum. Since these displacements will not move the minimum away from the stationary $SO(3)$ orbit, the (three) eigenmodes corresponding to these displacements are gapless (of the Goldstone type). Second, there exist two types of displacements that move the minimum away from the manifold $SO(3)$. These displacements correspond to gap modes.

Thus, independently of the actual form of the liquid-crystal energy, in the case of a biaxial nematic there are three Goldstone and two gap modes of fluctuation.²⁾

In the case of a stationary orbit, S^2 , there is another situation. This case corresponds to a certain singled-out direction in real space. Transformations of the rotation group around this axis splits the five-dimensional space of the order parameter into two two-dimensional and one one-dimensional manifolds. In these rotations, the components $\hat{\psi}_3$ and $\hat{\psi}_4$ are transformed to the stationary state in the manifold tangent to the stationary orbit, and the components $\hat{\psi}_1$ and $\hat{\psi}_2$ are transformed in the transverse space. A change of the component $\hat{\psi}_3$ moves the minimum away from the manifold S^2 . Similarly, the components $\hat{\psi}_1$ and $\hat{\psi}_2$ alter the order parameter in the manifold transverse to the stationary orbit, and consequently move the minimum away from this orbit, whereas the elements $\hat{\psi}_3$ and $\hat{\psi}_4$ acting in the tangential manifold S^2 leave the minimum on the orbit.

Thus, in the considered case of a preferred direction, (i.e., of a uniaxial nematic) there exist two zero-gap modes, two degenerate (in the limit as $q \rightarrow 0$, where \mathbf{q} is a wave vector) gap modes, and one more gap mode with a frequency different from those of the preceding ones.

4. POISSON BRACKETS

As already noted in the Introduction, we are interested in the present paper only in those nematic properties which are independent of the actual form of the thermodynamic potential, but are determined by the symmetry and topology of the order parameter. One of the most important characteristics of this type of system are the Poisson brackets, which determine in turn its dynamics.

It is known^{1,10} that the dynamic variables of a nematic liquid crystal are all the variables of an isotropic liquid (densities of the mass, momentum, and energy) and the degeneracy parameter. This parameter is specified by two independent components of the director unit vector in the case of a uniaxial nematic, while in the case of a two-axis nematic it is given by three rotation angles of a triplet of mutually perpendicular unit vectors that parametrize the displacements, described above and in the preceding section, of the minimum of the free energy in a manifold tangent to $SO(3)$.

However, in the case of weak first-order transitions (such as are all the known transitions from an isotropic liquid to a nematic) and with allowance for the narrow temperature range ΔT in which the nematic phase exists ($\Delta T/T_c \sim 10^{-2}$, where T_c is the temperature of the nematic \rightarrow isotropic liquid transition), all the components of the order parameters should be regarded as dynamic variables. We need thus a system of Poisson brackets which includes all five components of the matrix Q_{ij} .

It is convenient (following the analogous procedure¹¹ for uniaxial nematics) to introduce the components of the orbital momentum \mathbf{m} which are generators of infinitesimally small transformation of the type

$$\hat{Q} \rightarrow R\hat{Q}R^{-1}.$$

To avoid misunderstandings, we emphasize that in real nematics the variable \mathbf{m} has no macroscopic meaning. It must therefore be regarded as an auxiliary quantity. One must put $m = 0$ in the final dynamics equations. Introduction of \mathbf{m} makes it possible nonetheless to derive (and express in compact form) dynamics equations. In addition, one cannot exclude the possible (in principle) existence of nematic liquid crystals with a moment-of-inertia density that is small (compared with dissipation). In this case the density of the orbital momentum would have a macroscopic physical meaning.

The Poisson bracket $\{m_i, Q_{jk}\}$ is a commutator of $[Q, f_i]$, where f_i are generators, belonging to SO_3 , of infinitesimal rotations:

$$(f_i)_{ab} = -\varepsilon_{iab}, \quad (8)$$

where ε_{iab} is an entirely antisymmetric tensor:

$$\{m_i, \hat{Q}\} = [\hat{Q}, f_i]. \quad (9)$$

For an explicit calculation of the commutator in Eq. (9) we expand the order parameter in terms of the basis (3):

$$\hat{Q} = \sum_{i=1}^5 q_i \hat{\psi}_i. \quad (10)$$

Hence, with allowance for the normalization condition:

$$q_i = \text{Sp}(\hat{Q} \hat{\psi}_i). \quad (11)$$

Using these equations, we get:

$$\{m_i, q_j\} = q_k \text{Sp}(\hat{\psi}_j [\hat{\psi}_k, f_i]). \quad (12)$$

We put

$$\text{Sp} \hat{\psi}_j [\hat{\psi}_k, f_i] = -t_{ijk}. \quad (13)$$

The matrix t_{ijk} that determines the Poisson bracket $\{m_i, q_j\}$ can be easily obtained by direct calculation

$$\begin{aligned} t_{1jk} &= \frac{1}{2} \delta_{j1} \delta_{k3} - \frac{1}{2} \delta_{j2} \delta_{k4} + \frac{\sqrt{3}}{2} \delta_{j3} \delta_{k5}, \\ t_{2jk} &= \frac{1}{2} \delta_{j1} \delta_{k4} + \frac{1}{2} \delta_{j2} \delta_{k3} - \frac{\sqrt{3}}{2} \delta_{j4} \delta_{k5}, \\ t_{3jk} &= -\delta_{j1} \delta_{k2} + \frac{1}{2} \delta_{j3} \delta_{k4}. \end{aligned} \quad (14)$$

Thus, the Poisson bracket of interest to us is

$$\{m_i, q_j\} = -t_{ijk} q_k. \quad (15)$$

In this equation $i = 1, 2, 3$ and $j, k = 1, 2, 3, 4, 5$. The matrix t_{ijk} is defined by relations (14).

The Poisson brackets between the components of \mathbf{m} follow from the known commutation rules¹¹

$$\{m_i(r_1), m_j(r_2)\} = \varepsilon_{ijk} m_k(r_1) \delta(r_1 - r_2). \quad (16)$$

A separate and quite not trivial problem is the derivation of Poisson brackets between the momentum-density vector \mathbf{j} and the quantities q_i and \mathbf{m} . If, however, only the fluctuations of the order parameter are of interest (and it is just they which make the main contribution, for example, to light scattering), one can neglect in first-order approximation the contribution of all dynamic degrees of freedom except \hat{Q} and \mathbf{m} . The physical basis for this approximation is that the characteristic \hat{Q} fluctuation times are considerably longer in real liquid crystals than the fluctuation times for other hydrodynamic quantities, such as \mathbf{j} . Consideration of only the order-parameter fluctuations can be justified also if spatial gradients can be neglected. The group of problems connected with the structure of the Poisson brackets $\{j, q_i\}$ and $\{j, \mathbf{m}\}$ calls for a separate investigation. Here we note only that the right-hand sides of the corresponding brackets should contain both the usual terms, corresponding to displacement of \mathbf{m} and q_i along the current lines, also the additional terms connected with the rotations \mathbf{m} and q_i .

5. CONCLUSION

At present there is no need to convince any one of the usefulness of topological ideas in the physics of condensed media. As a rule, however, topological methods are used to describe and classify defects of various types (see, e.g., the reviews¹²). In the present paper, with nematic liquid crystals

as the example, we call attention to the feasibility of using topological method also for investigations of order-parameter fluctuations in complicated systems.

Roughly speaking, the procedure of topological analysis of order-parameter fluctuations is the following. We must separate all the orbit possible in the system's order-parameter space. Each orbit corresponds to possible equilibrium phases of the system. It is necessary next to determine the tangential and transverse manifolds. The first correspond to gapless (Goldstone) modes of the fluctuations, and the second to gap modes. Degeneracy of the gap modes is determined in a transverse manifold from symmetry considerations.

Clearly, such an analysis can in principle be confirmed directly by computations for which one must use, however, some model form of the thermodynamic potential.

A similar classification can be used in principle also to describe the dynamics of the fluctuations. It is known^{12,10} that relaxation dynamics can be described in the framework of some supersymmetrical action. It can thus be stated that the dynamics of fluctuations in nematic liquid crystals evolves from a supersymmetric generalization of the $SO(3)$ group. It is necessary to separate in this group the superorbits and find the tangents to the transverse supermanifolds (see the Appendix).

Finally, we note also that the obtained relation of the rotation "velocity" rates of the order-parameter components in tangential and transverse manifolds (and the corresponding ratio for the superorbits) denotes feasibility of the following phenomenon. Following a system-parameter change corresponding to motion along a closed contour in a normal manifold, the system state will not return to the initial point, because the rate of change of the components in a transverse manifold is half as large [this property is connected only with local symmetry of the minimum of the thermodynamic potential and is not a consequence of the known $\mathbf{n} \rightarrow \mathbf{n}$ global symmetry that reduces $SO(3)$ to RP^2]. The order parameter acquires thus a certain phase of geometric or topological type (a classical analog of the so-called Berry phase¹³), which can in principle be measured by optical methods.

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APPENDIX

The variation of the order parameter and of the momentum vector \mathbf{m} with time is described by the Langevin equations, which can be symbolically written in the form

$$\dot{X}_i = \{X_i, H\} + f_i + \eta_i \quad (\text{A1})$$

Here X_i ($i = 1, \dots, 8$) specifies the components of Q_{ij} and \mathbf{m} , H is the Hamiltonian, $\{x_i, H\}$ is a Poisson bracket, f_i is a generalized force, and η_i is noise, assumed to be Gaussian.

To calculate the mean values (correlators) it is necessary to average over the noise:

$$\overline{\Phi(X(\eta))} = c \int D\eta \Phi(X(\eta)) \exp\left(-\frac{1}{2} \int \eta^2 dt\right), \quad (\text{A2})$$

where c is a normalization factor and Φ is an averageable quantity. It is convenient, following Ref. 14, to make in (A2) the change of variables

$$D\eta = \frac{\delta\eta}{\delta X} DX, \quad (\text{A3})$$

and to obtain the random noise formally from the Langevin equation:

$$\eta_i = \dot{X}_i - \{X_i, H\} - f_i \quad (\text{A4})$$

The determinant $\|\delta\eta/\delta X\|$ which defines the functional derivative in (A3) can be expressed as an integral over the Fermi fields ψ_i :

$$\|\frac{\delta\eta}{\delta X}\| = \int D\bar{\psi} D\psi \exp\left[-\bar{\psi}_i \frac{\delta\eta_i}{\delta X_j} \psi_j\right]. \quad (\text{A5})$$

Substituting these equations in (A2), we represent the result of the averaging in the form:

$$\overline{\Phi(X(\eta))} = \int DX D\bar{\psi} D\psi \times \exp\left\{-\frac{1}{2} \int [(\dot{X} - \{X, H\} - f)^2 + \bar{\psi} \frac{\delta\eta}{\delta X} \psi] dt\right\}. \quad (\text{A6})$$

Averaging over the noise can thus be reduced to averaging with an effective supersymmetric action:

$$S^{\text{eff}} = \frac{1}{2} \int dt \left\{ [\dot{X} - \{X, H\} - f]^2 + \bar{\psi} \frac{\delta\eta}{\delta X} \psi \right\}. \quad (\text{A7})$$

The main ideas, definitions, and properties of the supersymmetric broadening of the groups can be found in Refs. 15. Here, in the context of the main content of our paper, we discuss only the possible types of stationary superorbits of the effective action (A7).

The case of thermodynamic equilibrium states can be obtained from (A7) by neglecting in the super-action S^{eff} the derivatives with respect to time, and putting $m = 0$ and $\Psi = 0$. We arrive thus at a minimization of the free energy F and to the orbits $SO(3)$ and S^2 obtained in the main text.

A more complicated situation takes place when the saddle trajectory in S^{eff} is determined from the conditions

$$\dot{X}_i = 0, \quad \mathbf{m} = 0, \quad \psi \neq 0.$$

In this case it is necessary to minimize the supersymmetric potential

$$U = \sum_i [\{X_i, H\} + f_i]^2 - \sum_{i,j} \bar{\psi}_j \left[\frac{\partial}{\partial X_j} \{X_i, H\} + \frac{\partial f_i}{\partial X_j} \right] \psi_i \quad (\text{A8})$$

The conditions for the extremum of U are of the form

$$\frac{\partial U}{\partial X_i} \Big|_{\mathbf{m}=0} = 0, \quad \frac{\partial U}{\partial \psi_j} \Big|_{\mathbf{m}=0} = 0. \quad (\text{A9})$$

They determine all the possible superorbits of this type.

The usual orbit (free-energy minimum) is contained as a submanifold in the superorbit specified by (A9). Then it is just the first equation of (A9) which contains the usual orbit, and the second

$$\left[\frac{\partial^2 F}{\partial X_k \partial X_j} (X_i, X_k) + \frac{\partial F}{\partial X_k} \frac{\partial}{\partial X_j} (X_i, X_k) + \frac{\partial f_l}{\partial X_j} \right] \psi_j = 0 \quad (\text{A10})$$

specifies the vector ψ_i —the section of the linear stratification above the orbit.

If the condition $(\partial F / \partial X) = 0$ that the free energy be stationary is satisfied, it follows from (A10) that this stratification above the orbit is directly connected with the gap modes of the system, since only for such modes do we have $\partial^2 F / \partial X_k \partial X_j \neq 0$.

In the general case $\dot{X} \neq 0$ the classification of all the possible types of superorbits and of all the possible types of superbroadening of the group $SO(3)$ is a problem outside the scope of the present paper.

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²¹All the foregoing pertains to an unbounded liquid-crystal sample in the absence of external volume forces. Otherwise only gap modes are present.

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