Large-scale dynamics of cholesterics and smectics C^*

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(Submitted 7 April 1986)
Zh. Eksp. Teor. Fiz. **91**, 878–890 (September 1986)

We consider the large-scale properties of cholesteric liquid crystals. A procedure based on eliminating the fast degrees of freedom is developed and permits a crossover from small- to large-scale characteristics of any system. A distribution function expressed in terms of the effective action is used to investigate dynamic properties within the framework of this model. It is shown that both the static and the dynamic large-scale properties of a cholesteric coincide with those of a smectic. We calculate all the static and dynamic large-scale characteristics that can be expressed in terms of the small-scale elastic moduli, kinetic coefficients, and the helical pitch of the cholesteric. A similar procedure is used also to describe the properties of chiral smectics C^*.

1. INTRODUCTION

Cholesteric liquid crystals are known to be produced in systems consisting of molecules that have no inversion center. The absence of the inversion center causes the ground state of a cholesteric to be inhomogeneous: the director **n** is twisted into a helix in this state.[1] The pitch L of the helix is usually much larger than the atomic dimensions. The properties of an inhomogeneously deformed cholesteric depend substantially on the ratio of the inhomogeneity scale λ and the helix pitch L. At scales $\lambda \ll L$ the absence of an inversion center is immaterial and the cholesteric has the same properties as an ordinary nematic. It should be expected from general considerations that for scales $\lambda \gg L$ the properties of a cholesteric, just like those of any other layered structure, coincide with those of a smectic. An attempt to corroborate this statement by using an averaging procedure was undertaken in Ref. 2, where the large-scale static properties of a cholesteric were considered (see also Ref. 3). The main purpose of the present paper is a systematic derivation of the equations of large-scale dynamics of cholesterics. Since, however, a rigorous procedure for obtaining large-scale static properties has likewise not been published heretofore, we derive parallel equations also for the static properties.

We develop here a method based on eliminating the rapid degrees of freedom, enabling us to calculate both static and dynamic large-scale properties of a cholesteric. We shall show that from the large-scale viewpoint a cholesteric is actually equivalent to a smectic, and express the parameters of a nematic that exists at scales $\lambda \ll L$.

The energy associated with the inhomogeneity of the director field is given for cholesterics by

$$E_F = {}^1/_2 [K_1 (\nabla_i n_i)^2 + K_2 (\varepsilon_{ijn} n_i \nabla_j n_n - q_0)^2 + K_3 (n_i \nabla_i n_j)^2]. \quad (1)$$

Here K_1, K_2, and K_3 are the Frank moduli. The presence in expansion (1) of a term linear in the derivatives is due to the abovementioned absence of an inversion center in cholesterics. The energy (1) reaches a minimum for

$$\mathbf{n} = \mathbf{u} \cos \Phi + \mathbf{u}' \sin \Phi, \quad (2)$$

where

$$\Phi = q_0 \mathbf{l r} + \Phi_0, \quad (3)$$

$$\mathbf{l} = [\mathbf{u}' \mathbf{u}]. \quad (4)$$

The vectors **u** and **u**′ in (2) are mutually orthogonal unit vectors, while Φ_0 is an arbitrary initial phase. The solution (2) describes a helix with pitch $L = 2\pi/q_0$ and with an axis directed along the unit vector **l**. The director field with large-scale deformation will also be expressed, following Refs. 2 and 3, in the form (2), where **u** and **u**′ are mutually orthogonal unit vectors as before, but now they vary in space slowly (i.e., for scales $\lambda \gg L$), and Φ is a phase no longer having the simple form (3). We shall assume, however, that the Fourier expansion of Φ has no rapid harmonics (i.e., having wave vectors $q \gtrsim L^{-1}$). This allows us to put

$$\overline{\cos \Phi} = \overline{\sin \Phi} = 0, \quad (5)$$

where the averaging is over scales $\sim L$.

The variation of the direction can be resolved along the unit vectors **l** and **n**×**l**. For the derivatives with respect to time and to the coordinates we have, using this expansion,

$$\frac{\partial \mathbf{n}}{\partial t} = s_0 [\mathbf{n l}] - \left(\mathbf{n} \frac{\partial}{\partial t} \mathbf{l} \right) \mathbf{l}, \quad (6)$$

$$\nabla_i n_j = s_i [\mathbf{n l}]_j - (\mathbf{n} \nabla_i \mathbf{l}) l_j. \quad (7)$$

Here

$$s_0 = \partial \Phi / \partial t + \mathbf{u}' \partial \mathbf{u} / \partial t, \qquad s_i = \nabla_i \Phi + \mathbf{u}' \nabla_i \mathbf{u}. \quad (8)$$

In view of the definitions (8), the quantities s_0 and s_j are connected by relations similar to those for the superfluid velocity in the A-phase of ^{3}He (Ref. 4):

$$\nabla_i s_j - \nabla_j s_i = \varepsilon_{mkn} l_m \nabla_i l_k \nabla_j l_n, \quad (9)$$

$$\nabla_i s_0 - \partial s_i / \partial t = \varepsilon_{mjn} \nabla_i l_j \partial l_n / dt. \quad (10)$$

As a result of (9) and (10), we can represent s_0 and s_j in the form

$$s_i = \nabla_i W + (\nabla_l / \nabla^2) (\varepsilon_{mjn} l_m \nabla_l l_j \nabla_i l_n), \quad (11)$$

$$s_0 = \partial W / \partial t + (\nabla_l / \nabla^2) (\varepsilon_{mjn} l_m \nabla_l l_j \partial l_n / \partial t). \quad (12)$$

The representations (11) and (12) demonstrate explicitly that three degrees of freedom appear in our large-scale de-

0038-5646/86/090518-07$04.00

scription of a cholesteric; the smectic one, related to the variable W, and two degrees connected with the unit vector **l**. It will be shown below that only the smectic degree of freedom is of the Goldstone type and it should therefore be the only one to enter in the large-scale description of a cholesteric.

2. LARGE-SCALE STATIC PROPERTIES

In principle, the procedure needed to obtain the large-scale characteristics of any system is well known and entails effective elimination of the rapid degrees of freedom. For cholesterics, this elimination is implemented in accordance with the definition

$$\exp\left(-\frac{1}{T}\int d^3rF_M\right)=\int D\tilde{n}\exp\left(-\frac{1}{T}\int d^3rE_F\right). \quad (13)$$

Here T is the temperature, F_M is the large-scale free energy of the cholesteric, and integration with respect to $\tilde{\mathbf{n}}$ means integration with respect to the director's rapid degrees of freedom, which contain Fourier harmonics with wave vectors $q \gtrsim L^{-1}$. Relations of type (13) are usually the starting point for taking fluctuation effects into account, as is the case in renormalization-group approach to the theory of phase transitions. In cholesterics, the fluctuations are weak and integration with respect to $\tilde{\mathbf{n}}$ is equivalent to minimizing E_F with respect to $\tilde{\mathbf{n}}$. This is usually a trivial procedure and reduces to substituting $\tilde{\mathbf{n}} = 0$ in E_F. In the case of cholesterics, however, the "slow" part of the director (2) contains rapidly oscillating factors, causing coupling of fast and slow degrees of freedom. Minimization of E_F with respect to $\tilde{\mathbf{n}}$ becomes therefore a nontrivial task.

By virtue of the condition $\mathbf{n}^2 = 1$ the director **n** contains two rapid degrees of freedom,[1] which we shall define by angles θ and φ containing Fourier harmonics with wave vectors $q \gtrsim L^{-1}$. An expression for the director in terms of these angles is

$$\mathbf{n}+\tilde{\mathbf{n}}=[\mathbf{u}\cos(\Phi+\varphi)+\mathbf{u}'\sin(\Phi+\varphi)]\cos\theta+\mathbf{l}\sin\theta. \quad (14)$$

We retain the symbol **n** for the slow part of the director (2). Note that Φ and φ in (14) differ in that Φ is the slow and φ the rapid variable. In the approximation linear in φ and θ, the only one we shall need hereafter, we get

$$\tilde{\mathbf{n}}=[\mathbf{nl}]\varphi+\mathbf{l}\theta. \quad (15)$$

We note that θ does not contain a term of the form

$$\psi_i n_i, \quad (16)$$

where ψ_i is a slow function. The point is that such a term can be eliminated by the following redefinition of the unit vectors in (3):

$$\delta\mathbf{u}=[[\mathbf{l}\boldsymbol{\psi}]\mathbf{u}], \quad \delta\mathbf{u}'=[[\mathbf{l}\boldsymbol{\psi}]\mathbf{u}']. \quad (17)$$

It follows from (3) and (8) that in equilibrium

$$s_i=q_0 l_i, \quad (18)$$

where l_i is a unit vector. Thus, the large-scale deviations from the equilibrium state of a cholesteric are characterized by the quantities

$$s_\parallel-q_0, \quad \mathbf{s}_\perp. \quad (19)$$

Here $s_\parallel$ and $\mathbf{s}_\perp$ are the **s**-components parallel and perpendicular to **l**. To calculate the direct contribution, due to the presence of the nonzero quantities (19), to the free energy F we must express the derivatives of **n** in (1) in accordance with (17) and average the results over scales $\sim L$. This averaging is carried out with the aid of the relations

$$\overline{n_in_j}={}^1/_2\delta_{ij}^\perp, \quad \delta_{ij}^\perp=\delta_{ij}-l_il_j, \quad (20)$$

$$\overline{n_in_jn_mn_n}={}^1/_8(\delta_{ij}^\perp\delta_{mn}^\perp+\delta_{im}^\perp\delta_{jn}^\perp+\delta_{in}^\perp\delta_{jm}^\perp). \quad (21)$$

These operations yield

$$\begin{aligned}F={}&{}^1/_4(K_1+K_3)s_\perp^2+{}^1/_2K_1[\mathbf{sl}]\nabla_\parallel\mathbf{l}\\&+{}^1/_2K_2(s_\parallel-q_0)^2+{}^1/_8(K_3-K_2)(\nabla_il_i)^2\\&+{}^1/_{16}(3K_2+K_3)(\nabla_il_n)^2+{}^1/_{16}(4K_1-3K_2-K_3)(\nabla_\parallel l_i)^2\\&\qquad+{}^1/_2K_2(s_\parallel-q_0)\varepsilon_{mjn}l_m\nabla_jl_n. \quad (22)\end{aligned}$$

Here and henceforth

$$\nabla_\parallel=l_i\nabla_i. \quad (23)$$

The linear term of the expansion of E_F [Eq. (1)] in terms of θ is, in the leading approximation,

$$K_1\nabla_\parallel\theta([\mathbf{nl}]_is_i-n_j\nabla_\parallel l_j)+K_3q_0\theta s_in_i. \quad (24)$$

The mean value of this expression is zero, since θ does not contain terms of the form (16). Thus θ is not coupled with the slow degrees of freedom, and minimization with respect to θ reduces to the substitution $\theta = 0$. The leading terms of the expansion of E_F in terms of φ are

$$E_F={}^1/_2K_2(\nabla_\parallel\varphi)^2+K_2(s_\parallel-q_0+[\mathbf{nl}]_jn_n\nabla_jl_n)\nabla_\parallel\varphi. \quad (25)$$

It follows from the structure of this expression that the slow degrees of freedom are linked with φ components of the form

$$\varphi=\varphi_{ik}n_in_k, \quad \varphi_{ik}=\varphi_{ki}, \quad \varphi_{ii}=0, \quad \varphi_{ik}l_k=0, \quad (26)$$

where φ_{ik} is a slow function. Substituting (26) in (25), retaining in $\nabla_\parallel\varphi$ the leading order, and averaging, we get

$$\bar{E}_\varphi={}^1/_2K_2q_0\varphi_{ij}\nabla_il_j+{}^1/_2K_2q_0^2\varphi_{ij}^2. \quad (27)$$

Minimization of $\bar{E}_\varphi$ with respect to φ_{ij} yields the following contribution to the free energy:

$$E_\psi=-{}^1/_{16}K_2[(\nabla_il_j)^2+(\nabla_il_j)(\nabla_jl_i)-(\nabla_il_i)^2-(\nabla_\parallel l_j)^2]. \quad (28)$$

To obtain the large-scale free energy of the cholesteric, we must add expressions (28) to expression (22).

It follows from (22) that the degrees of freedom connected with **l** are not of the Goldstone type and should be excluded from consideration. In the approximation in L/λ of interest to us we can neglect the second term in the right-hand side of (11), so that minimizing the free energy with respect to **l** reduces to minimization at constant $s_i = \nabla_i W$. The minimization yields, in the leading approximation in L/λ,

$$l_i=\nabla_iW/|\nabla W|, \quad (29)$$

$$\mathbf{s}_\perp=[K_1/(K_1+K_3)][\nabla_\parallel\mathbf{l}, \mathbf{l}]. \quad (30)$$

The end result of the minimization is replacement of the first two terms of (22) by

$$-[K_1^2/4(K_1+K_3)](\nabla_\| \mathbf{l})^2.$$

As for the remaining terms with **l** in (22) and (28), they are small corrections. They should therefore be retained simply with l_i replaced by (29). This replacement causes $\varepsilon_{ijn} l_i \nabla_j l_n$ to vanish, and with it the last term of (22). Collecting and transforming the remaining terms of (22) and (28), we ultimately get

$$F_W=(K_2/8q_0^2)[(\nabla W)^2-q_0^2]^2+(3K_3/16q_0^2)(\nabla^2 W)^2+\dots \quad (31)$$

The ellipsis stands here for inessential terms proportional to $(\nabla_\| \nabla W)^2$. The first term of (31) was transformed with allowance for the fact that $|\nabla W| \approx q_0$.

Expression (31) describes the elasticity theory for a smectic, with moduli

$$B=K_2 q_0^2, \quad K={}^3/_8 K_3. \quad (32)$$

The terms proportional to $(\nabla_\| \nabla W)^2$ [the ellipsis in (31)] play no role whatever in smectics; this becomes particularly clear when the long-wave fluctuations of the latter are considered.[6,7]

We conclude thus that from the standpoint of large-scale statics a cholesteric is indeed the equivalent of a smectic. Expressions (32) for the moduli of this smectic were obtained by de Gennes, who postulated the foregoing statement and compared the energies of a smectic and a cholesteric for two particular director configurations.[1]

3. HYDRODYNAMICS OF CHOLESTERICS

The hydrodynamic variables of cholesteric are, as for nematics, the director **n** and the densities of the energy E, of the mass ρ, and of the momentum j_i. These quantities are connected with the density of the entropy S by the following thermodynamic identity

$$dE=TdS+\mu d\rho+\mathbf{v}d\mathbf{j}+h_i dn_i+\nabla_k(\Phi_{kn} dn_n). \quad (33)$$

Here μ is the chemical potential, $\mathbf{v}=\mathbf{j}/\rho$ is the velocity, and

$$h_i=(\partial E/\partial n_j-\nabla_k \partial E/\partial \nabla_k n_j)(\delta_{ij}-n_i n_j), \quad (34)$$

$$\Phi_{ki}=(\partial E/\partial \nabla_k n_j)(\delta_{ij}-n_i n_j). \quad (35)$$

The last factor in (34) and (35) stems from the fact that the longitudinal parts of h_i and Φ_{ki} have no physical meaning in view of the identity $n_i dn_i=0$.

The pressure P is defined as usual as

$$P=\mu\rho+TS+\mathbf{vj}-E. \quad (36)$$

The thermodynamic identity for the pressure is

$$dP=SdT+\rho d\mu+\mathbf{j}d\mathbf{v}-h_i dn_i-\nabla_j(\Phi_{jn} dn_n). \quad (37)$$

The system of nonlinear hydrodynamic equations is of the same form for a cholesteric as for a nematic[2)]:

$$\partial\rho/\partial t+\nabla \mathbf{j}=0, \quad (38)$$

$$\partial j_i/\partial t+\nabla_j T_{ij}-\nabla_j(\eta_{ijnm}\nabla_n v_m)=0, \quad (39)$$

$$\partial n_i/\partial t+f_i+h_i/\gamma_1=0, \quad (40)$$

$$\partial E/\partial t+\nabla_i Q_i-\nabla_k(v_i \eta_{iknm}\nabla_n v_m+T\varkappa_{ik}\nabla_i T-\Phi_{ki}h_i/\gamma_1)=0. \quad (41)$$

The reactive terms in (39)–(41) are defined by the expressions

$$T_{ij}=\rho v_i v_j+P\delta_{ij}+\Phi_{jm}\nabla_i n_m+{}^1/_2(1-\lambda)n_i h_j-{}^1/_2(1+\lambda)h_i n_j, \quad (42)$$

$$f_i=v_j\nabla_j n_i+{}^1/_2(\nabla_i v_j-\nabla_j v_i)n_j-{}^1/_2\lambda(\delta_{im}-n_i n_m)n_j(\nabla_j v_m+\nabla_m v_j), \quad (43)$$

$$Q_i=(P+E)v_i-{}^1/_2(1+\lambda)n_i(\mathbf{hv})+{}^1/_2(1-\lambda)h_i(\mathbf{nv})+\Phi_{ij}f_j. \quad (44)$$

Here λ is the reactive coefficient. The divergence of the stress tensor (42) can be reduced to a divergence of a symmetrical stress tensor,[8] thereby ensuring conservation of the angular momentum. The dissipative terms in (39)–(41) are determined by kinetic coefficients, viz., the torsional viscosity γ_i, the thermal-conductivity tensor

$$\varkappa_{ik}=\varkappa_0\tilde\delta_{ik}+\varkappa_1 n_i n_k, \quad \tilde\delta_{ik}=\delta_{ik}-n_i n_k \quad (45)$$

and the viscosity tensor

$$\begin{aligned}\eta_{iklm}=&\eta_1(\tilde\delta_{il}\delta_{km}+\delta_{kl}\tilde\delta_{im})+(2\eta_2-\eta_1)\tilde\delta_{ik}\tilde\delta_{lm}\\ &+2\eta_3(n_i n_k\tilde\delta_{lm}+n_l n_m\tilde\delta_{ik})\\ &+{}^1/_2\eta_4(n_i n_l\tilde\delta_{km}+n_k n_l\tilde\delta_{im}+n_i n_m\tilde\delta_{kl}+n_k n_m\tilde\delta_{il})+2\eta_5 n_i n_k n_l n_m.\end{aligned} \quad (46)$$

The energy E of a cholesteric can be expressed to the required accuracy in the form

$$E=\mathbf{j}^2/2\rho+E_0(\rho,\sigma)+E_F, \quad (47)$$

where $\sigma=S/\rho$ is the specific entropy, and E_F is given by Eq. (1). Rewriting now in terms of pressure in accordance with (36), we obtain to lowest approximation

$$P=P_0+K_2\gamma(\varepsilon_{ijn}n_i\nabla_j n_n-q_0)q_0, \quad (48)$$

where

$$P_0=\rho(\partial E_0/\partial\rho)_\sigma-E_0, \quad \gamma=-(\partial \ln q_0/\partial \ln \rho)_\sigma. \quad (49)$$

Calculating Φ_{ij} and h_i in accordance with the definitions (34) and (35), we get

$$\Phi_{ij}=K_1\nabla\mathbf{n}\tilde\delta_{ij}+K_3 n_i n_m\nabla_m n_j+K_2\varepsilon_{ijm}n_m(\varepsilon_{kln}n_k\nabla_l n_n-q_0), \quad (50)$$

$$\begin{aligned}h_i=\tilde\delta_{ij}[&-K_1\nabla_j\nabla\mathbf{n}+K_3\nabla_j n_l n_k\nabla_k n_l-K_3\nabla_l n_l n_m\nabla_m n_j\\ &+2K_2(\varepsilon_{pqr}n_p\nabla_q n_r-q_0)\cdot\varepsilon_{jkm}\nabla_k n_m-K_2\nabla_m(\varepsilon_{pqr}n_p\nabla_q n_r-q_0)\cdot\varepsilon_{lmj}n_l].\end{aligned} \quad (51)$$

Finally, in accordance with identity (33) we obtain for the temperature, to lowest order

$$T=T_0-\rho^{-1}K_2 q_0(\partial \ln q_0/\partial\sigma)\cdot(\varepsilon_{ijm}n_i\nabla_j n_m-q_0), \quad (52)$$

where

$$T_0=\rho^{-1}(\partial E_0/\partial\sigma)_\rho. \quad (53)$$

All the coefficients in (48) and (50)–(52) are functions of ρ and σ, whereas to close the system (38)–(41) they should be expressed in terms of ρ and E. To the required accuracy, the specific entropy σ can be expressed in terms of E, using the relation $E = E_0(\rho, \sigma)$, without affecting the dependence of expressions (48) and (50)–(52) on the director.

4. LARGE-SCALE DYNAMICS

The equations of large-scale dynamics of cholesterics can, of course, not be obtained by averaging the equations given above over scales $\sim L$. We shall derive these equations by a procedure of eliminating the fast variables, similar to that used in the static case. We must integrate over the fast variables the distribution function $\exp(iI)$ that permits the dynamic correlators of the system to be calculated, just as the distribution function $\exp(-F/T)$ has enabled us to calculate the static correlators. The effective action for such a distribution function was constructed in Refs. 8 and 9 (see also Refs. 10 and 11). The action I includes a dependence both on the hydrodynamic variables and on the auxilliary fields, with both Bose and Fermi parts, but we shall not need the Fermi part of the action. The nonlinear hydrodynamic equations can be obtained, as usual, as extremals of I.

The effective action I for cholesterics can be constructed from Eqs. (38)–(41) in accordance with the procedure of Refs. 8 and 9. The Bose part of this action is

$$I = \int dt\, d^3r \mathscr{L}, \tag{54}$$

where the Lagrangian density $\mathscr{L}$ takes, to our specified accuracy, the form

$$\begin{aligned}\mathscr{L} = {} & p_i \partial j_i/\partial t - \nabla_k p_i T_{ik} + \nabla_k p_i \eta_{iknm} \nabla_n v_m + iT\eta_{iknm} \nabla_k p_i \nabla_n p_m \\ & + y_i \frac{\partial n_i}{\partial t} + y_i f_i + y_i h_i/\gamma_1 + iTy_i^2/\gamma_1 + p\frac{\partial E}{\partial t} \\ & - \nabla_i p Q_i + \nabla_i p T \varkappa_{ik} \nabla_k T + iT^3 \varkappa_{ik} \nabla_i p \nabla_k p.\end{aligned} \tag{55}$$

Here p_i, p, and y_i are auxilliary Bose fields; p_i is conjugate to j_i, p is conjugate to E, and y_i is conjugate to n_i; furthermore y_i must satisfy the orthogonality condition

$$y_i n_i = 0. \tag{56}$$

We can accordingly always expand y_i in terms of the vectors $\mathbf{l}$ and $\mathbf{n} \times \mathbf{l}$:

$$y_i = y[\mathbf{nl}]_i + y_\parallel l_i, \quad y_i^2 = y^2 + y_\parallel^2. \tag{57}$$

We have left out of (55) the dependence on ρ, since Eq. (38) can be regarded as an expression for ρ in terms of j.

We must now separate in (55) the contribution of the rapid degrees of freedom. We specify these degrees for the director, as before, by means of the angles φ and θ defined in Eqs. (14) and (15). All the other variables in (55), viz., p_i, $j_i = \rho v_i$, y, $y_\parallel$, p, E, simply break up into sums of slow and rapid parts (the latter will be marked with tildes). The effective action I_M, which describes the large-scale dynamics of cholesterics, can be obtained now by analogy with (13) by integrating over the rapid degrees of freedom:

$$\exp(iI_M) = \int D\varphi\, D\theta\, D\tilde{p}_i\, D\tilde{j}_i\, D\tilde{y}\, D\tilde{y}_\parallel\, D\tilde{p}\, D\tilde{E} \exp(iI). \tag{58}$$

Just as in statics, fluctuations play no role whatever in the dynamics of cholesterics. The integration in (58) reduces therefore to a search for the extremum of I with respect to all the variables listed in (58). This search is not trivial since, just as in statics, the slow part of the director (2) contains rapidly oscillating factors, so that the rapid and slow degrees of freedom become linked.

It is easy to verify that in view of the presence of the term $-c^2\nabla_i \tilde{p}_i \tilde{p}$ (where c is the speed of sound) in the Lagrangian density (55) the integration over the rapid acoustic degrees of freedom makes negligible contribution to I_M. We therefore disregard these degrees of freedom and put

$$\nabla_i \tilde{p}_i = \nabla_i \tilde{v}_i = 0. \tag{59}$$

It is also easy to verify that, just as in statics, θ is not coupled with the slow degrees of freedom, so that evaluating the extremum with respect to θ reduces to the condition $\theta = 0$. As for the remaining fast variables, their components coupled with the slow variables are

$$y_\parallel = \beta_i n_i, \quad \tilde{y} = m_{ik} n_i n_k, \tag{60}$$

$$\begin{aligned}\tilde{p}_i = {} & -(1/2q_0)(n_i[\mathbf{nl}]_j + n_j[\mathbf{nl}]_i)\xi_j \\ & + (1/8q_0^2) l_i (n_j n_k - [\mathbf{nl}]_j [\mathbf{nl}]_k) \cdot (\nabla_j \xi_k + \nabla_k \xi_j),\end{aligned} \tag{61}$$

$$\tilde{v}_i = -(1/2q_0)(n_i[\mathbf{nl}]_j + n_j[\mathbf{nl}]_i)\chi_j. \tag{62}$$

Here β_i, m_{ik}, ξ_i, χ_j are slow functions. For φ we have Eq. (26) as before. The tensor m_{ik} in (60) has the same properties as φ_{ik} in (26), and for the remaining quantities we have

$$\beta_i l_i = \xi_i l_i = \chi_i l_i = 0. \tag{63}$$

The second term is included in (61) to satisfy the condition $\nabla_i \tilde{p}_i = 0$ accurate to first order in L/λ (we need no such term in $\tilde{v}_i$).

We must now substitute the rapid parts of (15), (26), and (60)–(62) in the Lagrangian density (55), express the derivatives of the slow part of the director with the aid of (6) and (7), and average the result over scales $\sim L$ with the aid of (20) and (21). Integration of $\exp(iI)$ with the resultant Lagrangian density over the variables φ_{ik} and χ_i and with respect to the non-Goldstone variable[3] l_i, gives rise to the appearance of functional δ-functions that are eliminated by integrating with respect to the variables m_{ik}, β_i, and ξ_i. All these transformations are quite cumbersome and we leave out their details. The calculations yield the large-scale action I_M and the Lagrangian density consisting of a reactive part $\mathscr{L}_r$ and a dissipative part $\mathscr{L}_d$.

To lowest order, we have for the reactive part of the Lagrangian density

$$\begin{aligned}\mathscr{L}_r = {} & y(\partial W/\partial t + q_0 \mathbf{lv}) + p\partial E/\partial t - v_i \nabla_i p(E + P_0) + p_i \frac{\partial j_i}{\partial t} \\ & - \nabla_i p_i \{P_0 + {}^1/_2 K_2 \rho\gamma[(\nabla W)^2 - q_0^2]\} - {}^1/_2 K_2 \nabla_k p_i \\ & \times [(\nabla W)^2 - q_0^2] l_i l_k + {}^3/_8 K_3 \nabla_k p_i l_i \nabla^2 l_k.\end{aligned} \tag{64}$$

Here l_i is given by (29). We note that the variable $y_\parallel$ has dropped out completely, and also that we have discarded in (64) the terms proportional to $\nabla_\parallel^2 \mathbf{l}$ and $\nabla_\parallel \nabla \mathbf{l}$ (cf., the discussion in the static case). As for the dissipative part of the Lagrangian function, it is given in leading order by

$$
\begin{aligned}
\mathscr{L}_d=&-(K_2/2q_0\gamma_1)\,y\nabla_\parallel[(\nabla W)^2-q_0^2]+i(T/\gamma_1)y^2+\nabla_i pT\overline{\varkappa}_{ik}\nabla_k T_1\\
&+iT^3\overline{\varkappa}_{ik}\nabla_i p\nabla_k p+\nabla_k p_i\overline{\eta}_{iknm}\nabla_n v_m+iT\nabla_k p_i\overline{\eta}_{iknm}\nabla_n p_m\\
&+{}^1/_{16}\lambda^2\gamma_1\nabla_k p_i\cdot(\delta_{ij}{}^\perp\delta_{kn}{}^\perp+\delta_{in}{}^\perp\delta_{kj}{}^\perp)(\nabla_n v_j+iT\nabla_n p_j)\\
&-\eta_6\nabla_h p_i(\delta_{in}{}^\perp l_k l_m\\
&+\delta_{im}{}^\perp l_k l_n+\delta_{kn}{}^\perp l_i l_m+\delta_{km}{}^\perp l_i l_n)(\nabla_n v_m+iT\nabla_n p_m).
\end{aligned}\tag{65}
$$

Here

$$\overline{\varkappa}_{ik}={}^1/_2(\varkappa_0+\varkappa_1)\delta_{ik}{}^\perp+\varkappa_0 l_i l_k,\tag{66}$$

$$
\begin{aligned}
\overline{\eta}_{iklm}=&(2\eta_2+\eta_1)l_i l_k l_l l_m+{}^1/_2(2\eta_3+2\eta_2-\eta_1)[l_i l_k\delta_{lm}{}^\perp+l_l l_m\delta_{ik}{}^\perp]\\
&+{}^1/_2({}^1/_2\eta_4+\eta_1)(l_i l_l\delta_{km}{}^\perp+l_k l_l\delta_{im}{}^\perp+l_i l_m\delta_{kl}{}^\perp+l_k l_m\delta_{il}{}^\perp)\\
&+({}^1/_4\eta_5+{}^7/_4\eta_2-{}^1/_4\eta_4+{}^1/_8\eta_1)\delta_{ik}{}^\perp\delta_{lm}{}^\perp\\
&+({}^1/_4\eta_5-{}^1/_2\eta_3+{}^1/_4\eta_4+{}^1/_4\eta_2+{}^1/_8\eta_1)(\delta_{il}{}^\perp\delta_{km}{}^\perp+\delta_{im}{}^\perp\delta_{kl}{}^\perp),
\end{aligned}\tag{67}
$$

$$\eta_6=\frac{8({}^1/_2\eta_4-\eta_1)^2+(1-\lambda)^2\gamma_1({}^1/_2\eta_4-5\eta_1)}{16({}^1/_2\eta_4+\eta_1)+2(1-\lambda)^2\gamma_1},\tag{68}$$

$$T_1=T_0-\frac{1}{2\rho}K_2\frac{\partial\ln q_0}{\partial\sigma}[(\nabla W)^2-q_0^2]=\frac{\partial F_M}{\partial S}.\tag{69}$$

The structure of (65) is such that it leads automatically to satisfaction of the fluctuation-dissipation theorem (cf., Refs. 8 and 9). Note that in the derivation of (65) we have included in $\mathscr{L}_d$ a term $-(1/2)\beta_i\partial l_i/\partial t$, having transformed the derivative of $\mathbf{l}$ by using the reactive equation $W=-q_0\mathbf{l}\cdot\mathbf{v}$ and the representation (29).

Expression (64) with allowance for (29) agrees fully with the reactive part of the Lagrangian obtained in Ref. 12, and accords with smectic dynamics with the same smectic moduli (32) as obtained in statics. Thus, from the standpoint of large-scale dynamics, a cholesteric is equivalent to a smectic whose dynamics is consistent with the expression for its free energy. Expression (64) is connected with the dissipative part of the hydrodynamic equations and permits the large scale viscosity, heat-conduction, and permeation coefficients.

The hydrodynamic equations corresponding to the extremals I are of the form

$$
\begin{aligned}
\partial E/\partial t=&-\nabla_i[(P+E)v_i+M_i(v_k\nabla_k W)+M_{ik}\nabla_k(v_n\nabla_n W)]\\
&+\nabla_k[v_i\partial R/\partial\nabla_k v_i\\
&+T\partial R/\partial\nabla_k T+M_{ik}\nabla_i\partial R/\partial\nabla_j M_j+M_k\partial R/\partial\nabla_j M_j],
\end{aligned}\tag{70}
$$

$$\partial j_i/\partial t=-\nabla_k T_{ik}+\nabla_k\partial R/\partial\nabla_k v_i,\tag{71}$$

$$\partial W/\partial t=-\mathbf{v}\nabla W+\partial R/\partial\nabla_j M_j,\tag{72}$$

where we have used the notation

$$
\begin{gathered}
M_i={}^1/_2K_2[q_0^{-2}(\nabla W)^2-1]\nabla_i W-{}^3/_8\nabla_n(K_3/q_0^2)\nabla_n\nabla_i W,\\
M_{ij}={}^3/_8(K_3/q_0^2)\nabla_i\nabla_j W,\\
T_{ij}=P\delta_{ij}+\rho v_i v_j+M_j\nabla_i W+M_{jn}\nabla_n\nabla_i W,
\end{gathered}
$$

T_{ij} is the stress tensor, P is the pressure, and we have introduced the large-scale dissipative function

$$
\begin{aligned}
R=&{}^1/_2\overline{\varkappa}_{ij}\nabla_i T\nabla_j T+{}^1/_2\overline{\eta}_{ijnm}\nabla_j v_i\nabla_n v_m-{}^1/_2\eta_6[\nabla_\parallel v_i\nabla_i v_\parallel\\
&+(\nabla_i v_\parallel)^2+(\nabla_i v_\parallel\nabla_\parallel v_i)-4(\nabla_\parallel v_\parallel)^2]\\
&+\xi_1(\nabla_i M_i)^2-(\xi_2 q_0/T)l_i\nabla_i T\nabla_j M_j.
\end{aligned}
$$

In the large-scale limit we get thus the usual hydrodynamic equations for smectics.[3,12] The coefficient ξ_1 describes a phenomenon known as percolation,[1] i.e., flow of a layered system (smectic or cholesteric) with constrained layers. In accordance with (65) and (72), the percolation coefficient is $\xi_1=1/\gamma_1 q_0^2$ (the physical meaning of the terms with the kinetic coefficient ξ_2 will be discussed below).

The large-scale viscosity coefficients $\eta_i^{(M)}$ are expressed according to (65), (67) and (68) in terms of the small-scale bare viscosities of the cholesterics:

$$
\begin{gathered}
\eta_5^{(M)}={}^1/_2(2\eta_2+\eta_1),\qquad \eta_3^{(M)}={}^1/_4(2\eta_3+2\eta_2-\eta_1),\\
\eta_4^{(M)}={}^1/_2\eta_4+\eta_1-2\eta_6,\\
\eta_1^{(M)}={}^1/_4\eta_5-{}^1/_2\eta_3+{}^1/_4\eta_4+{}^1/_4\eta_2+{}^1/_8\eta_1+{}^1/_{16}\lambda^2\gamma_1,\\
\eta_2^{(M)}={}^1/_2\eta_5+2\eta_2-{}^1/_2\eta_3+{}^1/_4\eta_1.
\end{gathered}
$$

It is important to note that the large-scale viscosities are independent of the pitch $2\pi/q_0$ of the cholesteric helix. This conclusion can be experimentally verified by measuring the dependence of the viscosity on the concentration of the optically active additive in ordered nematic-cholesteric mixtures.

Unfortunately, there are at present for the viscosity coefficients of cholesterics nowhere near complete data, for any scale, to compare with the equations presented above. One can only expect from general considerations the coefficients η_1 and η_2 to be somewhat larger than the remaining ones (since they are not related to the presence of liquid-crystal order). It follows therefore, in the large-scale limit, that the coefficient $\eta_5^{(M)}$, expressed in terms of "large" bare viscosities, must be relatively large. No such increase is observed for the coefficient $\eta_4^{(M)}$, in which the "large" viscosity coefficient is canceled out by the term, η_6.

Let us discuss some of the terms left out above. Thus, we have disregarded the term $(1/2)\,y\varepsilon_{nij}l_n\nabla_i v_j$, that stems from $y_i f_i$ and leads to the appearance, in the right-hand side of (72), of a term

$$-{}^1/_2\varepsilon_{nij}l_n\nabla_i v_j.\tag{73}$$

Accordingly, account must be taken in the right-hand side of (71) of the omitted term

$${}^1/_2\varepsilon_{inj}\nabla_j\{l_n\nabla_m[(K_2/2q_0)\ln((\nabla W)^2-q_0^2)]\}.\tag{74}$$

We have also omitted above from the equations for the momentum density and the director the reactive purely cholesteric terms (i.e., those which are not invariant under spatial inversion). Thus, Eq. (40) may acquire a term proportional to

$$\nabla_n v_j(\varepsilon_{jlm}\nabla_m n_n+\varepsilon_{nlm}\nabla_m n_j)(\delta_{li}-n_i n_l).\tag{75}$$

An anlogous term should be added also to Eq. (39) for j_i. Averaging of term (75) and also of many others of like structure adds to the right-hand side of (72) a term proportional to (73), while averaging of a corresponding term to the equation for j_i adds to the right-hand side of (71) a term proportional to (74). The terms (73) and (74), multipled by certain coefficients, enter thus in the right-hand sides of (72) and (71). Although these terms are of the same order in L/λ as the dissipative terms in (71) and (72), it is easy to verify that they entail negligible corrections to the spectrum of the modes obtained in the large-scale theory, and can indeed be omitted.

Besides these purely cholesteric reactive terms, we have also left out some purely cholesteric dissipative terms. We shall need only the pair of such terms with coefficient ξ_2, which must be added to the right-hand sides of Eqs. (40) and (41), respectively:

$$-\xi_2 q_0 \varepsilon_{ijn} n_j \nabla_n T, \qquad \nabla_m (T \xi_2 q_0 \varepsilon_{jim} n_j h_i).$$

These are precisely the terms that generate the kinetic terms we included in (70) and (72) and containing the coefficient ξ_2 that has in the large-scale theory the meaning of the crossover thermomechanical percolation coefficient.

5. CHIRAL SMECTIC *C* (SMECTIC *C**)

A helicoidal structure similar to that of cholesterics is possessed also by the smectics known as chiral C and usually designated C^*. Besides spontaneous violation of rotational invariance, smectics are known to be subject also to violation of translational invariance along one of the directions, and the result is a layered structure. The unit vector normal to the layer, which we designate **l** in the present section, is not collinear in smectic C with the director **n**. Smectic layers are therefore not iosotropic: they have a preferred direction which we designate by the unit vector

$$\mathbf{v}=[\mathbf{nl}]/|[\mathbf{nl}]|, \quad \mathbf{v}^2=1, \ \mathbf{vl}=0. \tag{76}$$

Smectics C^*, just as cholesterics, consist of molecules that have no inversion centers. As a result they have in the ground state, in analogy with (2),

$$\mathbf{v}=\mathbf{u}\cos(q_0\mathbf{lr})+\mathbf{u}'\sin(q_0\mathbf{lr}), \tag{77}$$

where **u** and **u**′ are mutually orthogonal unit vectors that are also orthogonal to **l**.

We shall describe smectic ordering with the aid of a function W similar to the one introduced above for the large-scale description of a cholesteric, and having the meaning of the density-wave phase in smectics. By virtue of its definition, the function W is such that the equation $W=\text{const}$ describes the position of a certain smectic layer in space, such that the vector ∇W is normal to the layer. Hence it follows that

$$\mathbf{l}=\nabla W/|\nabla W|. \tag{78}$$

In view of the conditions (76) imposed on it, the vector $\mathbf{v}$ has one degree of freedom, so that its variation can always be represented in the form

$$\delta v_i=-(l_i/|\nabla W|)\mathbf{v}\nabla\delta W+{}^1/_2[\mathbf{vl}]_i\delta\varphi. \tag{79}$$

The angle φ introduced in this manner is a nonholonomic variable, so that only its variation is meaningful. For the commutator of the variations it is easy to obtain from (79)

$$[\delta_1, \ \delta_2]\varphi=(2/|\nabla W|^2)\varepsilon_{nik} l_n \nabla_i \delta_1 W \nabla_k \delta_2 W. \tag{80}$$

The angle φ is the cause of the additional Goldstone mode considered in Ref. 13 and present in smectics C.

In real smectics C the director makes a small angle with the normal **l** to the layer. Hereafter we regard this inclination angle θ_0 as a small parameter. With allowance for this smallness, we can confine ourselves to the following terms of the expansion of the free energy that describes the smectic and orientational orders:

$$F={}^1/_8 B[q_s^{-2}(\nabla W)^2-1]^2+(1/2|\nabla W|^2)K_{ikmn}\nabla_i\nabla_k W\nabla_m\nabla_n W$$
$$+{}^1/_2\alpha_{ik}\left(\nabla_i\varphi-2\frac{q_0}{q_s}\nabla_i W\right)\left(\nabla_k\varphi-2\frac{q_0}{q_s}\nabla_k W\right), \tag{81}$$

where

$$K_{ikmn}=K_1\delta_{ik}^{\perp}\delta_{mn}^{\perp}+K_3\delta_{im}^{\perp}l_k l_n, \quad \delta_{ik}^{\perp}=\delta_{ik}-l_i l_k, \tag{82}$$

$$\alpha_{ik}=\alpha_1[\mathbf{vl}]_i[\mathbf{vl}]_k+\alpha_2 v_i v_k+\alpha_3 l_i l_k. \tag{83}$$

Here, B, K_1, and K_3 are moduli characterizing the smectic ordering, and K_1, $K_3 \sim \rho c^2 a^2$, where ρ is the density, c the speed of sound, and a the atomic dimension. $B/\rho c^2$ is a small quantity (usually 10^{-3}–10^{-2}), meaning that the smectics are close to a point of second-order transition into the nematic state. The quantities α_{ik} can be naturally estimated at $\alpha \sim \rho c^2 a^2 \theta_0^2$. The wave vector q_s in (81) represents the thickness of the smectic layer and is of the oder of 10^7–10^8 cm^{-1}, while the vector q_0 represents in accordance with (77) the pitch of the helix in smectics C^*. The ratio q_0/q_s is the small parameter of the problem and is a measure of the weak perturbation of the symmetry center.

An important feature of smectics C^* is the presence of dipole (besides the smectic and orientational) ordering. This ordering is allowed just in smectic C^* and is due, first, to the angle made by the molecules with **l** and, second, to their chirality (i.e., to the absence of inversion centers). The dipole moment produced in smectics C^* is directed along the vector v introduced above [see Eq. (76)]. In view of the presence of the dipole moment, it is necessary to take into account in the free energy, besides the local term (81), also a nonlocal dipole term, which we write in the form

$$F_d=-2\mu(\nabla_i v_i)\nabla^{-2}(\nabla_j v_j). \tag{84}$$

Here μ is a constant of order $\rho c^2(q_0 a)^2\theta_0^2$.

To obtain the large-scale free energy of smectics C^* we must follow the procedure described above for cholesterics. The free energy (81) is easily averaged over scales $\sim q_0^{-1}$ with the aid of the relation

$$\overline{v_i v_k}={}^1/_2\delta_{ik}^{\perp}. \tag{85}$$

Before we average the dipole term (84), we note that if $\nabla_i v_i$ is expressed in accordance with (79) it is easily seen that ∇^{-2} can be replaced by $-q_0^{-2}$. This makes the averaging trivial. It can be readily estimated that the only substantial term from among the resultant ones reduces to a renormalization of α_{ik}. A direct verification shows that elimination of the fast degrees of freedom $\widetilde{W}$ and $\widetilde{\varphi}$ makes no substantial contribution to the large-scale free energy. The large-scale free energy of smectic C^* coincides with (81), where now

$$\alpha_{ik}=\alpha_{\perp}\delta_{ik}^{\perp}+\alpha_3 l_i l_k, \quad \alpha_{\perp}={}^1/_2(\alpha_1+\alpha_2+\mu/q_0^2). \tag{86}$$

Nonlinear dynamic equations for smectics C were obtained in Ref. 13 using the Poisson-bracket formalism. With an insignificant modification due to the presence of the dipole term (84), they are suitable also for smectics C^*. These equations for W, φ, and the momentum density $\mathbf{j}$ are:

$$\frac{\partial W}{\partial t}=-\mathbf{v}\nabla W-\xi_1|\nabla W|^2\frac{\delta E}{\delta W}-\xi_3|\nabla W|\frac{\delta E}{\delta\varphi}-\xi_2|\nabla W|\frac{l_i\nabla_iT}{T}, \tag{87}$$

$$\frac{\partial\varphi}{\partial t}=-v_i\nabla_i\varphi-\varepsilon_{jni}l_j\nabla_nv_i-\Gamma\frac{\delta E}{\delta\varphi}-\xi_3|\nabla W|\frac{\delta E}{\delta W}-\xi_4\frac{l_i\nabla_iT}{T}, \tag{88}$$

$$\partial j_i/\partial t=-\nabla_kT_{ik}+F_i+\nabla_k(\eta_{ikmn}\nabla_mv_n). \tag{89}$$

Here ξ_i, Γ, η_{ikmn} are kinetic coefficients, and in view of the smallness of θ_0 the tensor η_{ikmn} has the same structure as the viscosity tensor in cholesterics, subject to the obvious substitution $\tilde{\delta}_{ik}\to\delta_{ik}^{\perp}$ in (46). The stress tensor T_{ik} determined from the local part of the energy density E (see Ref. 13) is

$$\begin{aligned}T_{ik}=\rho v_iv_k&+\left(\rho\frac{\partial}{\partial\rho}-1\right)E\delta_{ik}+\frac{dE}{d\nabla_kW}\nabla_iW+\frac{\partial E}{\partial\nabla_k\nabla_nW}\nabla_i\nabla_nW\\&-\nabla_n\frac{\partial E}{\partial\nabla_n\nabla_kW}\nabla_iW+\frac{\partial E}{\partial\nabla_k\varphi}\nabla_i\varphi+\varepsilon_{ikn}l_n\nabla_m\frac{\partial E}{\partial\nabla_m\varphi}\\&+\varepsilon_{kjn}\frac{\partial E}{\partial\nabla_m\varphi}\frac{2}{|\nabla W|}\nabla_j\nabla_mWl_nl_i-\frac{1}{2}\varepsilon_{ikm}l_m[\mathbf{v}\mathbf{l}]_n\frac{\partial E}{\partial\mathrm{v}_n},\end{aligned} \tag{90}$$

where

$$\frac{dE}{d\nabla_kW}\equiv\frac{\partial E}{\partial\nabla_kW}-\frac{\partial E}{\partial\mathrm{v}_n}\frac{l_n}{|\nabla W|}\mathrm{v}_k. \tag{91}$$

The force F_i in the right-hand side of (89) is due to the presence of the dipole term (84):

$$F_i=\frac{\delta F_d}{\delta\mathrm{v}_k}\nabla_i\mathrm{v}_k+\nabla_k\left[\mathrm{v}_kl_il_m\frac{\delta F_d}{\delta\mathrm{v}_m}+\frac{1}{2}\varepsilon_{ikm}l_m[\mathbf{v}\mathbf{l}]_n\frac{\delta F_d}{\delta\mathrm{v}_n}\right]. \tag{92}$$

To obtain the large-scale dynamics of smectics C^* we must follow the procedure described by us for cholesterics. Averaging of the effective action over scales $\sim q_0^{-1}$ reduces in this case to averaging of Eqs. (87)–(89). It is obvious that averaging of (87) and (88) is equivalent to replacing E by $\overline{E}$ in these equations, and the latter coincides in accordance with the foregoing with the large-scale energy density. As for Eq. (89), its last term is not affected at all by the averaging. When T_{ik} [Eq. (90)] is averaged the term $dE/d\nabla_kW$ is replaced by $\partial\overline{E}/\partial\nabla_kW$, the last term of (90) vanishes, and the averaging in the remaining terms reduces to the substitution $E\to\overline{E}$. Allowances for the term F_i of (89), averaged in the same manner, leads to a final equation

$$\partial j_i/\partial t=-\nabla_kT_{ik}^{(M)}+\nabla_k(\eta_{ikmn}\nabla_mv_n). \tag{93}$$

The tensor $T_{ik}^{(M)}$ is determined here from the large-scale energy density by the procedure (90). It can be directly verified that elimination of the fast degrees of freedom leads to inessential corrections to the averaged equations; these corrections are small in the parameter k/q_0, where k is the characteristic wave vector.

Thus, from the large-scale standpoint, the entire difference between C and C^* smectics reduces to the presence of terms proportional to q_0 in the expansion of the free energy (81), and to the presence of the kinetic coefficients ξ_3, $\xi_4\propto q_0$ in Eqs. (87) and (88). These terms lead, in view of the smallness of q_0/q_s, to small efects. Therefore, in particular, the orientational-mode spectrum in smectics C^* has the same diffusive form as in ordinary smectics C:

$$\omega=-i\Gamma(\alpha_3q_z^2+\alpha_\perp q_\perp^2)[1+q_\perp^2/\Gamma(\eta_1q_\perp^2+{}^1/_2\eta_4q_z^2)]. \tag{94}$$

In the derivation of (94) we used the smallness of $\alpha\sim\theta_0^2$.

Thus, the situation in smectics C^* differs radically from the case of cholesterics, considered in the preceding sections, where the transition to large scales restructured the natural-mode spectrum (one propagating mode was obtained in lieu of the two small-sale diffusive nematic modes).

In conclusion, the authors thank G. E. Volovik for supplying his unpublished results on large-scale statics of cholesterics.

[1)] Note that the number of slow Goldstone degrees of freedom is not necessarily equal to the number of fast degrees of freedom.

[2)] This statement will be made more precise below.

[3)] Just as in statics, the second terms in the right-hand sides of (11) and (12) can be neglected here.

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Translated by J. G. Adashko