Orientational transitions in block-copolymer melts under shear flow
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Chapter 2

The lamellar phase under steady shear flow

In this chapter we develop the basic formalism which will serve us in explaining the orientational transitions. We consider the lamellar phase under steady shear flow. It is difficult to trace the origin of the idea underlying our theory. The earliest publication we are aware of is by de Gennes [50], who claims that the idea of reduction of the critical fluctuations by shear flow belongs to Bergé. Later, and possibly independently of the work [50], Onuki and Kawasaki discussed the same effect for binary mixtures [51]. The first application of these ideas to the orientational behaviour in block copolymers by Cates and Milner [53] was subsequently developed by Fredrickson [56]. There are several other works that contribute to our understanding of the phenomenon but do not deal with it explicitly [45, 119, 120, 55].

The key idea of the theory is that shear flow strongly interacts with the critical fluctuations near the ODT. In equilibrium such fluctuations are responsible for lowering the ODT temperature. Shear flow anisotropically suppresses the critical fluctuations. As a result, the ODT temperature also becomes spatially anisotropic, and the direction of the strongest suppression of the fluctuations corresponds to the highest ODT temperature. Thus, below the ODT we expect the lamellar phase to orient in this direction.

Our main task is to find a spectrum of the critical fluctuations in the presence of shear flow. In the following sections we introduce a simple dynamic equation for the order parameter and use it to derive an equation for the fluctuation intensity. This equation can be solved in the limits of high and low shear rates. For intermediate shear rates we interpolate in-between those two limits to obtain a cross-over from strong to weak shear. Then we discuss the influence of the viscosity contrast between different monomers on the stability of the orientations. Afterwards we relax the steady-state assumption and analyze the early stages of the orientational transition.

We want to note that the main results of Sections 2.1–2.3 and 2.5 were ob-
tained by G. Fredrickson [56].

2.1 Basic equations

To describe the dynamics of the order parameter we neglect as many details of the system behaviour as possible, leaving the only essential processes: diffusional relaxation and convection due to the flow. The simplest dynamical equation of this type is

$$\frac{\partial \phi}{\partial t} + \mathbf{v} \cdot \nabla \phi = -\hat{L} \frac{\delta H[\phi]}{\delta \phi} + \zeta$$

(2.1)

where \(\mathbf{v}\) is the velocity profile, \(H[\phi]\) is the Brazovskii Hamiltonian (eq.(B.3)), and \(\zeta\) is a noise term with the following properties

$$\langle \zeta(\mathbf{r}, t) \rangle = 0 \quad , \quad \langle \zeta(\mathbf{r}, t)\zeta(\mathbf{r}', t') \rangle = 2 \hat{L} \delta(\mathbf{r} - \mathbf{r'}) \delta(t - t')$$

(2.2)

and \(\hat{L}\) is an operator whose structure depends on the properties of the order parameter

$$\hat{L} = \left\{ \begin{array}{ll}
\gamma \nabla^2 & , \phi \text{ is conserved} \\
\mu & , \phi \text{ is not conserved}
\end{array} \right.$$ (2.3)

The equation (2.1) corresponds to models A and B in the notation of Hohenberg and Halperin [121] (see also [122]). We will see that the structure of the operator \(\hat{L}\) is irrelevant for our analysis: we will make use of the primary-harmonic approximation and set the typical length-scale in the system to \(k_0^{-1}\). Then in both cases \(\hat{L}\) reduces to a multiplication with a constant in Fourier space. To be specific we will use the non-conserved version of the eq.(2.1).

Such a Langevin equation is very good for illustrative purposes (since the origin of each term is transparent) but not for actual calculations. It is convenient to transform it into the Fokker-Planck equation since there is a whole arsenal of methods to solve the latter [123]. The derivation of Appendix A gives

$$\frac{\partial P[\phi, t]}{\partial t} = \int_k \frac{\delta}{\delta \phi_k} \left[ \mu \left( \frac{\delta}{\delta \phi_k} + \frac{\delta H[\phi]}{\delta \phi_k} \right) - D k_x \frac{\partial \phi_k}{\partial k_y} \right] P[\phi, t]$$

(2.4)

The functional \(P[\phi, t]\) describes the probability of creating a spatial order-parameter profile at time \(t\), the velocity profile is chosen to be \(\mathbf{v} = Dy \mathbf{e}_z\), where \(D\) is a shear rate, and the Onsager mobility coefficient \(\mu\) is approximated by \(\mu = \mu(k_0)\) (see [124, 125, 54, 56, 55] for discussion).

Because of the anharmonic terms in eq.(1.31) the Fokker-Planck equation (2.4) cannot be solved exactly in the presence of the flow (if \(D = 0, P[\phi] \sim \exp(-H[\phi])\)). There are several methods to construct its approximate solution
in a systematic way (see, for example, [126, 127, 128]). Here we employ the approach of Zwanzig [129] and Fredrickson [56], and derive a set of equations for the first two cumulants (all higher correlations are neglected)

\[ C(k) = \langle \phi_k \rangle \]  \hspace{1cm} (2.5)
\[ S(k) = \langle \phi_k \phi_{-k} \rangle - \langle \phi_k \rangle \langle \phi_{-k} \rangle \]

Generally, the average density profile can be represented as a superposition of plane waves

\[ C(k) = \sum_{i=1}^{m} a_i(t) \left[ \delta_{k,k_0 n^{(i)}} + \delta_{k,-k_0 n^{(i)}} \right] \]  \hspace{1cm} (2.6)

where the set of vectors \( n^{(i)} \) determine the lattice symmetry. The equation (2.6) tell us that all structures in the system have the same typical size \( \sim k_0^{-1} \).

The equations for the cumulants are derived in Appendix B. In the steady state they read

\[ h_i = \tau a_i + 2i + \frac{1}{2} a_i \int_k \lambda(k_0 n^{(i)}, -k_0 n^{(i)}, k, -k) S(k) + \mathcal{B}_i \]  \hspace{1cm} (2.7)
\[ 1 = -\frac{D}{2\mu} k_x \frac{\partial}{\partial k_y} S(k) + S(k) \left[ \tau + (k - k_0)^2 + \frac{1}{2} \int_q \lambda(k, q, -k, -q) S(q) + \mathcal{C}(k) \right] \]  \hspace{1cm} (2.8)

where we defined the structure constants:

\[ \mathcal{A}_i = \frac{1}{2} \int_{k_1} \int_{k_2} \xi(-k_0 n^{(i)}, k_1, k_2) C(k_1) C(k_2) \]
\[ \mathcal{B}_i = \frac{1}{2} \int_{k_1} \int_{k_2} \int_{k_3} \lambda(-k_0 n^{(i)}, k_1, k_2, k_3) C(k_1) C(k_2) C(k_3) \]  \hspace{1cm} (2.9)
\[ \mathcal{C}(k) = \frac{1}{2} \int_q \lambda(k, q, -k, -q) C(q) C(-q) \]

In equation (2.7) we introduced artificial external fields \( h_i \). This is equivalent to an introduction of the additional term in the Hamiltonian \( H_{\text{ext}} = - \int_k h_k \phi_{-k} \), \( h_k = \sum_{i=1}^{m} h_i [\delta_{k,k_0 n^{(i)}} + \delta_{k,-k_0 n^{(i)}}] \), which describes the interaction with the external field \( h_k \). The fields \( h_i \) will allow us to construct a potential \( \mathcal{F} \) governing the dynamics. In equilibrium the potential \( \mathcal{F} \) has the meaning of the free energy of the system. Thus, by introducing the fields \( h_i \) we obtain an analytic continuation of the free energy to the dynamic case [56].

As our last simplification we introduce an approximation for the vertex functions \( \xi \) and \( \lambda \). Since we use the principle-harmonic approximation in (2.6), we assume that all wave-vectors have the same modulus \( |k| = k_0 \). Moreover, following [130, 131, 120], we take into account the weak angular dependence in the
4th-order vertex function $\lambda$, so

$$\xi(k_1, k_2, k_3) = \xi \delta(\mathbf{k}_1 + \mathbf{k}_2 + \mathbf{k}_3) \quad (2.10)$$

$$\lambda(k_1, k_2, -k_1, -k_2) = \lambda \left[ 1 - \beta (\mathbf{k}_1 \cdot \mathbf{k}_2)^2 \right] \quad (2.11)$$

$$\beta \ll 1$$

where $\hat{k} = k/k$ denotes the unit vector in the direction of $k$.

The lamellar phase is described by eq. (2.6) with $m = 1$. In this case the structure constants are

$$\mathfrak{A} = 0$$

$$\mathfrak{B} = \frac{1}{2} \lambda a^3 (1 - \beta) \quad (2.12)$$

$$\mathcal{C}(k) = \lambda a^2 \left( 1 - \beta (\mathbf{k} \cdot \mathbf{n})^2 \right)$$

Here $\mathbf{n}$ is a unit vector perpendicular to the lamellae. It parameterizes the lamellar orientation: $n_y^2 = 1$ and $n_z^2 = 1$ correspond to the parallel and perpendicular orientation, respectively.

We introduce the notation:

$$\sigma(\mathbf{p}) = \frac{\lambda}{2} \int S(\mathbf{q}) \left[ 1 - \beta (\mathbf{p} \cdot \mathbf{q})^2 \right] \quad (2.13)$$

$$r - \mathbf{k} \cdot \mathbf{e} \cdot \mathbf{k} = \tau + \sigma(\mathbf{k}) + \lambda a^2 \left[ 1 - \beta (\mathbf{k} \cdot \mathbf{n})^2 \right] \quad (2.14)$$

$$S_0(k) = \left[ r - \mathbf{k} \cdot \mathbf{e} \cdot \mathbf{k} + (k - k_0)^2 \right]^{-1} \quad (2.15)$$

and rewrite the steady-state equations for the lamellar phase in the final form:

$$- \frac{D}{2 \mu} k_x \frac{\partial}{\partial k_y} S(k) + S(k) S_0^{-1}(k) = 1 \quad (2.16)$$

$$h = (r - \mathbf{n} \cdot \mathbf{e} \cdot \mathbf{n}) a - \frac{1}{2} \lambda a^3 (1 - \beta) \quad (2.17)$$

Notation (2.13)-(2.15) has a clear physical meaning. The fluctuation integral (2.13) takes into account the fluctuations of the order parameter $\left| \mathbf{1} \right| \left| \mathbf{1} \right|$ and renormalizes the temperature in the system. Because of the angular dependence of the 4th-order vertex function $\lambda$ (eq.2.11), the renormalized temperature also has an angular dependence. Expanding it to first order in $\beta$ (one can easily check that the anisotropy tensor $e_{ij}$ is of order $O(\beta)$), we extract this angular dependence and get (2.14), where $\mathbf{r}$ denotes the $k$-independent part of the renormalized temperature and $-\mathbf{k} \cdot \mathbf{e} \cdot \mathbf{k}$ absorbs the other terms. Finally, $S_0(k)$ is an equilibrium structure factor, which, in the limit $\beta = 0$ reduces to the one studied by Brazovskii [111], Fredrickson and Helfand [68], and discussed in Section 1.6.
2.2 Strong-shear regime

In order to determine the stable orientation of the lamellae, we need to solve eq.(2.16) for the structure factor. Its general solution can be found by the method of characteristics (see Appendix C)

\[
S(k, t) = 2\mu \int_0^t dt \exp \left[ -2\mu \int_0^t ds S_0^{-1}(k(s)) \right]
\]

(2.18)

where \( k(s) = (k_x, k_y + D s k_x, k_z) \). The steady state corresponds to the limit \( t \to \infty \). Integration in eq.(2.18) in the case \( D \to \infty \) is performed in Appendix D. It gives

\[
S_\infty(k) = c_0 \left( \frac{\mu \sqrt{\alpha}}{|k_x k_y|} \right)^{2/3}, \quad c_0 = \frac{(48\pi)^{1/3}}{3} \Gamma \left( \frac{1}{3} \right)
\]

(2.19)

For finite \( D \) the structure factor can be interpolated (Cates and Milner [53]) between equilibrium and infinitely fast flow

\[
S^{-1}(k) = S_0^{-1}(k) + S_\infty^{-1}(k) = \tau - \hat{k} \cdot \hat{e} \cdot \hat{k} + (k - k_0)^2 + \frac{1}{c_0} \left( \frac{D |k_x k_y|}{\mu \sqrt{\alpha}} \right)^{2/3}
\]

(2.20)

This approximation allows one to calculate the fluctuation integral eq.(2.13) for small \( r \) and \( \beta \). Using a method similar to that of Appendix 1.F and the formulas (Gradshteyn & Ryzhik [132])

\[
\int_0^{\pi/2} dx \sin^a x = \frac{1}{2} \frac{\Gamma \left( \frac{1}{2} \right) \Gamma \left( \frac{a+1}{2} \right)}{\Gamma \left( \frac{a+2}{2} \right)}
\]

(2.21)

\[
\int_0^{\pi/2} dx \sin^a x \cos^b x = \frac{1}{2} \frac{\Gamma \left( \frac{a+1}{2} \right) \Gamma \left( \frac{b+1}{2} \right)}{\Gamma \left( \frac{a+b+2}{2} \right)}
\]

(2.22)

we obtain

\[
\sigma(k) = (\alpha \lambda)^{2/3} \mathcal{D} \left[ 1 - \frac{\beta}{7} \left( 2(\hat{k}_x^2 + \hat{k}_y^2) + 3\hat{k}_z^2 \right) \right]
\]

(2.23)

where

\[
\mathcal{D} = \left( \frac{12}{\pi} \right)^{1/6} \sqrt{\Gamma \left( \frac{1}{3} \right)} \left( \frac{D_s}{D} \right)^{1/3}, \quad D_s = \mu \lambda \sqrt{\alpha}
\]

(2.24)

The spinodal temperature \( \tau_s \), marking the absolute limit of stability of the disordered phase, is found from the condition

\[
\left. r - n \cdot \hat{e} \cdot n \right|_{a=0} = 0
\]

(2.25)
and gives
\[
\tau_s(n) = - (\alpha \lambda)^{2/3} \Phi \left[ 1 - \frac{\beta}{7} \left( 2n_y^2 + 3n_z^2 \right) \right]
\] (2.26)

This is a remarkable result. Unlike the equilibrium fluctuation theory, where the spinodal temperature is suppressed to \( \tau_s = -\infty \) (see eq. (1.48)), the high-shear rate limit has a finite spinodal temperature. This temperature scales as \( \tau_s \sim D^{-1/3} \) and asymptotically reaches the mean-field value \( \tau_s = 0 \) as \( D \to \infty \). This demonstrates that the fast flow suppresses the critical fluctuations and restores the mean-field critical behaviour.

Moreover, eq. (2.26) shows that the spinodal temperature becomes anisotropic – it depends on the orientation of the density profile \( n \). The highest spinodal temperature corresponds to the perpendicular orientation with \( n = (0,0,1) \). Thus, we predict the perpendicular orientation to appear below the spinodal.

**Remark:** Our way of locating the spinodal temperature is equivalent to the linear stability analysis of the amplitude equation

\[
- \frac{1}{\mu} \frac{\partial a}{\partial t} = (r - n \cdot \mathbf{e} \cdot n) a - \frac{1}{2} \lambda a^3 (1 - \beta)
\] (2.27)

To identify the instability of the uniform solution \( a = 0 \) we write \( a = 0 + \delta a e^{s t} \) and to the first order in \( \delta a \)

\[
s \sim r - n \cdot \mathbf{e} \cdot n \bigg|_{a=0}
\] (2.28)

The growth rate changes its sign at \( s = 0 \), which is eq. (2.25).

### 2.3 Hydrodynamic effects

In the previous section we assumed that the velocity profile in melt is identical to the applied Couette flow

\[
\mathbf{v} = D \gamma \mathbf{e}_x
\] (2.29)

Thus, we have ignored local alterations of the flow caused by the inhomogeneous structure of melt. Here we present one way to incorporate these alterations into the theory discussed [56].

If we assume that the velocity profile instantaneously adapts to changes in the density profile \( \phi(\mathbf{r}) \), we can use the following momentum balance (Navier-Stokes) equation

\[
\nabla_j \left[ \eta(\phi) \left\{ \nabla_i u_j + \nabla_j u_i \right\} \right] = \nabla_i p
\] (2.30)
where $\mathbf{u}$ is a velocity field, $\eta(\phi)$ is a viscosity which depends on the local value of the order parameter, and $p$ is pressure. To the first order in $\phi$ we can write

$$
\eta(\mathbf{r}) = \eta_0 + \eta_1 \phi(\mathbf{r}) \tag{2.31}
$$

with $\epsilon = \eta_1/\eta_0$ being a small number. Assuming that block copolymer melts are incompressible, we use

$$
\nabla \cdot \mathbf{u} = 0 \tag{2.32}
$$

to exclude pressure from eq.(2.30). If we separate the applied Couette flow

$$
\mathbf{u} = D y \mathbf{e}_x + \mathbf{w}, \tag{2.33}
$$

then eq.(2.30) reads in Fourier space

$$
\hat{w}_i(k) = -\frac{iD\epsilon\phi_k}{k}z_i(k) + \frac{\epsilon}{k^2} \int_{q} \phi_{k-q}\Omega_{ij}(k, q)w_i(q)q \tag{2.34}
$$

where

$$
z_i(k) = \hat{k}_iT_{ij}(k) + \hat{k}_jT_{ix}(k) \tag{2.35}
$$

$$
\Omega_{ij}(k, q) = 2\hat{k}_i\hat{q}_j(k \cdot \hat{q}) - \hat{k}_j\hat{q}_i - \delta_{ij}(k \cdot \hat{q}) \tag{2.36}
$$

and

$$
T_{ij}(k) = \delta_{ij} - \hat{k}_i\hat{k}_j \tag{2.37}
$$

Its solution is found as a power series in $\epsilon$

$$
\hat{w}_i(k) = \sum_{n=1}^{\infty} \hat{w}_i^{(n)}(k)\epsilon^n \tag{2.38}
$$

$$
\hat{w}_i^{(1)}(k) = -\frac{iD\phi_k}{k}z_i(k)
$$

$$
\hat{w}_i^{(n>1)}(k) = -\frac{iD}{k} \int_{p_1,...,p_{n-1}} \phi_{k-p_1}p_{p_1-p_2}...\phi_{p_{n-1}}
\times \Omega_{i,i_1}(k, p_1)\Omega_{i_2,i_3}(p_1, p_2)\Omega_{i_3,i_4}(p_2, p_3)\Omega_{i_4,i_5}(p_3, p_4)...\Omega_{i_{n-2},i_{n-1}}(p_{n-2}, p_{n-1})z_{i_{n-1}}(p_{n-1})
$$

This solution can, in principle, be used in the dynamic equation (2.1). It will, however, significantly increase the difficulty in finding the potential $\mathcal{F}$. Therefore, we choose another way to introduce the hydrodynamic effects into our theory.

Let us assume that the real velocity profile $\mathbf{u}$ has the same triangular shape as the applied one (eq.(2.29)) with an effective shear rate $D_e$

$$
\mathbf{u} = D_e y \mathbf{e}_x \tag{2.39}
$$
Here the role of the hydrodynamic effects is to renormalize the shear rate only: 
\( D \rightarrow D_e \). The effective shear rate \( D_e \) can be found from the relation

\[
D_e = \lim_{k \rightarrow 0} \nabla_y u_x
\]

(2.40)

which simply tells us that the effective shear rate times the average viscosity \( \eta_0 \) is equal to the shear stress. The latter can be calculated with the help of eq.(2.34)

\[
D_e = D \left[ 1 + \varepsilon^2 \lim_{k \rightarrow 0} \int_q \phi_{k-q} \phi_q \Omega_{z_0}(k, q) z_\alpha(q) \right] 
\]

(2.41)

where we have used \( \lim_{k \rightarrow 0} \phi_k = 0 \). Since we are interested in the effective shear rate, we can replace \( \phi_{k-q} \phi_q \) with \( \langle \phi_{k-q} \phi_q \rangle \), which is averaged over the solution of eq.(2.4). In the limit \( k \rightarrow 0 \) it reduces to

\[
\langle \phi_{-q} \phi_q \rangle = S(q) + C(q)C(-q)
\]

(2.42)

The first term in the previous equation can be omitted since the integral \( f_q S(q) \) depends on the shear rate (see eq.(2.23)), and, therefore, contributes a higher order correction to \( D_e \). After integration we obtain

\[
D_e = D \left[ 1 + 2a^2 \varepsilon^2 n_y^2 \right] 
\]

(2.43)

This equation can be used to construct the potential \( \mathcal{F} \) in the strong-shear limit. Replacing \( D \) with \( D_e \) in eqs.(2.17), (2.14), and (2.23)-(2.26), we find to the leading order in \( \varepsilon \)

\[
h = [\tau - \tau_s(n)] a + \left[ \frac{1}{2} \lambda(1 - \beta) + \frac{2}{3} \varepsilon^2 n_y^2 \tau_s(n) \right] a^3
\]

(2.44)

Since the coefficients in eq.(2.44) do not depend on \( a \) (unlike eq.(2.17)), the potential \( \mathcal{F} \) is easily obtained by integration

\[
\mathcal{F} = 2 \int_0^a da' \ h(a') = [\tau - \tau_s(n)] a^2 + a^4 \left[ \frac{1}{2} \lambda(1 - \beta) + \frac{2}{3} \varepsilon^2 n_y^2 \tau_s(n) \right]
\]

(2.45)

Optimization with respect to the amplitude \( a \) gives

\[
a^2 = \frac{\tau_s(n) - \tau}{\frac{2}{3} \lambda(1 - \beta) + \frac{2}{3} \varepsilon^2 n_y^2 \tau_s(n)}
\]

(2.46)

and

\[
\mathcal{F}(n) = -\frac{(\tau_s(n) - \tau)^2}{\lambda(1 - \beta) + \frac{4}{3} \varepsilon^2 n_y^2 \tau_s(n)}
\]

(2.47)
The ODT transition occurs when $\mathcal{F} = 0$, and, therefore, $\tau = \tau_s(n)$, which is identical to eq.(2.26). In order to find the stable orientation we compare

$$
\mathcal{F}_\parallel = -\frac{(\tau_\parallel - \tau)^2}{\lambda(1 - \beta) + \frac{4}{3}\epsilon^2\tau_\parallel}
$$

and

$$
\mathcal{F}_\perp = -\frac{(\tau_\perp - \tau)^2}{\lambda(1 - \beta)}
$$

Eq.(2.26) gives

$$
\tau_\perp \equiv \tau_s\left(n_z^2 = 1\right) > \tau_\parallel \equiv \tau_s\left(n_y^2 = 1\right),
$$

and we confirm the prediction of the previous section that the perpendicular lamellae are stable immediately below the spinodal. However, as the temperature reaches some value $\tau_i$, the perpendicular orientation becomes unstable and transforms into the parallel one. The temperature $\tau_i$ is found from the condition

$$
\mathcal{F}_\perp = \mathcal{F}_\parallel \Rightarrow \tau_i \approx -\frac{3\lambda}{2e^2}\left(1 - \frac{\tau_\perp}{\tau_\parallel}\right) \approx -\frac{3\lambda}{14e^2\beta}
$$

The last equation has two peculiar features: i) the $\perp \rightarrow \parallel$ transition temperature does not depend on the shear rate as $D \rightarrow \infty$; ii) if we neglect the hydrodynamic effects and put $\epsilon = 0$, then $\tau_i \rightarrow -\infty$ and the perpendicular lamellae are stable in the whole temperature range in the strong-shear limit.

### 2.4 Cross-over from small- to large-shear rate behaviour

In this section we analyze the transition from the perpendicular to parallel orientation caused by decrease of the shear rate. We start with eq.(2.20) for the structure factor $S(k)$, which interpolates between $S_0$ and $S_\infty$

$$
S^{-1}(k) = S_0^{-1}(k) + S_\infty^{-1}(k) = \frac{\epsilon - k \cdot \hat{e} \cdot \hat{k} + (k - k_0)^2}{c_0 \left( \frac{D}{\mu \sqrt{\alpha}} \right)^{2/3}}
$$

One should realize that the previous equation is an analytic continuation of the $D \rightarrow \infty$ behaviour to $D < \infty$ values. As a result, a small-$D$ behaviour of eq.(2.20) does not correspond to the $D \rightarrow 0$ behaviour of eq.(2.18). On the contrary, it describes the $D \sim O(1)$ region. Since we expect the cross-over to
lay in-between the $D \sim O(1)$ and $D \to \infty$ regions, we need to calculate the fluctuation integral $\sigma(\hat{k})$ in-between these regions. This can be done in several steps. First, we use $S(\mathbf{k})$ from eq.(2.20) to perform integration over the radial part in eq.(2.13). This gives

$$
\sigma(\hat{k}) \approx \frac{\lambda k_0^2 c_1}{16\pi^3} \left( \frac{\mu}{D\sqrt{\alpha}} \right)^{1/3} \times \int d\Omega \frac{1 - \beta (\hat{k} \cdot \hat{q})^2}{|\hat{q}_x \hat{q}_y|^{1/3}} \left[ \frac{\pi}{2} + \arctan \left( \frac{k_0 c_1}{|\hat{q}_x \hat{q}_y|^{1/3}} \left( \frac{\mu}{D\sqrt{\alpha}} \right)^{1/3} \right) \right] (2.52)
$$

where $c_1 = \sqrt{\Gamma \left( \frac{1}{3} \right) / (9\pi)^{1/3}}$. As a next step we expand the integrand for $D \ll 1$ and $D \gg 1$ and sum these expressions keeping only the few first terms. Integration over the orientations of the unit vector $\hat{q}$ ($\int d\Omega = \int_0^\pi d\theta \sin \theta \int_0^{2\pi} d\phi$) then gives

$$
\sigma(\hat{k}) = \frac{\lambda \sqrt{\alpha}}{8\pi^{3/2}} \left\{ -4\pi \left( 1 - \frac{\beta}{3} \right) + \frac{1}{3Z^3} \left[ I_1 - \beta \left( I_2 (\hat{k}_x^2 + \hat{k}_y^2) + I_3 \hat{k}_z^2 \right) \right] 
+ 4\pi^2 2^{1/3}\sqrt{3Z} \left[ 1 - \beta \frac{2}{7} \left( 2\hat{k}_x^2 + 2\hat{k}_y^2 + 3\hat{k}_z^2 \right) \right] 
+ Z^2 \left[ I_4 - \beta \left( I_5 (\hat{k}_x^2 + \hat{k}_y^2) + I_6 \hat{k}_z^2 \right) \right] \right\} (2.53)
$$

where

$$
I_1 = 2 \frac{\sqrt{\pi} \Gamma \left( \frac{5}{6} \right)^2}{\Gamma \left( \frac{10}{6} \right)} \quad I_2 = 2 \frac{\sqrt{\pi} \Gamma \left( \frac{7}{6} \right) \Gamma \left( \frac{11}{6} \right)}{\Gamma \left( \frac{10}{6} \right)} \quad I_3 = 2 \frac{\Gamma \left( \frac{3}{6} \right)^2 \Gamma \left( \frac{5}{6} \right)}{\Gamma \left( \frac{10}{6} \right)}
$$

$$
I_4 = \frac{\Gamma \left( \frac{1}{6} \right)^3}{\sqrt{\pi}} \quad I_5 = 2 \frac{\sqrt{\pi} \Gamma \left( \frac{1}{6} \right) \Gamma \left( \frac{5}{6} \right)}{\Gamma \left( \frac{11}{6} \right)} \quad I_6 = 2 \frac{\Gamma \left( \frac{3}{6} \right) \Gamma \left( \frac{1}{6} \right)^2}{\Gamma \left( \frac{11}{6} \right)}
$$

This procedure relies on a number of approximations. However, any direct analysis of eq.(2.16) is impossible and we use eq.(2.53) for moderate shear rates.

The order-disorder transition (ODT) occurs when $\mathcal{F}$ becomes negative. The corresponding transition temperature follows from eq.(2.47)

$$
\tau_s(\mathbf{n}) = -\sigma(\mathbf{n}) (2.54)
$$

The orientation with the lowest $\sigma$ will appear immediately below the ODT temperature. The cross-over is then located at such a value of $D$ that $\sigma_\parallel - \sigma_\perp$ changes
its sign. From eq.(2.53) this point is given by
\[ \sigma_{\parallel} - \sigma_{\perp} \approx \frac{1}{3Z^2}(I_3 - I_2) + \frac{4\pi^2}{l}2^{1/3}\sqrt{3}Z + Z^2(I_6 - I_3) = 0 \] (2.55)
and is found to be
\[ D_{cr} \approx 4 \cdot 10^3 \mu \alpha \] (2.56)

From eq.(2.55) it also follows that \( \tau_{s\parallel} < \tau_{s\perp} \) for \( D > D_{cr} \), which fits the strong-shear behaviour discussed in the previous section. When \( D < D_{cr} \), the parallel orientation first appears below the spinodal. This cross-over is depicted in Fig.2.1.

As it was noticed before [53, 56], the mean-field behaviour is restored in the limit \( D \to \infty \). On the other hand, the small-\( D \) region is dominated by fluctuations. Thus, \( D_{cr} \) can be interpreted as the position of a cross-over from the fluctuation to mean-field behaviour. The scaling properties of \( D_{cr} \) follow from eq.(2.56) and are determined by the Onsager coefficient \( \mu \). Using the results of [125, 124, 45, 55] \( \mu \equiv k_0^2\Lambda(k_0)/N \) with \( \Lambda \) from [124, 45] we obtain
\[ D_{cr} \sim N^{-3} \] (2.57)
which shows that the fluctuation region disappears in the limit \( N \to \infty \). In equilibrium the same conclusion was drawn in [67, 68].

Finally, we emphasize that these results are independent of the hydrodynamic corrections discussed in the previous section.
2.5 Weak-shear regime

When the shear rate is very small we expect that the flow only slightly modifies the equilibrium fluctuation spectrum. Therefore, we can develop the solution of eq.(2.16) as a perturbation series in $D$

$$S(k) = S_0(k) + \frac{D}{2\mu} S_1(k) + \left( \frac{D}{2\mu} \right)^2 S_2(k) + \cdots$$  \hspace{1cm} (2.58)

where

$$S_1(k) = k_x S_0(k) \frac{\partial S_0(k)}{\partial k_y}$$  \hspace{1cm} (2.59)

$$S_2(k) = \left( k_x S_0(k) \frac{\partial S_0(k)}{\partial k_y} \right)^2 S_0(k)$$  \hspace{1cm} (2.60)

Substituting the expression for $S_0$ (eq.(2.15)) we obtain

$$S_1(k) = 2S_0^3(k) \hat{k}_x T_{yi}(\hat{k}) e_{ij} \hat{k}_j$$  \hspace{1cm} (2.61)

$$S_2(k) = 2S_0^4(k) \hat{k}_y^2 \hat{k}_z^2 \left[ 6S_0(k)(k - k_0)^2 - 1 \right]$$  \hspace{1cm} (2.62)

where we have dropped all the terms that do not contribute to the fluctuation integral (see [56] for details). Evaluation of $\sigma$, eq.(2.13), leads to

$$\sigma(k) = \sigma_0(k) + \left( \frac{D}{2\mu} \right)^2 \sigma_2(k) + \cdots$$  \hspace{1cm} (2.63)

with

$$\sigma_0(k) = \frac{\alpha \lambda}{\sqrt{r}} \left[ 1 - \frac{\beta}{3} (\hat{k} \cdot \hat{k}) \right] + \frac{1}{6 r^{3/2}} e_{ii}$$  \hspace{1cm} (2.64)

$$\sigma_2(k) = - \frac{\alpha^2 \lambda^2 5\pi}{8 r^{7/2}} \left[ \frac{1}{15} - \frac{\beta}{35} \left( \hat{k}_x^2 + \hat{k}_y^2 + \frac{1}{3} \hat{k}_z^2 \right) + \frac{7\beta}{90} \frac{\alpha \lambda}{r^{3/2}} \right]$$

$$+ \frac{\beta}{10} \frac{\lambda a^2}{r} \left( n_y^2 + \frac{1}{3} n_z^2 \right)$$  \hspace{1cm} (2.65)

where $e_{ii}$ denotes the trace of $e_{ij}$, and $\hat{k} \cdot \hat{k} = 1$ is left in order to separate the orientation-dependent terms in eq.(2.14). The separation gives

$$r = \tau + \lambda a^2 + \frac{\alpha \lambda}{\sqrt{r}} + \frac{1}{6 r^{3/2}} e_{ii}$$

$$- \left( \frac{D}{2D_s} \right)^2 \left( \frac{\alpha \lambda}{r^{7/2}} \right) \left[ \frac{1}{15} + \frac{7\beta}{90} \frac{\alpha \lambda}{r^{3/2}} + \frac{\beta}{10} \frac{\lambda a^2}{r} \left( n_y^2 + \frac{1}{3} n_z^2 \right) \right]$$  \hspace{1cm} (2.66)

$$e_{ij} = \beta \left( \lambda a^2 n_i n_j + \frac{1}{3} \frac{\alpha \lambda}{\sqrt{r}} \delta_{ij} \right)$$

$$- \beta \left( \frac{D}{2D_s} \right)^2 \left( \frac{\alpha \lambda}{r^{7/2}} \right) \left[ \delta_{ij} \delta_{ij} + \delta_{ij} \delta_{ij} + \frac{1}{3} \delta_{ij} \delta_{ij} \right]$$  \hspace{1cm} (2.67)
To keep the lowest order in $D$ we substitute the equilibrium values for $\gamma = \alpha \lambda / r^3 = 0.92$, $\zeta = \alpha \lambda / r_0^3 = 11.11$, and $a^2 = 2r / \lambda$ (see eqs.(1.46-1.49)) into the terms $\sim D^2$. Omitting the angular-independent terms $O(D^0 \beta)$ and $O(D^2 \beta n^0)$ we obtain

$$
r = r - \Delta_1 + \lambda a^2 + \frac{\alpha \lambda}{\sqrt{r}}
$$

$$
h = a(r + \Delta_2) - \frac{1}{2} \lambda a^3
$$

where

$$
\Delta_1 = \frac{5\pi}{32} \gamma^{7/3} (\alpha \lambda)^2 / 3 \left( \frac{D}{D_c} \right)^2 \left[ \frac{1}{15} + \frac{2}{5} \left( n_y^2 + \frac{1}{3} n_z^2 \right) \right]
$$

$$
\Delta_2 = \frac{5\pi}{32} \gamma^{7/3} (\alpha \lambda)^2 / 3 \left( \frac{D}{D_c} \right)^2 \left[ \frac{\beta}{35} \left( n_y^2 + \frac{1}{3} n_z^2 \right) \right]
$$

We see that shear interacts stronger with the parallel orientation and we can foresee that it will be the stable one. To show it we evaluate the potential $F$ and solve the equation $F = 0$ up to $O(D^2)$. This gives for the ODT temperature

$$
\tau_{ODT} (n) = -2.0308 (\alpha \lambda)^2 / 3 \\
+ \frac{5\pi}{32} \gamma^{7/3} (\alpha \lambda)^2 / 3 \left( \frac{D}{D_c} \right)^2 \left[ \frac{1}{15} + \beta \left( \frac{1}{5} - \frac{2}{35} \frac{\zeta^{2/3}}{\gamma^{2/3}} \right) \left( n_y^2 + \frac{1}{3} n_z^2 \right) \right]
$$

or

$$
\frac{\tau_{ODT} (n)}{(\alpha \lambda)^2 / 3} = -2.0308 + \left( \frac{D}{D_c} \right)^2 \left[ 0.0268 + 0.0520 \beta \left( n_y^2 + \frac{1}{3} n_z^2 \right) \right]
$$

The last equation shows that the ODT temperature in the weak-shear limit is larger than the equilibrium one. We again identify this effect with the suppression of the critical fluctuations. Additionally, it shows that the ODT temperature is higher for the parallel orientation $n_y^2 = 1$ and we, therefore, predict its stability below the order–disorder transition.

In this section we have neglected the hydrodynamic corrections to the shear rate and put $D_c \approx D$. Since the hydrodynamic corrections also favour the parallel orientation, this assumption does not modify our predictions.

### 2.6 Early stages of phase separation

In this section we consider some dynamical aspects of the orientational transitions. Mainly, we relax the steady-state assumption employed in the previous sections and study the early stages of the order–disorder transition. Our goal is to discriminate between two possibilities: i) the orientations exchange their relative
stability in time, or $ii$) once some orientation becomes stable it remains stable until the steady state is reached. We perform our calculations in the strong-shear limit since it is technically easier.

In Appendix D we have shown that the structure factor in the large-shear rate regime is given by

$$S_\infty(k, t) = \frac{2\mu}{3x^{1/3}} \left[ \Gamma \left( \frac{1}{3} \right) - \Gamma \left( \frac{1}{3}, xt^3 \right) \right], \quad x = \frac{2\mu}{3} \left( \frac{Dk_xk_y}{k_0^3} \right)^2 \tag{2.74}$$

For $xt^3 \ll 1$ it can be approximated by $[133]

$$S_\infty(k, t) = 2\mu t \left( 1 - \frac{xt^3}{4} \right) \tag{2.75}$$

Employing the approximation eq.(2.20) we obtain for the fluctuation integral

$$\sigma(k, t) = \sigma_1 \sqrt{t} - \sigma_2(k) t^{7/2} \tag{2.76}$$

where

$$\sigma_1 = \alpha \lambda \sqrt{\frac{\mu}{2}} \left( 1 - \frac{\beta}{3} \right) \tag{2.77}$$

$$\sigma_2(k) = \frac{\pi}{3\sqrt{2}} \alpha^2 \mu^{3/2} D^2 \left[ \frac{1}{15} - \frac{\beta}{35} \left( \hat{k}_x^2 + \hat{k}_y^2 + \frac{1}{3} \hat{k}_z^2 \right) \right] \tag{2.78}$$

In eq.(2.76) we could have added $\sigma_0$ to the r.h.s. to assure the correct behaviour at $t = 0$. This, however, would not modify our results.

Evolution of the amplitude $a$ is described by eq.(2.17) if we restore there the time derivative (see also eq.(B.6))

$$- \frac{1}{\mu} \frac{da}{dt} = [\tau + \sigma(n, t)]a + \frac{1}{2}\lambda a^3 \tag{2.79}$$

where we ignored some subdominant terms $O(\beta)$. This equation can be analyzed for the linear stability. We want to see how the homogeneous phase with $a = 0$ loses its stability and for which orientation the amplitude grows faster. We perform a small perturbation: $a = 0 + \delta a$ and linearize the amplitude equation (2.79)

$$- \frac{1}{\mu} \frac{d\delta a}{dt} = [\tau + \sigma(n, t)] \delta a \tag{2.80}$$

Its solution is$^1$

$$\delta a \sim \exp \left[ -\mu \left( \tau t + \frac{2}{3} \sigma_1 t^{3/2} - \frac{2}{3} \sigma_2(n) t^{9/2} \right) \right]$$

$$\sim e^{-\mu t} \exp \left[ -\frac{\sqrt{2\pi \beta}}{9!!} \alpha^2 \mu^{5/2} \theta^{9/2} D^2 \left( n_y^2 + \frac{1}{3} n_z^2 \right) \right] \tag{2.81}$$

$^1$Here we employ the double factorial notation as follows:

$$(2n)!! = 2 \cdot 4 \cdot 6 \ldots (2n)$$

$$(2n - 1)!! = 1 \cdot 3 \cdot 5 \ldots (2n - 1)$$
We see that the amplitude grows faster for \( n_x^2 = 1 \). Moreover, before we have shown that the perpendicular orientation is also stable immediately below the ODT in the steady state as \( D \to \infty \). Thus, we have an indication that the stable orientation appears as an instability of an homogeneous phase and keeps growing until the steady state is reached. (Since we have considered only \( t \to 0 \) and \( t \to \infty \) limits we cannot be sure that the same orientation is stable over the whole \((0, \infty)\) range. However, it is very unlikely that there is a range \((t_1, t_2)\) where the other orientation would be stable, since there is no physical mechanism in the system to select two special moments of time \( t_1 \) and \( t_2 \).)

It is interesting to note that despite the fact that we have considered the strong-shear limit, the fluctuation integral in eq. (2.78) scales as \( D^2 \), which is typical for the weak-shear regime. This is not surprising since shortly after the beginning of shearing even very strong shear would not have time to develop. Thus we effectively have a weak-shear behaviour. However, the orientation stays specific to strong shear – the perpendicular one.
Appendix 2.A Derivation of the Fokker-Planck equation

Here we show how to pass from the Langevin description (eq. (2.1)) to the Fokker-Planck equation.

We start from the Langevin equation for the non-conserved order parameter field \( \psi(r, t) \)

\[
\frac{\partial \psi(r, t)}{\partial t} = -D_y \frac{\partial \psi(r, t)}{\partial x} - \mu \frac{\delta H[\psi]}{\delta \psi(r, t)} + \zeta(r, t)
\]

\[
\equiv -\mu F[\psi(r, t)] + \zeta(r, t)
\]  

(A.1)

with

\[
\langle \zeta(r, t) \rangle = 0 \quad , \quad \langle \zeta(r, t) \zeta(r', t') \rangle = 2 \mu \delta(r - r') \delta(t - t')
\]  

(A.2)

Let us introduce a functional \( P[\phi(r), t] \), which gives the probability that at time \( t \) the order parameter profile will be given by \( \phi(r) \). This statement can be written as

\[
P[\phi(r), t] = \left\langle \delta[\psi(r, t) - \phi(r)] \right\rangle
\]  

(A.3)

where the angular brackets denote the average over the noise. Taking the time derivative of eq. (A.3) and using the Langevin equation for \( \psi(r, t) \) we obtain

\[
\frac{\partial P[\phi(r), t]}{\partial t} = \int dr \left\langle \left( -\mu F[\psi(r, t)] + \zeta(r, t) \right) \frac{\delta}{\delta \psi(r, t)} \delta[\psi(r, t) - \phi(r)] \right\rangle
\]  

(A.4)

Since the \( \delta \)-function is symmetric, we can replace \( \frac{\delta}{\delta \psi(r, t)} \) with \( -\frac{\delta}{\delta \phi(r)} \):

\[
\frac{\partial P[\phi(r), t]}{\partial t} = \int dr \frac{\delta}{\delta \phi(r)} \left[ \mu F[\phi(r)] P[\phi(r), t] - \left\langle \zeta(r, t) \delta[\psi(r, t) - \phi(r)] \right\rangle \right]
\]  

(A.5)

In order to calculate the correlation function \( \langle \zeta(r, t) \delta[\psi(r, t) - \phi(r)] \rangle \) we notice the following relation. The average of an arbitrary functional \( G[\zeta] \zeta(r, t) \) over the Gaussian distribution of \( \zeta(r, t) \)

\[
\rho[\zeta] = \int D\zeta \exp \left[ -\frac{1}{4\mu} \int_{-\infty}^{\infty} d\tau \int d\mathbf{r}' \zeta(\mathbf{r}', \tau)^2 \right]
\]

(A.6)

can be evaluated as

\[
\left\langle G[\zeta] \zeta(r, t) \right\rangle = \int D\zeta G[\zeta] \zeta(r, t) \exp \left[ -\frac{1}{4\mu} \int_{-\infty}^{\infty} d\tau \int d\mathbf{r}' \zeta(\mathbf{r}', \tau)^2 \right]
\]

\[
= -2\mu \int D\zeta G[\zeta] \frac{\delta}{\delta \zeta(r, t)} \exp \left[ -\frac{1}{4\mu} \int_{-\infty}^{\infty} d\tau \int d\mathbf{r}' \zeta(\mathbf{r}', \tau)^2 \right]
\]  

(A.7)

\[
= 2\mu \int D\zeta \frac{\delta G[\zeta]}{\delta \zeta(r, t)} \exp \left[ -\frac{1}{4\mu} \int_{-\infty}^{\infty} d\tau \int d\mathbf{r}' \zeta(\mathbf{r}', \tau)^2 \right] = 2\mu \left\langle \frac{\delta G[\zeta]}{\delta \zeta(r, t)} \right\rangle
\]
where integration by parts was used in the last step. As a result,

\[
\left\langle \zeta(r, t) \delta [\psi(r, t) - \phi(r)] \right\rangle = 2\mu \left( \frac{\delta}{\delta \zeta(r, t)} \delta [\psi(r, t) - \phi(r)] \right)
\]

\[
= -2\mu \frac{\delta}{\delta \phi(r)} \left( \frac{\delta \psi(r, t)}{\delta \zeta(r, t)} \delta [\psi(r, t) - \phi(r)] \right)
\]

(A.8)

The Langevin equation can be formally integrated

\[
\psi(r, t) = \psi(r, 0) - \mu \int_0^t d\tau F[\psi(r, \tau)] + \int_0^t d\tau \zeta(r, \tau)
\]

(A.9)

which gives

\[
\frac{\delta \psi(r, t)}{\delta \zeta(r, t')} = -\mu \int_0^t d\tau \frac{\delta F[\psi(r, \tau)]}{\delta \zeta(r, t')} + \theta(t - t')
\]

(A.10)

Here \(\theta(t - t')\) is a step function the role of which is to preserve causality: the order parameter \(\psi(r, t)\) can only depend on the noise \(\zeta(r, t')\), if \(t > t'\). If we now put \(t' = t\) in eq.(A.10), we see that its r.h.s. becomes undefined. This problem is connected with some features of our derivation and discussed in [114, 122]. It can be avoided if we choose

\[
\left\langle \zeta(r, t) \zeta(r', t') \right\rangle = 2\mu \delta(r - r') \eta(t - t')
\]

(A.11)

where \(\eta(t - t')\) is sharply peaked around \(t = t'\) and \(\int_{-\infty}^{\infty} dt \eta(t) = 1\). Then one can show [114, 122] that

\[
\frac{\delta \psi(r, t)}{\delta \zeta(r, t)} = \frac{1}{2}
\]

(A.12)

Finally,

\[
\frac{\partial P[\phi(r), t]}{\partial t} = \mu \int dr \frac{\delta}{\delta \phi(r)} \left[ F[\phi(r)] + \frac{\delta}{\delta \phi(r)} \right] P[\phi(r), t]
\]

(A.13)

Restoring the original notation we obtain

\[
\frac{\partial P[\phi(r), t]}{\partial t} = \int d\mathbf{r} \frac{\delta}{\delta \phi(\mathbf{r})} \left[ \mu \left( \frac{\delta}{\delta \phi(\mathbf{r})} + \frac{\delta H[\phi]}{\delta \phi(\mathbf{r})} \right) + D \frac{\partial \phi(\mathbf{r})}{\partial x} \right] P[\phi(\mathbf{r}), t]
\]

(A.14)

or in Fourier space

\[
\frac{\partial P[\phi, t]}{\partial t} = \int k \frac{\delta}{\delta \phi_k} \left[ \mu \left( \frac{\delta}{\delta \phi_{-k}} + \frac{\delta H[\phi]}{\delta \phi_{-k}} \right) - D k_x \frac{\partial \phi_k}{\partial k_y} \right] P[\phi, t]
\]
Appendix 2.B  Cumulant expansion

Here we present a method [129] which allows one to derive the equations for the cumulants (eq. (2.5)) from the Fokker-Planck equation (2.4). This can be done with the help of the generating functional

\[ G[\varsigma, t] = \log G[\varsigma, t], \quad G[\varsigma, t] = \int \mathcal{D}\varsigma \exp \left[ - \int_k \phi_k \varsigma_{-k} \right] P[\phi, t] \] (B.1)

First we derive an equation for \( G[\varsigma, t] \)

\[ \frac{\partial G[\varsigma, t]}{\partial t} = \int \mathcal{D}\varsigma \exp \left[ - \int_k \phi_k \varsigma_{-k} \right] \int_k \frac{\delta}{\delta \phi_k} \left[ \mu \left( \frac{\delta}{\delta \phi_k} \phi_k \right) + \frac{\delta H[\phi]}{\delta \phi_k} \right] - D k_x \frac{\partial \phi_k}{\partial k_x} \] (B.2)

The main idea is to remove all \( \phi_k \) from this equation. This can be achieved if we notice that an arbitrary number of \( \phi \)'s can be produced by functional differentiation of \( \exp \left[ - \int_k \phi_k \varsigma_{-k} \right] \) with respect to \( \varsigma \). Then, using integration by parts we can rewrite it as a functional derivative of \( G \). The equation for \( G \) is obtained by substitution \( G[\varsigma, t] = \exp (G[\varsigma, t]) \). Using the Brazovskii Hamiltonian with an external field \( h \)

\[ H[\phi] = \frac{1}{2} \int_k \left\{ \frac{\tau + (k - k_0)^2}{\mu} \phi_k \phi_{-k} + \frac{1}{3!} \int_{k_1} \int_{k_2} \int_{k_3} \xi(k_1, k_2, k_3) \phi_{k_1} \phi_{k_2} \phi_{k_3} \right. \]
\[ + \frac{1}{4!} \int_{k_1} \int_{k_2} \int_{k_3} \int_{k_4} \lambda(k_1, k_2, k_3, k_4) \phi_{k_1} \phi_{k_2} \phi_{k_3} \phi_{k_4} - \int_h \phi_{-k} \] (B.3)

we get

\[ \frac{1}{\mu} \frac{\partial G[\varsigma, t]}{\partial t} = \int_k \varsigma_{-k} \left\{ \varsigma_k + \left\{ \frac{D}{\mu} k_x \frac{\partial}{\partial k_x} - \left[ \frac{\tau + (k - k_0)^2}{\mu} \right] \right\} \right\} \frac{\delta G}{\delta \varsigma_{-k}} + h_k \]
\[ - \frac{1}{2} \int_j \xi(q, -k, k - q) \left[ \frac{\delta G}{\delta q} \frac{\delta G}{\delta q_{-k}} + \frac{\delta^2 G}{\delta q_{-k} \delta q} \right] \] (B.4)
\[ - \frac{1}{3!} \int_{q_1} \int_{q_2} \lambda(q_1, q_2, -k, k - q_1 - q_2) \left[ \frac{\delta G}{\delta q_{-k}} \frac{\delta G}{\delta q_{-q_1}} \frac{\delta G}{\delta q_{-q_2}} \right] \]
\[ + \frac{\delta^2 G}{\delta q_{-q_1} \delta q_{-q_2} \delta q_{-k+q_1+q_2}} + \frac{\delta^2 G}{\delta q_{-q_2} \delta q_{-q_1 \delta q_{-k+q_1+q_2}}} + \frac{\delta^2 G}{\delta q_{-k+q_1+q_2} \delta q_{-q_1 \delta q_{-q_2}} \delta q_{-k+q_1+q_2}} \]

The cumulants enter the Taylor expansion of \( G[\varsigma, t] \) in terms of \( \varsigma \)

\[ G[\varsigma, t] = \int_k C(k, t) \varsigma_{-k} + \frac{1}{2} \int_k S(k, t) \varsigma_k \varsigma_{-k} + \cdots \] (B.5)
Therefore, we expand eq.(B.4) in powers of \( \varsigma \) and separate the different-order terms. This gives the equations for the cumulants

\[
\frac{1}{\mu} \frac{\partial C(k)}{\partial t} = h_k + \frac{D}{\mu k_x} \frac{\partial C(k)}{\partial k_y} - \left[ \tau + (k - k_0)^2 \right] C(k)
\]

\[
-\frac{1}{2} \int_q \xi(q, -k, k - q) C(k - q) C(q) - \frac{1}{2} C(k) \int_q \lambda(q, -q, k, -k) S(q) \tag{B.6}
\]

\[
-\frac{1}{3!} \int_{q_1} \int_{q_2} \lambda(q_1, q_2, -k, k - q_1 - q_2) C(q_1) C(q_2) C(k - q_1 - q_2)
\]

and

\[
\frac{1}{2\mu} \frac{\partial S(k)}{\partial t} = 1 + \frac{D}{2\mu k_x} \frac{\partial S(k)}{\partial k_y} - S(k) \left\{ \tau + (k - k_0)^2 \right\}
\]

\[
+ \frac{1}{2} \int_q \lambda(q, -q, k, -k) S(q) + \frac{1}{2} \int_q \lambda(q, -q, k, -k) C(q) C(-q) \tag{B.7}
\]

In the steady state these equations reduce to eq.(2.7-2.8).

### Appendix 2.C  Method of characteristics

Our goal is to find the solution of the equation for the structure factor

\[
\frac{1}{2\mu} \frac{\partial S(k_y, t)}{\partial t} = 1 + \frac{D}{2\mu k_x} \frac{\partial}{\partial k_y} - S(k_y, t) - S(k_y, t) S_0^{-1}(k_y) \tag{C.1}
\]

where for the sake of clarity we suppressed \( k_x \) and \( k_z \) in the arguments of the functions. We use the method of characteristics [134] to obtain its solution in quadratures. First we find the characteristic curves in \((k_y, t)\) plane. They are given by

\[
-2\mu dt = \frac{2\mu d k_y}{D k_x} \quad \Rightarrow \quad k_y = C - D k_x t \tag{C.2}
\]

where \( C \) is an integration constant which determines the characteristic. The structure factor varies along each characteristic as

\[
-\frac{2\mu}{D k_x} d k_y = \frac{d S(k_y, t)}{1 - S(k_y, t) S_0^{-1}(k_y)} \tag{C.3}
\]

The solution of this equation is easily obtained

\[
S(k_y) = \frac{2\mu}{D k_x} \int_0^{k_y} dy \exp \left[ \frac{2\mu}{D k_x} \int_y^{k_y} dx S_0^{-1}(x) \right] \tag{C.4}
\]
Inspired by the form of the characteristic curves (C.2), we change the integration variables

\[ y = k_y + D k_x \tau \]
\[ x = k_y + D k_x s \]

and restore \( k_x \) and \( k_z \) in the arguments

\[
S(k, t) = 2\mu \int_0^t d\tau \exp \left[ -2\mu \int_0^r ds S_0^{-1}(k_x, k_y + D k_x s, k_z) \right]
\]  

(C.5)

The steady state is reached as \( t \to \infty \).

**Appendix 2.D  Structure factor at high shear rates**

The integration in eq.(2.18) can be performed for small \( r \) and finite \( D \). First we expand \( S_0^{-1}(k(s)) \), eq.(2.15), near \( k_x \approx 0 \) and \( k_y^2 + k_z^2 \approx k_0^2 \)

\[
S_0^{-1}(k(s)) \approx \left(\frac{D k_x k_y s}{k_0}\right)^2
\]

(D.1)

It gives

\[
S(k, t) = 2\mu \int_0^t d\tau \exp \left[ -\frac{2\mu}{3} \left(\frac{D k_x k_y}{k_0}\right)^2 \tau^3 \right]
\]

(D.2)

Using formulas from Gradshteyn & Ryzhik [132]

\[
\int dt \exp \left(-\alpha t^2\right) = -\frac{1}{3\alpha^{1/3}} \Gamma \left(\frac{1}{3}, \alpha t^3\right)
\]

\[ \Gamma \left(\frac{1}{3}, 0\right) = \Gamma \left(\frac{1}{3}\right) \]

\[ \Gamma \left(\frac{1}{3}, \infty\right) = 0 \]

we obtain in the steady state \( t \to \infty \)

\[
S_\infty(k) = \Gamma \left(\frac{1}{3}\right) \left(\frac{2\mu k_0}{3D k_x k_y}\right)^{2/3}
\]

(D.3)