

# Analytical calculation of the Stokes drag of the spherical particle in a nematic liquid crystal

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As an approach to the motion of particles in an anisotropic liquid, we analytically study the Stokes drag of spherical particles in a nematic liquid crystal. The Stokes drag of spherical particles for a general anisotropic case is derived in terms of multipoles. In the case of weak anchoring, we use the well-known distribution of the elastic director field around the spherical particle. In the case of strong anchoring, the multipole expansion may be also used by modifying the size of a particle to the size of the deformation coating. For the case of zero anchoring (uniform director field) we found that the viscosities along the director  $\eta_{\parallel}$  and perpendicular direction  $\eta_{\perp}$  are almost the same, which is quite reasonable because in this case the liquid behaves as isotropic. In the case of non-zero anchoring, the general ratio  $\eta_{\parallel}/\eta_{\perp}$  is about 2 which is satisfied by experimental observations.

**Key words:** *Stokes drag, liquid crystal, diffusion, viscosity*

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## 1. Introduction

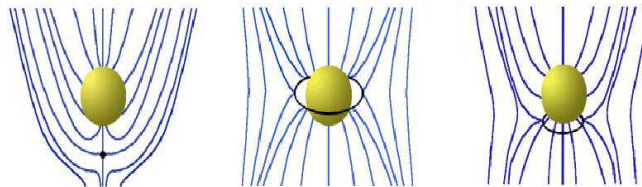
Colloidal particles in liquid crystals (LC) have attracted a great research interest during the recent years. Anisotropic properties of the host fluid-liquid crystal give rise to a new class of colloidal anisotropic interactions that never occur in isotropic hosts. Liquid crystal colloidal systems have shown much recent interest as the models for diverse phenomena in condensed matter physics. Particles suspended in a fluid are under the effect of the hits from the surrounding particles and perform Brownian motion. They perform random walk whose diffusion constant obeys the famous Stokes-Einstein relation. A simple Langevine approach predicts that the velocity autocorrelation function of random walkers decays exponentially [1]. The drag force can be derived from the Navier-Stokes equations with an additional assumption on the character of the random force. The Navier-Stokes equations, which describe the hydrodynamic behavior of fluids, assume that molecules are point particles or smooth spheres and, as a consequence, do not exert a torque on one another. These equations originate from the conservation of mass, linear momentum and energy during the collision processes. If the particles in a fluid are of non-spherical shape, they can induce rotation to each other during the collisions and the energy can be transferred from the translational motion to the rotational motion. During these collisions, the total angular momentum of colliding particles should be conserved. The requirement that the angular momentum should be conserved together with the Navier-Stokes equations leads to a complete hydrodynamic description of the fluid. Such a complete hydrodynamic description was applied to the fluid composed of finite-sized spherical particles with internal rotational degrees of freedom and it is shown that the friction force becomes memory dependent even for this simple liquid [2].

In anisotropic liquids, the rod-like organic molecules align, on average, along a common direction indicated by a unit vector  $\vec{n}$  called director. For this case, to find the drag force we need to solve the dynamic equations of a nematic liquid crystal LC, i.e., the Ericksen-Leslie equations. In these equations, the independent variables — the director and the fluid velocity — are coupled and this fact causes the

complexity of these equations. Thus, only a few examples with analytical solution exist, e.g., the flow between two parallel plates which defines the different Miesowicz viscosities [3], the Couette flow [4, 5], the Poiseuille flow [6] which was first measured by Cladis et al. [7], or back flow [8]. It is expected that the knowledge of the more or less general solutions of these equations will shed light upon some effects. The solutions of the Ericksen-Leslie equations are also of technological interest since they are indispensable for determining the switching times of liquid-crystal displays.

Every particle immersed in a liquid crystal produces a deformation director field around the particle if the LC molecules are specifically anchored to the closed surface. In the case of a weak anchoring, the area of deformation of the director field around the particle is small and every deformation of the director field can be presented as a small deformation of the ground state, which represents the orientation of all the molecules in one direction.

In the case of a strong anchoring, we have a distortion director field around the immersed particle, which can be called a dipole or quadrupole configuration [9] (figure 1). This configuration directly depends on the strength of coupling with the surface and on the size of particles. In this case, it is necessary to describe the possible configurations and to note that in the long-range distance we have a configuration which shows the same behavior as in the case of weak anchoring represented by a multiple expansion. There exist two approaches to describe the distribution of the director field at a short and long distance from the immersed particle. The first theoretical approach was developed in [9] combining the ansatz functions for the director and the use of the multiple expansion in the far field area. The authors investigated spherical particles with hyperbolic hedgehog and found dipole and quadrupole elastic interactions between such particles. Another approach [10] made it possible to find approximate solutions in terms of the geometrical shape of particles for the case of small anchoring strength and has provided the way to connect the type of the interaction potential with the local symmetry of the director field around the particles [11]. The concept of coating has been introduced that contains all the topological defects located inside and carries the symmetry of the director, and enables us to qualitatively determine the type of the interaction potential. However, the coating is not quantitatively exactly defined. The configuration of director distribution plays a crucial role when the particle moves through a liquid crystal.



**Figure 1.** (Color online) The distortion of the molecules around the spherical particle in the case of the strong anchoring. We can see that the change of the distortion of the director near the particle is very strong. The form of the distortion of the director field in the case of the strong anchoring was theoretically obtained in article [31].

The hydrodynamic solution for the flow of a nematic liquid crystal around a particle at rest, which is equivalent to the problem of a moving particle, still requires its full result. The experiment with the inverted nematic emulsion [12, 13] and investigations by Ruhwandl and Terentjev [14] urged Stark and Ventzki [15–17] to perform Stokes drag calculations for a particle in a nematic environment, especially for the particle-defect dipole. They concentrated on the low Eriksen numbers, where the director field is not affected by the velocity field. The authors presented streamline patterns, interpreted them, calculated Stokes drags for motions parallel and perpendicular to the overall symmetry axis, and compared the results to the Saturn-ring configuration and a uniform director-field. Heuer et al. presented analytical and numerical solutions for both the velocity field and the Stokes drag where the director field was kept uniform [18, 19]. They were the first to investigate a cylinder of infinite length [20]. Diogo [21] put

the velocity field to be the same as the one for an isotropic fluid and calculated the drag force for simple director configurations. He investigated the case where the viscous forces largely exceed the elastic forces from director distortions, i.e., Ericksen numbers much larger than one, as it was explained in the [15]. Roman and Terentjev, have focused on the opposite case. They obtained an analytical solution for the flow velocity in a spatially uniform director field by an expansion in the anisotropy of the viscosities [22]. Chono and Tsuji performed a numerical solution of the Ericksen-Leslie equations around a cylinder determining both the velocity and director field [23]. They found that the director field strongly depends on the Eriksen number, but for homeotropic anchoring their director fields did not exhibit any topological defects required by the boundary conditions signaling about some shortcomings in the exploration. Billeter and Pelcovits used molecular-dynamic simulations to determine the Stokes drag of very small particles [24]. They observed that the Saturn ring is strongly deformed due to the motion of the particles. Ruhwandl and Terentjev have investigated a nonuniform but fixed director configuration, and numerically calculated the velocity field and Stokes drag of a cylinder [25] or spherical particle [14]. The particle was surrounded by the Saturn-ring configuration, and the cylinder was accompanied by two disclination lines. It is known when a particle is surrounded by a disclination ring, the Stokes drag strongly depends on the presence of line defects. There are a few studies that determine both experimentally [26] and theoretically [27–29] the drag force of a moving disclination.

We cannot fully describe all the effects associated with the possible configurations of the director field around the immersed particle, but we attempt to find a general motive of the change dissipation energy of the moving particle in a liquid crystal. First of all, we focus on increasing the effective mass of the immersed colloidal particle and analytically calculate the Stokes drag for colloidal particles in a nematic liquid crystal.

## 2. Theory and details of calculations

The essence of this paper is the calculation of the Stokes drag of a spherical particle in a nematic liquid crystal when the angle between the director and particle velocity is arbitrary. In other words, we have calculated the Stokes drag of the spherical particle in a nematic liquid crystal for the fully anisotropic case. The drag force is caused by the interaction of the particles of the fluid and a foreign body immersed in it. As we mentioned in the introduction, every particle immersed in a liquid crystal is dressed in a deformation coating with the region of deformation of the director field at the distance of the correlation length. The efficacy of the coating was investigated in [30]. To describe this phenomena we can also use the results on the inertial characteristic and viscosity, which present a different approach to the motion of the immersed particles. The inertial characteristic is the effective mass which is an analogue of the hydrodynamical mass in the usual hydrodynamics. Every moving particle immersed in a liquid crystal has two principal different characteristics. One is an inertial characteristic as an effective mass and another characteristic determines the dissipative part. When the particle moves, the region of deformation — i.e., coating, moves too. This causes an increase of its inertia mass. Under these conditions, the effective mass becomes the anisotropic value and can be expressed via formulas [30]:

$$m_{\text{eff}}^{\perp} = m + I \int d\vec{r} \left\{ \left( \frac{\partial \vec{n}}{\partial x} \right)^2 + \left( \frac{\partial \vec{n}}{\partial y} \right)^2 \right\}, \quad (2.1)$$

$$m_{\text{eff}}^{\parallel} = m + I \int d\vec{r} \left( \frac{\partial \vec{n}}{\partial z} \right)^2, \quad (2.2)$$

where  $I$  is density of the moment inertia of the liquid crystal. To determine the inertial characteristic, we can use the distribution of the elastic director around the particles. As was shown in [31], in the case of a weak anchoring, when only small deviations of the director for homeotropic boundary conditions on the surface of a particle are expected, the problem can be linearized, and to describe the director field one can use the two principal angles of a spherical coordinate system  $n_z = \cos \beta(\vec{r})$ ,  $n_x = \sin \beta(\vec{r}) \cos \phi$ , and  $n_y = \sin \beta(\vec{r}) \sin \phi$ , where  $\phi$  is the azimuthal angle, thus respecting an obvious azimuthal symmetry

of the problem. At a small anchoring  $\beta \ll 1$ , the director rotation angle takes the form

$$\beta = \frac{WR}{4K} \left( \frac{R}{r} \right)^3 \sin 2\theta. \quad (2.3)$$

If we substitute the known director field distribution for weak anchoring, we get the value of the inertial effective mass:

$$m_{\text{eff}} = m + \frac{4I}{3} \left( \frac{W}{4K} \right)^2 R^3, \quad (2.4)$$

which can be by an order higher than the mass of the immersed particle [30]. It is analogue of the hydrodynamic mass for the moving particle in an ordinary liquid.

The friction force for a spherical region of a radius  $R/\varepsilon$  with a centered particle within is expressed via formula [15].

The same arguments relate to the viscosity coefficient. The theoretical calculations [16, 17] revealed that the viscosity coefficient depends on the configuration of the director distribution and is much bigger than in an ordinary viscose liquid. The essence of this phenomena can be understood from simple considerations. Every particle that moves in the viscose environment undergoes the action of the additional friction force, which is described by Stokes formula  $f = 6\pi\eta R$ , where  $\eta$  is the friction coefficient, which is associated with a diffusion coefficient of the Brownian particle via the relation  $D = (kT)/(6\pi\eta R)$ . The friction force for the spherical region of radius  $R/\varepsilon$  with a centered particle within is expressed via formula [15]

$$f = 6\pi\eta R \frac{1 - \frac{3\varepsilon}{2} + \varepsilon^3 - \frac{\varepsilon^5}{2}}{\left(1 - \frac{3\varepsilon}{2} + \varepsilon^3\right)^2}. \quad (2.5)$$

From this formula it is easy to see that the friction force increases if the particle is inside the shell. It can be a solvate shell and in the case of the liquid crystal this is the region of a strong change of a director deformation. If we now take into account the configuration of the director distribution around the spherical inclusion, then the diffusion of this inclusion will depend on the direction of the motion with regard to equilibrium director distribution. This leads to the anisotropy of the diffusion coefficient and to a dependence of these coefficients on the conditions of anchoring on the surface of the inclusion. The results of numerical calculations of these phenomena can be found in [16, 17].

To determine the character and the value of the Stokes drag of the spherical particle in a nematic liquid crystal, we can use different distributions of elastic director field around it. The stress tensor  $\sigma_{ik}$  is used to calculate the Stokes drag force [32]. From the known stress tensor  $\sigma_{ik}$ , the drag force can be calculated by the following formula [2]:

$$F_i = \int \sigma_{ij} ds_j. \quad (2.6)$$

The expression for the stress tensor  $\sigma_{ik}$  in a nematic environment is well known and can be found in the literature [32]

$$\sigma_{ik} = -p\delta_{ik} + \sigma_{ik}^{(r)} + \sigma'_{ik}. \quad (2.7)$$

Here,  $p$  is macroscopic pressure,  $\sigma_{ik}^{(r)}$  is "reactive" part of stress tensor and  $\sigma'_{ik}$  is a dissipative part of stress tensor. The expressions for "reactive" and dissipative parts of stress tensor can be found in [32]

$$\sigma_{ik}^{(r)} = -\pi_{kl}\partial_i n_l - \frac{\lambda}{2}(n_i h_k + n_k h_i) + \frac{1}{2}(n_i h_k - n_k h_i), \quad (2.8)$$

$$\begin{aligned} \sigma'_{ik} = & 2\eta_1 v_{ik} + (\eta_2 - \eta_1)\delta_{ik} v_{ll} \\ & + (\eta_4 + \eta_1 - \eta_2)(\delta_{ik} n_l n_m v_{lm} + n_i n_k v_{ll}) \\ & + (\eta_3 - 2\eta_1)(n_i n_l v_{kl} + n_k n_l v_{il}) \\ & + (\eta_5 + \eta_1 + \eta_2 - 2\eta_3 - 2\eta_4) n_i n_k n_l n_m v_{lm}. \end{aligned} \quad (2.9)$$

To find the stress tensor we need to know the solution of the Eriksen-Leslie equations that link the director field and the fluid velocity. The general solution of these equations is a challenge to a theorist.

Here we suggest an approach for finding the stress tensor. As the first step we use the director structure around a colloid particle suspended in a nematic liquid crystal, found in [31]. We assume here the situation when a spherical particle moves slowly and the nematic liquid crystal environment has enough time to relax to the equilibrium state during the motion of a spherical particle. We consider a smooth hard sphere which moves through the fluid with the velocity  $\vec{u}(t) = u(t)\vec{e}_z$ . The fact that it is smooth means that no torques and no force directed tangent to its surface can be exerted on it. Under these conditions, only the component  $\sigma_{rr}$  of the stress tensor contributes to the drag force. Since we use the director field for equilibrium state, the “reactive” part of stress tensor will not contribute to the drag force, but only a dissipative part. It is obvious that the drag force  $\vec{F}$  has the same direction as the velocity of a spherical particle  $\vec{u}$ , and formula (2.6) will reduce to the following:

$$F = \int \sigma'_{rr} \cos \theta ds. \quad (2.10)$$

Substituting the components of director field from [31] and the components for velocity field which are the same as the one for an isotropic fluid [33] in the stress tensor and keeping terms up to the first order of small parameter  $\beta$ , we have obtained the Stokes drag of spherical particle in a nematic environment at weak anchoring

$$\begin{aligned} F = & 4\pi R\eta_1 u + 3\pi R u (\eta_1 + \eta_2 - \eta_3 - \eta_4) \left( -0.27 - 0.02 \frac{WR}{K} \right) \\ & + 3\pi R u (\eta_5 + \eta_1 + \eta_2 - 2\eta_3 - 2\eta_4) \left( 0.11 + \frac{WR}{K} \right). \end{aligned} \quad (2.11)$$

The presented director field structure contains configurations of the director field at a small anchoring. We would like to determine the Stokes drag in the case of strong anchoring. The task of finding a director distribution around a spherical particle consists in minimizing the Frank free-energy functional with boundary conditions provided by it and by the surface energy. Generally, this class of problems is not solvable analytically due to its nonlinearity brought in by the unit-vector constraint  $|\vec{n}(\vec{r})|^2 = 1$ . In [34], in particular, the director distribution was obtained in the one-constant approximation in terms of the multipole expansion. However, the expansion coefficients were not associated with the physical and geometrical parameters of macroparticles. In [10], the director distribution is derived for the general case of different elastic Frank constants and, moreover, the multipole expansion parameters are found in terms of geometric and physical characteristics of macroparticles. Thus, both the behavior and the value of the pair interaction energy are described with no additional restrictions. However, only in [9] there was proposed a theoretical approach combining the ansatz functions for the director field and the use of the multiple expansion in the far field area that was a satisfactory solution to many problems. Thus, the use of the director field in terms of the multipole expansion becomes particularly relevant. In the framework of this approach we have

$$n_x = p_z \frac{x}{r^3} + 2c \frac{zx}{r^5}, \quad n_y = p_z \frac{y}{r^3} + 2c \frac{zy}{r^5}, \quad n_z = 1 - \frac{1}{2} (n_x^2 + n_y^2), \quad (2.12)$$

where the vector  $\vec{p}$  is the dipole moment of the droplet-defect configuration and the parameter  $c$  is the amplitude of the quadrupole moment tensor  $c_{ij}$  of the particle-defect configuration. Assuming that, at small anchoring, the director deviates from its uniform orientation  $\vec{n}_0 \parallel \vec{e}_z$  by only a small amount, we can consider  $n_x$  and  $n_y$  as small parameters. Repeating all the steps as in the first approach, except that now the small parameters are  $n_x$  and  $n_y$ , we obtain the Stokes drag of a spherical particle in a nematic environment at a weak anchoring in the framework of the present approach

$$\begin{aligned} F = & 4\pi R\eta_1 u + (\eta_4 + \eta_3 - \eta_2 - \eta_1) \left[ \cos^2 2\Omega \cdot \frac{0.06u\pi c}{R^2} + \cos 2\Omega \cdot \left( \frac{0.18u\pi c}{R^2} + 0.6u\pi R \right) + 0.2u\pi R \right] \\ & + (\eta_5 + \eta_1 + \eta_2 - 2\eta_3 - \eta_4) \left[ (1 + \sin 2\Omega) (-0.08 \cos^4 \Omega + 0.44 \cos^2 \Omega - 0.12 \sin \Omega - 0.24) \left( \frac{u\pi c}{R^2} \right) \right. \\ & + \left( \frac{u\pi c}{R^2} \right) (0.17 \cos 2\Omega + 0.6 \cos^6 \Omega - 0.12 \cos 4\Omega - 0.06 \cos 6\Omega) + 3u\pi R (0.08 \cos^2 \Omega \cdot \sin^2 \Omega \\ & \left. - 0.06 \sin^4 \Omega) + 0.06u\pi R \sin^2 2\Omega + 0.165u\pi R (3 \cos^4 \Omega - \cos^2 \Omega) \right]. \end{aligned} \quad (2.13)$$

**Table 1.** The viscosities for zero and planar anchoring. The middle column is obtained from equations (2.14) and (2.15) with the Leslie coefficients of 5CB and MBBA [38, 39]; the last one is derived from Stark's numerical calculations [36].

Uniform director		Present work	Numerically exact
5CB	$\eta_{\parallel}$	0.27	0.38
	$\eta_{\perp}$	0.24	0.75
MBBA	$\eta_{\parallel}$	0.28	0.38
	$\eta_{\perp}$	0.27	0.68
Planar anchoring			
5CB	$\eta_{\parallel}$	0.16	–
	$\eta_{\perp}$	0.34	–
MBBA	$\eta_{\parallel}$	0.18	–
	$\eta_{\perp}$	0.36	–

Here,  $\Omega$  is the angle between the director  $\vec{n}_0$  and the velocity of a spherical particle  $\vec{u}$ . This is a general expression for the drag force which includes anisotropy. We can note that it is a very good approximation for a spherical particle with the coating size  $R$ . Outside this region, there are only small deformations and we should take into account the multiple explanation. If we assume that the director field is equal to zero we see that expressions (2.11) and (2.13) become the same, which confirms the rightness of our calculations. In practice, only two directions are measured i.e., along and perpendicular to the director direction. We rewrite the expression (2.13) for these two directions

$$F_{\parallel} = 4\pi\eta_1 u + (\eta_4 + \eta_3 - \eta_2 - \eta_1) \left( \frac{0.24u\pi c}{R^2} + 0.8u\pi R \right) + (\eta_5 + \eta_1 + \eta_2 - 2\eta_3 - 2\eta_4) \left( \frac{0.71u\pi c}{R^2} + 0.33u\pi R \right), \quad (2.14)$$

$$F_{\perp} = 4\pi\eta_1 u + (\eta_4 + \eta_3 - \eta_2 - \eta_1) \left( \frac{-0.12u\pi c}{R^2} - 0.4u\pi R \right) + (\eta_5 + \eta_1 + \eta_2 - 2\eta_3 - 2\eta_4) \left( \frac{-0.59u\pi c}{R^2} - 0.18u\pi R \right). \quad (2.15)$$

Alternative investigation was recently conducted by [35]. In [35] the authors have developed a perturbative approach to the Leslie-Ericksen equations and related the diffusion coefficients to the Miesoviz viscosity parameters  $\eta_i$ . We present our results in the same order as in [35] in table 1. The value of quadrupole moment is used as in [9]. Two cases are considered, i.e., uniform director field and planar anchoring.

### 3. Conclusion

For uniform director field, our results differ from the results in [35, 36], but we believe that our results are reasonable. Our arguments are as follows: in case of a uniform director field, the field of the director is the same in space. The difference in the description of the dynamics of the usual liquid and the liquid crystal is in the expression of the free energy [32]. The expression for the free energy for the liquid crystal contains, in comparison with the expression for the free energy for an ordinary liquid, an additional term, i.e., deformation free energy which depends only on the derivatives of the director regarding the position [32]. In the case of a uniform director field, this term becomes zero and the liquid crystal behaves as an ordinary liquid. Thus, the viscosities along the director  $\eta_{\parallel}$  and perpendicular direction  $\eta_{\perp}$  should be the same. Our results confirm this fact in contradiction to the results of [35, 36]. The possible explanation of this discrepancy might be in the fact that the director evolution depends on the molecular field and the gradient of the velocities [32]. A situation is possible when the molecular field is zero but the gradient

of velocities is big enough (the particle moves quickly) and, consequently, the deviation of the director is not as small as in our approach. If the anchoring is not zero, we find that for both kinds of liquid crystals the ratio  $\eta_{\parallel}/\eta_{\perp}$  is about 2 which agrees well with the general tendency [35]. The approach used by us is valid for a weak anchoring when  $n_x$  and  $n_y$  are small parameters. However, it may be applied to the case of strong anchoring as well. As was shown in [11, 30, 37], under strong anchoring, the effective mass of an ion increases due to the formation of a polarization coating, moving together with the ion. Thus, we can consider the particle with the coating to be a new single moving particle. The anchoring for this “new particle” is weak [11, 30, 37] and the above approach can be applicable too. It should be noted that while extracting the viscosity, the size of the polarization coating should be taken into account. We can conclude that our approach works well for the two limiting cases, i.e., weak and strong anchoring, and it does not include the case of the “middle” anchoring. In [35] there was measured the ratio  $D_{\parallel}/D_{\perp} \approx 4$ . This might be the case of the “middle” anchoring. We cannot claim a complete theory of motion of immersed particles in a nematic liquid crystal, but we suggest the approach which makes the analytical calculation of the diffusion process of a particle in this viscose media possible. This process should take into account the change as an inertial effect and the Stokes drag of a particle in a liquid crystal which are linked with the deformation of the elastic director field.

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## Аналітичний розрахунок сили Стокса для сферичної частинки в нематичному рідкому кристалі

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Як одне з наближень до опису явища руху частинки в анізотропній рідині, ми аналітично рахуємо силу Стокса для сферичних частинок в нематичному рідкому кристалі. Сила Стокса для сферичної частинки порахована для загального анізотропічного випадку в термінах мультипольного розкладу. Для випадку слабого зчеплення ми використовуємо добре відомий розподіл поля директора навколо сферичної частинки. Для випадку сильного зчеплення мультипольний розклад також можна використовувати, якщо модифікувати розмір частинки до розміру шуби. При нульовому зчепленні (однорідне поле директора) ми знайшли, що вязкість вздовж та перпендикулярно до директора є однаковою. Цей результат є правдоподібним, оскільки в цьому випадку рідкий кристал веде себе як ізотропна рідина. Для випадку ненульового зчеплення загальне співвідношення  $\eta_{\parallel}/\eta_{\perp}$  є близько 2, що задовільняє експериментальні результати.

**Ключові слова:** сила Стокса, рідкі кристали, дифузія, вязкість