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MECHANISMS FOR ANOMALOUS DIFFUSION IN A NEMATIC ENVIRONMENT

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Mechanisms for anomalous diffusion of colloidal particles in a nematic environment are theoretically investigated. It is shown that thermal fluctuations of the nematic director may couple to the translational and orientational motions of particles, which leads to anomalous diffusion. Both superdiffusion, when the mean square displacement increases with the time faster than linearly, and subdiffusion, when this dependence is slower than linear, are possible. For micrometer-sized particles, the anomalous diffusion effects are expected on millisecond time scales.

K e y w o r d s: anomalous diffusion, nematic environment

1. Introduction

Transport phenomena are ubiquitous in nature and play the essential role in physics, chemistry, and biology. Microscopically, particles embedded in a fluid, be they molecules, agglomerates, or even live bacteria, are subject to a random motion as a result of their random collisions with the surrounding particles. This thermal motion, first described by Scottish botanist Robert Brown in 1828 [1], is strongly influenced by the properties of the surrounding medium. The physical approach to the Brownian motion, developed in the beginning of the 20th century by Einstein [2], Smoluchowski [3], and Langevin [4], still forms the basis for our understanding of this stochastic dynamics. The main result first derived by Einstein [2] consists in that the mean square displacement (MSD) $\langle \Delta \mathbf{r}^2 \rangle$ of a particle undergoing the Brownian motion in a Newtonian fluid increases linearly with the time,

$$\langle \Delta \mathbf{r}^2(t) \rangle = 6Dt, \tag{1}$$

where D is the diffusion constant. For a spherical particle with hydrodynamic radius R in a fluid with viscosity η, the diffusion coefficient is given by the Stokes–Einstein relation

$$D = k_{\mathrm{B}}T/\zeta, \tag{2}$$

where k_{B} is the Boltzmann constant, T the temperature, and ζ the Stokes friction coefficient, which is given under no-slip conditions by

$$\zeta = 6\pi\eta R. \tag{3}$$

These results are valid for the Brownian motion under the influence of two forces, the viscous frictional force linear in the particle velocity, $-\zeta \mathbf{V}$, and a random force $\mathbf{F}(t)$ with white-noise-spectrum due to random collisions with surrounding particles. The corresponding stochastic equation of motion for a Brownian particle with mass m, known as the Langevin equation [4], reads

$$m\frac{d\mathbf{V}}{dt} = -\zeta \mathbf{V} + \mathbf{F}(t), \tag{4}$$

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whose solution for the velocity autocorrelation function reads (see, e.g., [5])

$$C_{\mathbf{V}}(t) \equiv \langle \mathbf{V}(0) \cdot \mathbf{V}(t) \rangle = \langle V^2 \rangle e^{-t/\tau}, \tag{5}$$

where $\tau = m/\zeta$ is the correlation time, and the average thermal velocity V follows from the equipartition theorem, $m\langle V^2 \rangle/2 = 3k_{\mathrm{B}}T/2$. The MSD along, say, the x-axis is expressed through the velocity autocorrelation function as follows [6]:

$$\langle \Delta x^2(t) \rangle = 2 \int\limits_0^t dt' \int\limits_0^{t'} C_{V_x}(t'')\, dt''. \tag{6}$$

Conversely,

$$C_{V_x}(t) = \frac{1}{2} \frac{d^2 \langle \Delta x^2(t) \rangle}{dt^2}. \tag{7}$$

Evidently, the normal diffusion regime of Eq. (1), i.e., $\langle \Delta x^2(t) \rangle = 2Dt$, is established on the time scale $t \gg \tau$, where $C_{\mathbf{V}}(t) = 0$ (τ is usually a submicrosecond time). If $C_{\mathbf{V}}(t) \gtrless 0$ for some reason, then, depending on the sign of $C_{\mathbf{V}}(t)$, the MSD will increase slower ($C_{\mathbf{V}}(t) < 0$, subdiffusion) or faster ($C_{\mathbf{V}}(t) > 0$, superdiffusion) than linearly with the time. Such diffusion would be anomalous, as it deviates from the normal behavior expressed by Eq. (1).

If the Brownian motion occurs in a system with additional degrees of freedom that exhibit a relatively slow dynamics (such as the relaxational dynamics in polymers), then this dynamics may couple to the Brownian dynamics, introducing a certain correlation into the particle motion. For instance, the dynamics of colloidal particles in polymer networks may couple to the Rouse relaxation dynamics of the polymer and exhibit, as a result, the subdiffusive behavior $\langle \Delta \mathbf{r}^2(t) \rangle \propto t^{\alpha}$, where $\alpha < 1$ [7]. Superdiffusion with $\alpha > 1$ was observed in other kinds of polymer systems, the so-called "living polymers" [8]. Even in water, the interaction with hydrodynamic modes leads to long negative "tails" in $C_{\mathbf{v}}(t)$ [9], so that the Brownian motion is, in fact, (slightly) subdiffusive even in normal liquids. It is natural to expect deviations from the normal diffusion of Eq. (1) in soft matter systems, since soft matter typically possesses slow degrees of freedom, whose dynamics may couple with the Brownian dynamics.

Colloidal dispersions in nematic liquid crystals (NLC) have recently emerged as a novel type of soft matter [10]. A particle immersed in a nematic environment distorts a local ordering of the liquid crystal, which leads to a number of intricate effects even concerning the static properties of such particles [10]. The nematic phase is anisotropic [11], so that the translational diffusion is also anisotropic with two independent diffusion constants $D_{\parallel}$ and $D_{\perp}$ that correspond to the diffusion in parallel and perpendicularly to the nematic director $\mathbf{n}$ [12]. Compared to isotropic liquids, the liquid crystals exhibit a substantial additional dynamics of relatively slow director fluctuations. It is due to these fluctuations or rather the concomitant birefringence fluctuations that a bulk NLS strongly scatters light and, therefore, looks turbid. The characteristic relaxation time depends on the configuration of the NLC system. In a thin-film configuration such as that in liquid crystal displays, the characteristic time (which actually determines the display reaction time) is of the order of milliseconds. One then expects that, on such millisecond time scales, the liquid crystal dynamics may influence the diffusion dynamics of colloidal particles. In this work, we theoretically analyze possible mechanisms and consequences of such effects and show that they may lead to anomalous sub- and superdiffusion.

In the next section, we summarize the (known) results concerning the nematic relaxation dynamics; in the subsequent section, the coupling of these dynamics to the Brownian motion is analyzed.

2. Thermal Fluctuations in a Nematic Medium

In addition to hydrodynamic modes typical of normal liquids, the nematic environment supports director fluctuation modes, whose spectrum shall now be analyzed. The equations of nematodynamics are significantly non-linear [11] and do not allow for general analytical solutions. However, the relaxational dynamics of NLC and their power spectrum can be analytically obtained under simplifying assumptions. Let us consider a bulk NLC in the absence of external fields, and neglect flow and inertial effects. The equation of motion for the director fluctuations $\delta\mathbf{n}$ reduces to the torque equation that balances the viscous and elastic torques [11, 13, 14],

$$\gamma_1 \frac{\partial}{\partial t} \delta\mathbf{n} = K \nabla^2 \delta\mathbf{n}, \tag{8}$$

where γ_1 is the rotational viscosity, and K is the effective (average) elastic constant. Performing the space-time Fourier transformation of Eq. (8) yields the dispersion relation with a purely imaginary frequency $\omega = -iK\mathbf{q}^2/\gamma_1$ ($\mathbf{q}$ is the wave vector), which implies that the fluctuation modes are overdamped and, thus, purely relaxational with the relaxation time

$$\tau_{\mathbf{q}} = \gamma_1 / Kq^2. \tag{9}$$

The fluctuation dynamics thus occur on *all* time scales, depending on the length scales of fluctuations, as expressed by the wavenumber q. Adding an external source (force) to Eq. (8), one solves the equation for the response (susceptibility) function ([14]), which leads through the fluctuation-dissipation theorem to the following fluctuation power spectral density [14]:

$$I_{\mathbf{n},\mathbf{q}}(\omega) = \frac{k_{\mathrm{B}}T}{Kq^2} \frac{\tau_{\mathbf{q}}}{1 + (\omega\tau_{\mathbf{q}})^2}. \tag{10}$$

In what follows, we will be interested in the autocorrelation function of the director fluctuations, $C_{\mathbf{n},\mathbf{q}}(t) = \langle \delta\mathbf{n}_{-\mathbf{q}}(0)\delta\mathbf{n}_{\mathbf{q}}(t)\rangle$. Recalling that the fluctuation power spectrum equals the Fourier transform of the fluctuation autocorrelation function (Wiener-Khinchin theorem), the sought autocorrelation function is the inverse Fourier transform of Eq. (10),

$$C_{\mathbf{n},\mathbf{q}}(t) \equiv \langle \delta\mathbf{n}_{-\mathbf{q}}(0)\delta\mathbf{n}_{\mathbf{q}}(t)\rangle = \frac{2}{\pi}\int_0^\infty I_{\mathbf{n},\mathbf{q}}(\omega)\cos\omega t\, d\omega =$$

$$= \frac{k_{\mathrm{B}}T}{Kq^2}\, e^{-t/\tau_{\mathbf{q}}}. \tag{11}$$

It should be noted that, under the implied approximations, the correlation function of Eq. (11) is unphysical at short times, whereas a physically consistent, even in time, autocorrelation function may not have discontinuous derivatives at the origin. A related inconsistency is already seen in Eq. (9), which predicts an arbitrarily fast $\tau_{\mathbf{q}}$ as $q \to \infty$. Conversely, this yields fluctuations at arbitrarily high frequencies, which is a consequence of the neglect of inertial effects. Evidently, the description of Eq. (8) is invalid on the molecular length scales l (e.g., the molecular length of a classical liquid crystal 5CB is $l \sim 2$ nm), which corresponds to $q_{\max} \simeq 2\pi/l$ in the reciprocal space. With the typical values of viscous and elastic constants, $\gamma_1 \sim 20$ mPa and $K \sim 10$ pN [11], one then estimates from Eq. (9) that the short-time limit of the validity of the above description is $t_{\min} \sim 0.2$ ns, which is clearly beyond the time scales we are interested in. The correct asymptotic behavior of $C_{\mathbf{n},\mathbf{q}}(t)$ at $t \lesssim t_{\min}$ follows from the boundedness of its power spectrum in frequency. This requires the first initial derivative of $C_{\mathbf{n},\mathbf{q}}(t)$ be zero, and the second derivative be finite and negative. This has implications for the director angular velocity correlation function, discussed below.

By symmetry, there are two uncoupled fluctuation modes with orthogonal polarizations and similar spectra of Eq. (10) [14]. In the first mode, $\delta\mathbf{n}_1$ is parallel to the $(\mathbf{n}, \mathbf{q})$ plane. In the second mode, $\delta\mathbf{n}_2$ is orthogonal to that plane. Significantly, these modes involve different types of deformations and, thus, couple differently to the diffusion dynamics we are interested in. Specifically, mode 1 involves bend and splay deformations, whereas mode 2 involves bend and twist ones. (The deformations are classified as follows: $\nabla\mathbf{n} \neq 0$ for splay, $\nabla \times \mathbf{n} \perp \mathbf{n}$ for bend, and $\nabla \times \mathbf{n} \parallel \mathbf{n}$ for twist deformations.) Because of the different deformations involved, the effective elastic constant K in Eqs. (9), (10), and (11) will be different for the two modes, so their time scales will be somewhat different as well.

3. Coupling of Nematic Fluctuations and Brownian Dynamics

A particle embedded in NLC distorts the local director order, by creating a topological defect. In the case of a spherical particle with homeotropic boundary condition (i.e., the director at the particle surface is orthogonal to the surface), two commonly encountered director configurations around the particle are of the dipole and quadrupole symmetries, respectively [15]. We will mostly concentrate the attention on dipolar inclusions that behave themselves as elastic dipoles with the dipole moment $\mathbf{P}$, which is oriented in parallel to the (undistorted) local director $\mathbf{n}$, has the magnitude $P = aR^2$ (R is the particle radius, $a = 2.04$ [15]), and therefore interacts with inhomogeneities of the director field that arise due to thermal fluctuations. The effect is two-fold: the particle influences the local director field and its dynamics, and *vice versa*. To solve the problem mathematically, one would have to solve the coupled equations of motion of a particle (Newton's equation) and nematic (Ericksen–Leslie's equations) dynamics with the ap-

propriate boundary conditions at the particle and cell surfaces. Here, we do not attempt to rigorously solve this formidable problem, but rather analyze it under simplifying conditions. First, we neglect the effect of the particle dynamics on the nematic. Similarly, we assume that the nematic director dynamics at the particle is the same as that in the far field of a bulk NLC and is expressed by Eqs. (10) and (11).

The energy of interaction between $\mathbf{P}$ and $\mathbf{n}$ reads [15]:

$$U = -4\pi K \mathbf{P}\cdot\mathbf{n}\,\nabla\cdot\mathbf{n} = -4\pi K P\,\nabla\cdot\mathbf{n}, \tag{12}$$

where the last equality follows from the fact that $\mathbf{P}$ prefers to orient along the local director $\mathbf{n}$, and, therefore, $\mathbf{P}\cdot\mathbf{n} = P$. If $\nabla\cdot\mathbf{n} \neq 0$ (splay deformation) due to fluctuations, then there appears a force on the particle,

$$\mathbf{F} = -\nabla U = 4\pi K P \nabla(\nabla\cdot\mathbf{n}) = 4\pi K P\,\nabla(\nabla\cdot\delta\mathbf{n}), \tag{13}$$

where it is assumed that the director distortions are fluctuations $\delta\mathbf{n}$ in an otherwise uniform director field, so that $\nabla\cdot\mathbf{n} = \nabla\cdot\delta\mathbf{n}$. Under the action of this force and neglecting inertial effects, the particle will start to move with a velocity $\mathbf{v}$, so that the viscous force balances the force $\mathbf{F}$ from director fluctuations, $\mathbf{F} = \zeta\mathbf{v}$. Thus,

$$\mathbf{v} = \frac{4\pi K P}{\zeta}\nabla(\nabla\cdot\delta\mathbf{n}). \tag{14}$$

Fourier-transforming Eq. (14) into the reciprocal space, director fluctuation component $\delta\mathbf{n}_\mathbf{q}$ with wave vector $\mathbf{q}$ results in the particle velocity $\mathbf{v}_\mathbf{q}$ given by the following relation:

$$\mathbf{v}_\mathbf{q} = \frac{4\pi K P}{\zeta}\,\mathbf{q}(\mathbf{q}\cdot\delta\mathbf{n}_\mathbf{q}). \tag{15}$$

The particle velocity autocorrelation function is then

$$C_{\mathbf{v},\mathbf{q}}(t) = \langle \mathbf{v}_{-\mathbf{q}}(0)\cdot\mathbf{v}_\mathbf{q}(t)\rangle =$$

$$= \left(\frac{4\pi K P}{\zeta}\right)^2 q^2 \langle \mathbf{q}\cdot\delta\mathbf{n}_{-\mathbf{q}}(0)\,\mathbf{q}\cdot\delta\mathbf{n}_\mathbf{q}(t)\rangle. \tag{16}$$

Note that the splay deformations leading to the effects described above exist only in the 1st fluctuation mode (cf. the previous section), for which $\delta\mathbf{n}$ is in the $(\mathbf{n},\mathbf{q})$ plane and perpendicular to $\mathbf{n}$, so that $\mathbf{q}\cdot\delta\mathbf{n} = q\,\delta n\sin\theta$, where θ is the angle between $\mathbf{q}$ and $\mathbf{n}$. Equation (16) can thus be re-written as

$$C_{\mathbf{v},\mathbf{q}}(t) = \left(\frac{4\pi K P}{\zeta}\right)^2 q^4 \sin^2\theta\,\langle \delta\mathbf{n}_{-\mathbf{q}}(0)\,\delta\mathbf{n}_\mathbf{q}(t)\rangle =$$

$$= A\,q^2\sin^2\theta\; e^{-t/\tau_\mathbf{q}}, \tag{17}$$

where we substituted the director fluctuation correlation function $C_{\mathbf{n},\mathbf{q}}(t)$ from Eq. (11) and absorbed all the constant pre-factors into $A = (4\pi P/\zeta)^2 K k_\mathrm{B}T$.

Equation (17) describes the contribution of thermal director fluctuations with wavevector $\mathbf{q}$ to the particle velocity autocorrelation function. To obtain the full velocity correlation function, Eq. (17) has to be integrated over $\mathbf{q}$. (Note that fluctuations with different $\mathbf{q}$ are uncorrelated.) Integration has to only involve fluctuations occurring on length scales large as compared with the particle size $d = 2R$, which corresponds to wavenumbers smaller than $q_d = 2\pi/d$. Thus,

$$C_\mathbf{v}(t) = A \int\limits_{q<q_d} q^2\sin^2\theta\; e^{-t/\tau_\mathbf{q}}\,d\mathbf{q}. \tag{18}$$

Evidently, the correlation function $C_\mathbf{v}(t)$ is positive and thus corresponds to the superdiffusive behavior. On which time scales are such anomalous diffusion effects expected? In other words, what is the correlation time of the correlation function (18)? The correlation time of a normalized correlation function is, by definition, its time integral. For the correlation function above, it is easy to obtain

$$\tau_\mathrm{corr} = C_\mathbf{v}^{-1}(0)\int\limits_0^\infty C_\mathbf{v}(t)\,dt = \frac{5}{3}\,\frac{\gamma_1}{K q_d^2}. \tag{19}$$

With the typical values of viscous and elastic constants, $\gamma_1 \sim 20$ mPa and $K \sim 10$ pN [11], and for a 5-μm particle, $\tau_\mathrm{corr} \sim 2$ ms. Thus, the superdiffusive behavior of micron-sized particles due to the coupling to the director fluctuation dynamics is expected to occur on millisecond time scales.

The coupling mechanism between director fluctuations and Brownian dynamics, discussed above, is only functional for the fluctuations that involve splay deformations; in particular, such deformations only occur in the 1st (of the two) fluctuation mode (cf.

the previous section). Can the fluctuations that involve other types of deformations affect Brownian dynamics? Bend and twist fluctuations are basically director rotations. So, by symmetry, they cannot directly couple to the particle translation. However, they can obviously couple to the particle *rotation*, influencing thereby the *rotational* Brownian motion. Microscopically, a rotating director field exerts a torque on the particle, which is proportional to the director change rate $\dot{\mathbf{n}}$. In response, the particle acquires an angular velocity proportional to the applied torque, so that it is balanced by the viscous friction torque. As a result, the particle *angular* velocity correlation function will be proportional to the director angular velocity correlation function $C_{\dot{\mathbf{n}}}(t) = \langle \dot{\mathbf{n}}(0)\dot{\mathbf{n}}(t)\rangle$.

In the case of anisotropic diffusion, the translational and rotational motions of a diffuser are coupled (Ref. [5], p. 149; [16]). Another possible mechanism of rotation-translation coupling is specific to NLC and related to the nonlinear Stokes drag and its coupling to rotations [17, 18]. One more mechanism of rotational-translational coupling appears in a vicinity of boundaries, such as walls of the containing cell, through hydrodynamic interactions with the walls. Overall, we thus expect that the rotational Brownian dynamics, insofar it is influenced by the nematic fluctuations, is reflected in the *translational* Brownian motion. Specifically, the particle velocity v acquires a contribution proportional to the director angular velocity $|\dot{\mathbf{n}}(t)|$, so that the particle velocity autocorrelation function becomes proportional to the director angular velocity correlation function,

$$C_{\mathbf{v},\mathbf{q}}(t) = c\,\langle \dot{\mathbf{n}}_{-\mathbf{q}}(0)\dot{\mathbf{n}}_{\mathbf{q}}(t)\rangle, \tag{20}$$

where c is a coupling constant. Following the same reasoning as that prior to Eq. (18), the full correlation function is

$$C_{\mathbf{v}}(t) = c \int\limits_{q<q_d} \langle \dot{\mathbf{n}}_{-\mathbf{q}}(0)\dot{\mathbf{n}}_{\mathbf{q}}(t)\rangle\, d\mathbf{q}. \tag{21}$$

Recalling the well-known theorem of statistical physics, which states that, for any mechanical property X that is a function on the phase space of a classical many-particle system, there holds $\langle \dot{X}(0)\dot{X}(t)\rangle = -d^2/dt^2\,\langle X(0)X(t)\rangle$ [19], it follows that the director angular velocity correlation function is expressed through the correlation function of Eq. (11) as $C_{\dot{\mathbf{n}},\mathbf{q}}(t) = -d^2/dt^2\, C_{\mathbf{n},\mathbf{q}}(t)$, so that

$$C_{\dot{\mathbf{n}},\mathbf{q}}(t) = -\frac{k_{\mathrm{B}}T}{Kq^2\tau_{\mathbf{q}}^2}\, e^{-t/\tau_{\mathbf{q}}}. \tag{22}$$

Clearly, the director angular velocity autocorrelation function $C_{\dot{\mathbf{n}},\mathbf{q}}(t)$ is negative. This is easy to understand in view of the fact that the director fluctuations are only small angular excursions from the mean, so that if the director rotates in a certain direction at a given instant of time, it has to be rotating back at a later time, which means a negative angular velocity autocorrelation. At short times, however, the relation (22) is not valid, as the initial value of autocorrelation function must be positive. As was discussed in relation to Eq. (11), the correct asymptotic behavior of $C_{\mathbf{n},\mathbf{q}}(t)$ is such that its initial second derivative is negative, so that $C_{\dot{\mathbf{n}},\mathbf{q}}(t)$ is positive at short times, as it should be.

Substituting Eq. (22) in (21), we obtain

$$C_{\mathbf{v}}(t) = -B \int\limits_{q<q_d} q^2 e^{-t/\tau_{\mathbf{q}}}\, d\mathbf{q}, \tag{23}$$

where $B = ck_{\mathrm{B}}TK\gamma_1^{-2}$. The correlation function is *negative*, so that this mechanism of coupling between the director fluctuation and the Brownian dynamics leads to the subdiffusion. Similarly to Eq. (19), the correlation time of (23) is evaluated as $\tau_{\mathrm{corr}} = \frac{5}{3}\,\frac{\gamma_1}{Kq_d^2}$, so that this sort of anomalous Brownian dynamics is likewise expected on millisecond time scales.

Thus, there are two mechanisms of coupling between the nematic director and the Brownian dynamics that lead to anomalous diffusion. The first mechanism, discussed here for colloidal inclusions with dipolar symmetry, is a direct coupling through interactions of the elastic dipole with transient gradients of the director field that appear due to thermal fluctuations. This results in positive velocity autocorrelation tails and, thus, to the superdiffusive behavior of particles. The second mechanism is an indirect coupling, whereby director reorientations couple to particle rotations and further, through a rotation-translation coupling, to particle translation. This effect results in negative velocity autocorrelation tails and, therefore, to subdiffusion. Both effects occur on similar time scales. Thus, their interplay, depending on their

relative strength and characteristic times, may lead to various intricate scenarios of particle dynamics. Depending on material parameters, the time scales of the sub- and superdiffusive effects may actually be well separated, as they originate from different, uncoupled nematic fluctuation modes. In the case of a strong anisotropy, the relevant elastic constants may be significantly different for the two modes, so that their characteristic time scales will be different as well.

4. Conclusions

In conclusion, the Brownian dynamics in a nematic environment is theoretically analysed. It is shown that thermal fluctuations of additional, with respect to those of an isotropic fluid, degrees of freedom, namely the nematic director fluctuations, may couple to particle's translation and rotation, influencing thereby the Brownian dynamics. On certain time scales, this leads to anomalous diffusion effects. Both superdiffusion (when the mean square displacement increases with the time faster than linearly) and subdiffusion (when this dependence is slower than linear) are possible. For micrometer-sized particles, the anomalous diffusion effects are expected on millisecond time scales.

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МЕХАНІЗМИ АНОМАЛЬНОЇ ДИФУЗІЇ
В НЕМАТИЧНОМУ СЕРЕДОВИЩІ

Р е з ю м е

Теоретично досліджено механізми аномальної дифузії колоїдних частинок у нематичному середовищі. Показано, що теплові флуктуації директора рідкого кристала можуть зв'язуватись з поступальним та обертальним рухом частинок, приводячи до аномальної дифузії. Можлива як супердифузія, коли середньоквадратичне зміщення зростає з часом швидше, ніж лінійно, так і субдифузія, коли ця залежність повільніша від лінійної. Для мікронних частинок режим аномальної дифузії передбачається на мілісекундних часових масштабах.