# THE SPECTRUM OF THE CORRELATION FUNCTION FOR FLUCTUATIONS OF THE ANISOTROPY TENSOR OF A LAGRANGIAN LIQUID PARTICLE

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The spectrum of the correlation function (CF) for thermal hydrodynamic fluctuations of the anisotropy tensor of a liquid Lagrangian particle is studied in detail. Such a correlation function represents an asymptotic estimate for a molecular correlation function of orientational variables which is important for investigating the thermal motion of anisotropic liquid molecules. The Euler correlation function for the anisotropy tensor is obtained from the Langevin equations with random fluctuating fields of the velocity and its rotor. The corresponding Lagrangian correlation function was obtained on the basis of the approach proposed by N.P. Malomuzh.

## 1. Introduction

The mechanism of rotational heat motion of liquid molecules describes the phenomena of dielectric relaxation, IR absorption, Raman and Rayleigh light scattering, etc. in terms of temporal CFs of molecular variables which are responsible for the rotational motion of nonspherical liquid molecules. The accurate calculation of these or any other molecular CFs is impossible in view of the difficulties associated with a description of many-particle systems. It is usual to restrict oneself with searching for the Lagrangian CFs of the corresponding fluctuation fields that represent good long-time asymptotics of the CFs of molecular variables [1] and can be obtained with the help of various model conceptions [2–9]. According to experimental data and results of computer simulations carried out using the molecular dynamics technique [10], these asymptotics turn out to be sufficient for a rather wide range of hydrodynamically short time intervals. Moreover, according to [1], the range of applicability of these asymptotic estimates can be essentially expanded towards a short-time region under the condition of refining the investigated hydrodynamic models and the corresponding theoretical apparatus.

Hence, the investigation of the mechanism of heat motion of anisotropic liquid molecules requires constructing the Lagrangian CFs for fluctuations of orientational variables. The theory of thermal hydrodynamic fluctuations developed on the basis of hydrodynamic equations in the Lagrangian form turns out to be rather complicated in use as its equations are strongly nonlinear. The calculation of the Lagrangian CFs with the help of this system is associated with essential mathematical difficulties. That's why one tries to construct the Lagrangian CFs associating them with the corresponding Euler ones as the theory of thermal hydrodynamic fluctuations for the latter is well developed. Such an approach was first formulated in [1] and applied in a number of papers [6–9]. A modified technique of calculating the fluctuations of Lagrangian variables in terms of the fluctuations of the Euler field of hydrodynamic variables was recently proposed by N.P. Malomuzh and co-authors [11–15]. In the new approach, the operator which converts the Euler CFs for the fluctuation field to the Lagrangian ones is integral, in contrast to [1], where it is differential. This fact is not essential for the analysis of the long-time behavior of the Lagrangian CFs, and both versions of the Lagrangian theory of thermal fluctuations are equivalent. The

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difference in the temporal behavior appears only in the range of short time intervals (shorter than or comparable to the relaxation time of the shear viscosity of liquid). In this case, the approach proposed by N.P. Malomuzh has a number of advantages as it allows one to get rid of the problems concerning the appearance of singular contributions and the necessity of their physical interpretation. It must improve the agreement between the molecular and Lagrangian CFs. The choice of a specific Euler theory of thermal hydrodynamic fluctuations is also of importance.

One of the most perfect Euler theories, which are able to describe thermal hydrodynamic fluctuations of a liquid consisting of nonspherical molecules, is the phenomenological Leontovich theory [16]. In this theory, it is assumed that the state of liquid at any point can be described by a set of ordinary hydrodynamic variables and the anisotropy tensor that characterizes the deviation of the axes of anisotropic molecules in the volume element from the isotropic distribution. The introduction of the anisotropy tensor turns out to be very effective for describing the thermal motion of liquid and investigating the spectral composition of light scattered by it [4, 17]. A total system of hydrodynamic equations was recently constructed in [6] with the anisotropy tensor introduced as a new field variable and interpreted as a local inertia tensor of the volume element.

The Euler CFs for the thermal fluctuations of local macroscopic variables can be also studied by means of solving the inhomogeneous linear Langevin equations that describe the dynamics of fluctuations [10,16,18–20]. For this purpose, the macroscopic equations of the thermodynamics of irreversible processes linearized in the neighborhood of a locally equilibrium state are supplemented with the correlated Gaussian fields of fluctuating flow components (random extrinsic sources of thermal noise). Statistical properties of the sources are determined by the fluctuation-dissipative theorem.

In the present paper, we analyze the spectrum of the CFs for thermal hydrodynamic fluctuations of the anisotropy tensor of a Lagrangian liquid particle. Its insensibility to the dispersion of kinetic coefficients is proved. The corresponding Euler CF was obtained with the help of the Langevin technique of constructing a theory of equilibrium thermal fluctuations. In this case, the fluctuating velocity field and its rotor were considered as random fields. The presented results are based on the technique of converting the Euler CFs to the Lagrangian ones, which was proposed by N.P. Malomuzh.

## 2. Lagrangian Correlation Function

The method of derivation of Lagrangian CFs proposed by N.P. Malomuzh is based on the account of a non-local character of the relations between Lagrangian and Euler variables. According to [11], the transition from the Euler CFs for fluctuation fields to the Lagrangian ones is accomplished by the following rule:

$$\psi_A^{\mathrm{L}}(t) = \frac{1}{V_{\mathrm{L}}^2}\int\limits_{V_{\mathrm{L}}} d\vec{r}_1 \int\limits_{V_{\mathrm{L}}} d\vec{r}_2 \times$$

$$\times \int d\vec{s}\, W^{(\mathrm{G})}(\vec{s})\, \langle A(\vec{r_1}+\vec{s},t)A(\vec{r}_2,0)\rangle\,, \tag{1}$$

where $\psi_A^{\mathrm{E}}(\vec{r},t) = \langle A(\vec{r_1},t)A(\vec{r}_2,0)\rangle$ is the Euler autocorrelation function for the fluctuation field $A$, $\vec{r}=\vec{r}_2-\vec{r}_1$, $V_{\mathrm{L}}=\frac{4}{3}\pi r_{\mathrm{L}}^3$ is the volume of a Lagrangian particle, while the distribution function for the displacements $\vec{s}$ of a Lagrangian particle is assumed to be Gaussian:

$$W^{(\mathrm{G})}(\vec{s}) = \left(\frac{2}{3}\pi\Gamma(t)\right)^{-3/2} \exp\left(-\frac{3\vec{s}^{\,2}}{2\Gamma(t)}\right), \tag{2}$$

where $\Gamma(t)$ is the root-mean-square displacement of a particle.

Passing to the Fourier-transform of the Euler autocorrelation function

$$\langle A(\vec{r_1},t)A(\vec{r}_2,0)\rangle =$$

$$= \int \frac{d\vec{q}}{(2\pi)^3}\int\limits_{-\infty}^{+\infty}\frac{d\omega}{\pi}\psi_A(\vec{q},\omega)e^{i\vec{q}(\vec{r_1}-\vec{r}_2)-i\omega t}, \tag{3}$$

where $\psi_A(\vec{q},\omega)$ is the corresponding spectral intensity, and substituting (3) into (1) under the assumption that the displacement of a liquid Lagrangian particle can be neglected (like it was done in [1, 11]), we obtain the spectrum of the Lagrangian CF for the fluctuation field $A$ as

$$\psi_A^{\mathrm{L}}(\omega) =$$

$$= \frac{8r_{\mathrm{L}}^2}{\pi V_{\mathrm{L}}^2}\int\limits_0^{\infty}\frac{1}{q^2}\left(\cos qr_{\mathrm{L}} - \frac{\sin qr_{\mathrm{L}}}{qr_{\mathrm{L}}}\right)^2 \psi_A(\vec{q},\omega)dq. \tag{4}$$

Thus, if the spectral intensities of fluctuations of the Euler fields are known, one can recover the spectra of the Lagrangian CFs for these fluctuations. The explicit form of the spectra depends on the hydrodynamic model being used.

## 3. Spectral Intensity of Hydrodynamic Fluctuations of the Anisotropy Tensor

Let's proceed from the Langevin equation for the fluctuations of the anisotropy tensor in terms of spherical components

$$I_{\pm 2} = \frac{1}{4}(I_{xx} - I_{yy} \pm 2iI_{xy}), \quad I_{\pm 1} = \frac{1}{2}(I_{xz} \pm iI_{yz}),$$

$$I_0 = -(I_{xx} + I_{yy}). \tag{5}$$

In this case, the fluctuating velocity field and its rotor $\vec{\omega} = \frac{1}{2}\,\mathrm{rot}\vec{v}$ should be considered as random fields. The spatial and temporal evolutions of the density of the anisotropy tensor are connected to the convective transport and rotation and obey the stochastic equation

$$\frac{\partial I_m}{\partial t} = -v_a \nabla_a I_m - \frac{1}{\tau} I_m - i \sum_{b=-2}^{2} (\vec{\omega}\hat{\vec{L}})_{mb} I_b, \tag{6}$$

where $\hat{\vec{L}}$ is the operator of the orbital moment. In the general case, this equation must include the contributions proportional to the gradient of the flow velocity, but their role is insignificant. The stochastic properties of disturbances (those of the velocity field and its rotor) induce the convection and orientational relaxation and allow one to consider them to be rapidly fluctuating as compared to the response. This fact enables one to find the correlation function for fluctuations of the anisotropy tensor using the technique proposed in [21,22]. This technique consists in substituting the result of formal integration of (6) over time into the right-hand side of the same equation and carrying out the averaging over various realizations of the velocity field with the separation of averages by the rule

$$\langle v_a(\vec{r},\, t)\, v_b^*(\vec{r}, t')\, I_c(\vec{r}, t')\rangle \approx \langle v_a(\vec{r},\, t)\, v_b^*(\vec{r}, t')\rangle\, \overline{I}_c(\vec{r}, t'),$$

$$\langle v_a(\vec{r},\, t)\rangle = 0. \tag{7}$$

As a result, we obtain a coarsened equation for the Euler field of the anisotropy tensor [18,23]

$$\frac{\partial}{\partial t}\overline{I}_m(\vec{r},\, t) = \int_0^t dt' D(t-t')\Delta \overline{I}_m(\vec{r},\, t') -$$

$$- \int_0^t dt'\, L(t-t')\overline{I}_m(\vec{r},\, t'), \tag{8}$$

with the following notations introduced for the collective coefficients of translational diffusion and relaxation:

$$D(t-t') = \frac{1}{3}\,\langle \vec{v}(\vec{r},\, t)\vec{v}(\vec{r},\, t')\rangle\,,$$

$$L(t-t') = \frac{1}{\tau}\,\delta(t-t') + 2\,\langle \vec{\omega}\,(\vec{r}, t)\vec{\omega}(\vec{r}, t')\rangle\,. \tag{9}$$

They are determined by the Euler CFs taken at one spatial point. For the spectral intensity of fluctuations of the anisotropy tensor, the last equation easily yields

$$\psi_{I_m}(q, \omega) = \left\langle \overline{I}_m(\vec{r},\, t)\overline{I}_m^*(\vec{r}\,', t')\right\rangle_{q\omega} =$$

$$= \mathrm{Re}\frac{\left\langle \left|\overline{I}_m(q)\right|^2 \right\rangle}{-i\omega + L(\omega) + q^2 D(\omega)}. \tag{10}$$

The frequency-dependent diffusion coefficients and the relaxation time can be simulated, for example, by means of the spectra of the Lagrangian fluctuation fields of the flow velocity and the rotor of liquid neglecting the influence of the anisotropy field on the velocity one.

## 4. Spectra of the Lagrangian Fluctuation Fields of the Flow Velocity and the Rotor of Fluid

In the simplest case of incompressible fluid, where the evolution of transverse modes is described by the Navier—Stokes equation, the spectral function for the fluctuations of the velocity field $\psi_{\vec{v}}(\vec{q},\, \omega)$ has a form

$$\psi_{\vec{v}}(\vec{q}, \omega) = \frac{2k_{\mathrm{B}}T}{\rho}\frac{1}{-i\omega + \nu q^2}, \tag{11}$$

where $\nu$ is the kinematic coefficient of shear viscosity, $\rho$ is the density, $k_{\mathrm{B}}T$ is the energy of thermal motion. According to (4), if the Euler spectral function (11) is known, the corresponding spectrum of the Lagrangian CF for the fluctuation velocity field is determined by the expression:

$$\psi_{\vec{v}}^{\mathrm{L}}(\omega) = \frac{2k_{\mathrm{B}}T}{\pi\rho}\frac{8r_{\mathrm{L}}^2}{V_{\mathrm{L}}^2} \times$$

$$\times \int_0^\infty \frac{1}{q^2}\left(\cos qr_{\mathrm{L}} - \frac{\sin qr_{\mathrm{L}}}{qr_{\mathrm{L}}}\right)^2 \frac{1}{-i\omega + \nu q^2}\, dq, \tag{12}$$

so that the frequency dependence of the complex collective coefficient of translational diffusion is, by definition, equal to

$$D(\omega) = \frac{1}{3}\left(\int_0^\infty \psi_{\vec{v}}^{\mathrm{L}}(t)\, e^{i\omega t}\, dt\right) = \frac{1}{3}\,\psi_{\vec{v}}^{\mathrm{L}}(\omega). \tag{13}$$

Making use of the relationship between trigonometric and Bessel functions ($(\cos x - \frac{\sin x}{x})^2 = \frac{\pi}{2}\, x\, J^2_{3/2}(x)$) and integrating over $q$, we finally obtain $D$ in the form

$$D(\Omega) = \frac{3k_{\rm B}T}{2\pi^2\rho_0\nu r_{\rm L}}\ \frac{1}{-i\Omega}\ \left\{\frac{1}{3} - \frac{1}{2\sqrt{-i\Omega}}\ \times\right.$$

$$\left.\times\left[1+\frac{1}{i\Omega}+e^{-2\sqrt{-i\Omega}}\left(1+\frac{1}{\sqrt{-i\Omega}}\right)^2\right]\right\} \qquad (14)$$

with a dimensionless frequency introduced in terms of the radius of a Lagrangian particle and the kinematic viscosity $\Omega = \omega\frac{r_{\rm L}^2}{\nu}$.

The spectra of the Lagrangian CF for the fluctuation field of the fluid rotor or the rotational diffusion coefficient that determines the frequency dependence of the relaxation time can be obtained using the relationship between the Euler CFs for the fluid rotor field $\left\langle\vec{\omega}(\vec{r},t)\ \vec{\omega}(\vec{0},0)\right\rangle$ and the CFs for the transverse modes of the velocity field $\left\langle\vec{v}\,(\vec{r},\,t)\,\vec{v}(\vec{0},\,0)\right\rangle$ [9]:

$$\left\langle\vec{\omega}(\vec{r},t)\vec{\omega}(\vec{0},0)\right\rangle = -\frac{1}{4}\left\langle\Delta\vec{v}(\vec{r},\,t)\vec{v}(\vec{0},\,0)\right\rangle, \qquad (15)$$

where $\Delta$ is the Laplace operator. Allowing for (15) and passing to the Fourier-transform, we find the spectrum of the Lagrangian CF for the fluctuation field of the fluid rotor by analogy with (11)—(14) as

$$\psi^{\rm L}_{\vec{\omega}}(\omega) = -\frac{4k_{\rm B}T}{\pi\,\rho_0}\,\frac{r_{\rm L}^2}{V_{\rm L}^2}\times$$

$$\times\int\limits_0^\infty\left(\cos qr_{\rm L} - \frac{\sin qr_{\rm L}}{qr_{\rm L}}\right)^2\frac{1}{-i\omega+\nu q^2}\,dq. \qquad (16)$$

Hence, we finally derive

$$\psi^{\rm L}_{\vec{\omega}}(\Omega) = -\frac{9k_{\rm B}T}{16\pi^2\rho_0\nu r_{\rm L}^3}\,\frac{1}{(-i\Omega)^{3/2}}\times$$

$$\times\left[1+\frac{1}{i\Omega}+e^{-2\sqrt{-i\Omega}}\left(1+\frac{1}{\sqrt{-i\Omega}}\right)^2\right]. \qquad (17)$$

Thus, on the ground of expression (9), we obtain the frequency dependence of the relaxation coefficient as

$$L(\omega) = \frac{1}{\tau} + R(\omega) = \frac{1}{\tau} - \frac{9k_{\rm B}T}{8\pi^2\rho_0\nu r_{\rm L}^3}\ \frac{1}{(-i\Omega)^{3/2}}\times$$

$$\times\left[1+\frac{1}{i\Omega}+e^{-2\sqrt{-i\Omega}}\left(1+\frac{1}{\sqrt{-i\Omega}}\right)^2\right]. \qquad (18)$$

If it is necessary to investigate a more accurate hydrodynamic model, the spectra of the collective coefficients of translational diffusion and relaxation described by expressions (14), (18) can be refined by means of the replacement of the constant viscosity coefficient by a frequency-dependent one:

$$\nu(\omega) = \frac{\nu}{1-i\omega\tau_\nu}, \qquad (19)$$

where $\tau_\nu$ is the relaxation time for viscous stresses of fluid. Such a model allows for the fact that, in the range of Maxwellian times $\tau_{\rm M} \sim 2\cdot 10^{-13}$ s, a viscous response of the system to an external influence is changed to an elastic one, and the diffusion modes that determine the character of damping of the transverse modes in fluid become vibrational. Nevertheless, as will be shown below, the introduction of the relaxing viscosity coefficient does not essentially influence the character of the frequency dependence of the CF for the anisotropy tensor, which represents the ultimate aim of this investigation.

## 5. Spectrum of the Lagrangian CF for the Anisotropy Tensor

The spectrum of the Lagrangian CF for fluctuations of the anisotropy tensor is important for the interpretation of spectral experiments. It can be easily obtained from expressions (4),(10). Let's investigate the dimensionless relation of the spectra at an arbitrary and zero values of the frequency $\omega$:

$$I_\omega = \psi^{\rm L}_{I_m}(\omega)/\psi^{\rm L}_{I_m}(0) =$$

$$= A_0\,{\rm Re}\int\limits_0^\infty\frac{1}{q^2}\left(\cos qr_{\rm L} - \frac{\sin qr_{\rm L}}{qr_{\rm L}}\right)^2\times$$

$$\times\frac{1}{-i\omega+L(\omega)+q^2D(\omega)}\,dq, \qquad (20)$$

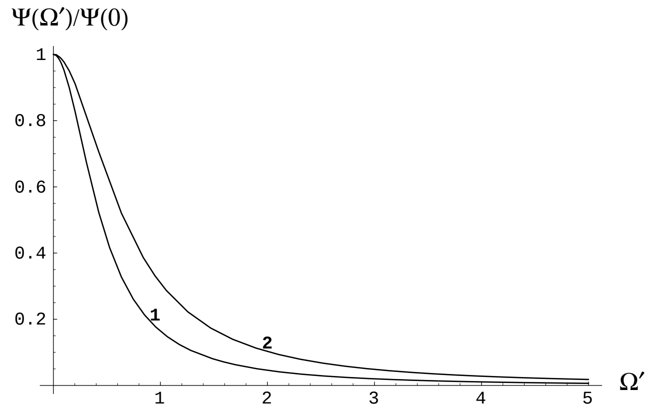
where $A_0^{-1}$ is equal to the integral on the right-hand side at $\omega = 0$.

Carrying out the substitution of the integration variable $x = qr_l$, one can represent (20) as

$$I_\omega = {\rm Re}\frac{1}{d_\omega}\ F(b_\omega)/F(b_0)\ , \qquad (21)$$

where

$$F(z) = \int\limits_0^\infty\left(\frac{\cos x}{x} - \frac{\sin x}{x^2}\right)^2\frac{dx}{x^2+z^2} \qquad (22)$$




Spectral density of the Lagrangian CF for fluctuations of the anisotropy tensor normalized to unity $\Psi(\Omega')/\Psi(0)$ (*1*), and the Lorentz curve (*2*) as functions of the dimensionless frequency. Curve *1* is obtained for the value of the dimensionless parameter $C = \frac{r_{\rm L}^2}{D(0)\tau} = 2,6 \cdot 10^4 \Omega'$.

$$b_\omega^2 = \frac{1}{d_\omega}\,\frac{r_L^2}{D(0)}\left[-i\omega + \frac{1}{\tau} + l_\omega\, R(0)\right],$$

$$b_0^2 = \frac{1/\tau + R(0)}{D(0)}\, r_{\rm L}^2, \tag{23}$$

$$d_\omega = \frac{D(\omega)}{D(0)} = \frac{F(c_\omega)}{F(0)}, \tag{24}$$

and

$$l_\omega = \frac{R(\omega)}{R(0)}, \quad R(\omega) = \int\limits_0^\infty \left(\cos x - \frac{\sin x}{x}\right)^2 \frac{dx}{x^2 + c_\omega^2} \tag{25}$$

are the dimensionless coefficients of translational and rotational diffusion, respectively; the dimensionless parameter $c_\omega^2 = -i\Omega$.

The explicit form of $d_\omega$ *and* $l_\omega$ is determined by the expressions (14) i (18), so that

$$F(c_\omega) = \frac{1}{c_\omega^2} \times$$

$$\times \left\{\frac{1}{3} - \frac{1}{2c_\omega}\left[1 - \frac{1}{c_\omega^2} + e^{-2c_\omega}\left(1 + \frac{1}{c_\omega}\right)^2\right]\right\}, \tag{26}$$

and

$$d_\omega = \frac{15}{2}\, F\left(c_\omega\right), \quad l_\omega = 3F(c_\omega) - \frac{1}{c_\omega^2}. \tag{27}$$

In order to find out which multiplier influences the character of the frequency behavior of $I_\omega$ most of all, let's consider the dependence of $b_\omega$ on the frequency in more details. In view of (23), it can be represented in the form

$$b_\omega^2 = \frac{1}{d_\omega}\left[-i\Omega' + \lambda\, l_\omega + \frac{r_{\rm L}^2}{D(0)\tau}\right], \tag{28}$$

where $\lambda = \frac{r_{\rm L}^2\, R(0)}{D(0)} = \frac{3}{4}$, and there appears the new parameter, the dimensionless frequency $-i\Omega' = -i\omega\, \frac{r_{\rm L}^2}{D(0)}$. One can easily correlate it with the function $c_\omega^2$:

$$c_\omega^2 = -i\Omega = -i\omega\ \frac{r_{\rm L}^2}{\nu} = -i\Omega'\ \frac{D(0)}{\nu}. \tag{29}$$

As $D(0) \sim 10^{-5}$ cm$^2$/s, $\nu \sim 10^{-2}$ cm$^2$/s and $\frac{D(0)}{\nu} \sim 10^{-3} \sim 1$, $c_\omega^2$ can be considered small as compared to the parameter $-i\Omega'$ of the anisotropy tensor. That's why, when expanding the dimensionless kinetic coefficients contained in (21) and (28) in terms of $c_\omega^2$, only first terms should be allowed for. Without giving the corresponding expressions, it's worth noting that, in the zero-order approximation according to (24), (25), $d_\omega$ and $l_\omega$ are equal to unity, while the following summands represent the terms of the first order of smallness with respect to $c_\omega^2$. Hence,

$$b_\omega^2 \approx -i\Omega' + \lambda + \frac{r_{\rm L}^2}{D(0)\,\tau} = -i\Omega' + {\rm const}, \tag{30}$$

and $F(b_\omega)$ represents the principal contribution to (21). It is also clear that, due to the smallness of $c_\omega^2$, the specific form [as for (19)] of the frequency dependence of the viscosity coefficient that determines the former will not essentially influence the character of the frequency dependence of the anisotropy tensor. The spectrum of the Lagrangian CF for fluctuations of the anisotropy tensor appears to be low-sensitive to the dispersion of the kinetic coefficients contained in its definition. The figure represents the behavior of the spectrum $I_\omega$ normalized to unity (curve *1*) depending on the dimensionless frequency $\Omega'$. In order to compare, curve *2* shows the dynamics of the Lorentz line.

The anisotropy tensor can be interpreted, for example, as a local inertia tensor of the volume element of fluid like it was done in [6–8]. In [8], its spectral intensity was investigated on the basis of the total system of hydrodynamic equations for anisotropic fluid [6]. The conversion of the Euler CFs to the Lagrangian ones was carried out using the approach proposed by I.Z. Fisher. In this case, a slight dependence of the spectrum on the dispersion of the kinetic coefficients was observed. Nevertheless, the form of its frequency dependence completely corresponds to that obtained in the present paper.

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## СПЕКТР КОРЕЛЯЦІЙНОЇ ФУНКЦІЇ ФЛУКТУАЦІЙ ТЕНЗОРА АНІЗОТРОПІЇ ЛАГРАНЖЕВОЇ ЧАСТИНКИ РІДИНИ

*О.І. Сахненко*

Р е з ю м е

Детально вивчено спектр кореляційної функції (КФ) теплових гідродинамічних флуктуацій тензора анізотропії лагранжевої частинки рідини. Ця КФ є асимптотичною оцінкою для молекулярної КФ орієнтаційних змінних, яка дуже важлива для дослідження теплового руху анізотропних молекул рідини. Ейлерівську КФ тензора анізотропії знайдено з рівняння Ланжевена з випадковими флуктуючими полями швидкості та його вихору. Відповідну лагранжеву КФ отримано на підставі підходу, запропонованого М.П. Маломужем.