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ON NON-EQUILIBRIUM STATISTICAL THEORY
OF SIMPLE FLUIDS: THE BASIC EQUATIONS

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До нерівноважної статистичної теорії простих рідин:
Основні рівняння.

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Анотація. На основі методу нерівноважного статистичного Зубарева для довільного набору динамічних змінних отримані загальні рівняння теорії лінійної релаксації простих рідин. Одержані результати проаналізовані для різних наборів динамічних змінних, а також отримані співвідношення між нижчими і вищими функціями пам'яті. Показано, що, фактично, рівняння лінійної релаксації є точними, якщо використовуються точні вирази для функцій пам'яті. Проведені порівняння із попередніми роботами.

**On non-equilibrium statistical theory of simple fluids:
The basic equations**

I.M.Mryglod, A.M.Hachkevych

Abstract. The general equations of linear relaxation theory of a fluid for arbitrary set of dynamic variables are derived on the base of Zubarev's nonequilibrium statistical operator method. The obtained results are analyzed for different sets of dynamic variables, and the relations between lower- and higher-order memory functions are derived. It is shown that the linear relaxation equations are in fact exact for arbitrary set of dynamic variables if the explicit expressions for memory functions are used. The comparison with the previous works is made.

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1. Introduction

In the past twenty years many successful studies have been made of the dynamical properties of classical fluids. Special attention has been paid to hard-sphere and Lennard-Jones fluids as the most simple models of realistic liquids. The various methods have been developed with the aim to investigate the time-correlation functions (TCFs), the collective mode spectrum and the generalized transport coefficients (see, for instance, [1–3]). The subject of special interest was the studies of lower-order TCFs, namely, the density-density, the density-energy and the energy-energy TCFs as well as the transverse momentum-momentum TCF, because these TCFs allow a unique determination of the generalized transport coefficients. The most studied of them is the density-density TCF which can be observed by scattering experiments. The all other TCFs noted above were considered mainly in connection with computer experiments.

Among the most used methods of theoretical investigation of the dynamical properties of simple fluids the following main ones may be noted: (i) the method of kinetic equations [2–4]; (ii) approaches based on the sum rules [5] or the linear response theory [7]; (iii) the formalism of memory functions including so-called method of k - and t -dependent memory functions (hydrodynamic description [1–3], different modifications of viscoelastic theory [1,6], the generalized mode approach [8–11], etc.); (iv) the mode-coupling theory of fluids [12]; (v) the methods of computer simulations [13,14]. The most part of these methods obtained also a wide application for studying of complex fluids such as molecular fluids with internal structure, mixtures, suspensions etc. However, as far as we can judge a lot of problems remain to be solved yet even in the case of simple liquids. Some of them can be formulated as follows: What is relation between various methods? What is the range of their applications? In which a way the memory functions should be modified for description of intermediate range of wave-vector k and frequency ω ? These questions could be supplemented with other ones related to the theoretical foundations of the non-equilibrium statistical theory of fluids. To recognize all these problems it is necessary to reconsider the theory of fluids from very beginning in such a way that the results would be presented in the enough general form for the subsequent analysis of the methods noted above (or at least some of them) on this basis. From the other side, the formulation of the theory have to be rather simple in order to be applied for calculations of the dynamic quantities such as generalized transport coefficients for instance. To develop of such viewpoint is the goal of our study. In this paper some general results of the

non-equilibrium statistical theory of simple fluids are presented. We used in our study the non-equilibrium statistical operator method in the form proposed by Zubarev [15,16]. Some findings (see, for instance [17–19]) of the next development of this method are also applied in our study.

The outline of this paper is as follows. In Sec.2 the general ideas of nonequilibrium statistical operator method are formulated, the solution of Liouville equation for arbitrary set of dynamic variables is found, and the generalized transport equations are derived. A weak nonequilibrium case is considered in Sec.3, and the equations of linear relaxation theory in matrix form are derived. The problem of the dynamic variables choice is discussed in Sec.4. Here too one of the most general case when the set of dynamic variables besides the hydrodynamic variables includes their derivatives up to order s is considered, and the recurrent relations for memory functions of fluids are derived. A discussion and concluding remarks are given in Sec.5.

2. Method of nonequilibrium statistical operator: General relations

2.1. Liouville equation with boundary conditions

The nonequilibrium state of a system is described by the nonequilibrium statistical operator $\rho(\mathbf{x}^N; t)$ which is a solution of the Liouville equation

$$\frac{\partial}{\partial t}\rho(\mathbf{x}^N; t) + i\hat{L}\rho(\mathbf{x}^N; t) = 0, \quad (2.1)$$

where $i\hat{L}$ is the Liouville operator, and $\mathbf{x}^N = \{\mathbf{r}, \mathbf{p}\}^N$. For a simple classical fluid with the Hamiltonian

$$H = \sum_{f=1}^N \frac{\mathbf{p}_f^2}{2m} + \frac{1}{2} \sum_{f \neq l} V(|\mathbf{r}_{fl}|), \quad (2.2)$$

the Liouville operator is

$$i\hat{L} = \sum_{f=1}^N \frac{\mathbf{p}_f}{m} \frac{\partial}{\partial \mathbf{r}_f} - \frac{1}{2} \sum_{f \neq l} \frac{\partial}{\partial \mathbf{r}_f} V(|\mathbf{r}_{fl}|) \left(\frac{\partial}{\partial \mathbf{p}_f} - \frac{\partial}{\partial \mathbf{p}_l} \right), \quad (2.3)$$

where $V(|\mathbf{r}_{fl}|)$ is a potential of interparticle interactions. As it usually is the nonequilibrium statistical operator $\rho(\mathbf{x}^N; t)$ is normalized to unity

$$\text{Sp } \rho(\mathbf{x}^N; t) = 1, \quad (2.4)$$

where in the case of classical treatment of a fluid

$$\text{Sp}(\dots) = \int \dots \int \frac{(d\mathbf{r}d\mathbf{p})^N}{N!(2\pi\hbar)^{3N}}(\dots).$$

In order to determine the nonequilibrium statistical operator $\rho(\mathbf{x}^N; t)$ from the Liouville equation (2.1) which is reversible with respect to time, one has to pose the boundary conditions corresponding to the physics of a system under consideration. In general case it can be done using two fundamental ideas of Bogolubov. First, this is conception of quasi-averages according to which one can introduce the infinitesimal source in the equation (2.1) which destroy its symmetry with respect of time inversion. The term with infinitesimal source should be tended to zero, the thermodynamic limit having been performed. In such a way one can find the retarded (or advanced) solution of the Liouville equation. Second, it is the Bogolubov's idea about the hierarchy of relaxation times which can be also presented as an abbreviate description hypothesis. From the physical point of view this means that a weak nonequilibrium state of a system can be described via evolution of the most slowly varying dynamic variables. With respect to the dynamic properties both of these physical principles were best used by Zubarev in his formulation of the nonequilibrium statistical operator method [15,16,20].

With Zubarev's method of nonequilibrium statistical operator, the retarded solutions of the Liouville equation (2.1) can be obtained from the Liouville equation with an infinitesimal source ($\epsilon \rightarrow +0$) which has the following form

$$\frac{\partial}{\partial t}\rho(\mathbf{x}^N; t) + i\hat{L}\rho(\mathbf{x}^N; t) = -\epsilon(\rho(\mathbf{x}^N; t) - \rho_q(\mathbf{x}^N; t)), \quad (2.5)$$

where $\rho_q(\mathbf{x}^N; t)$ is so-called quasi-equilibrium statistical operator.

According to the hypothesis of an abbreviated description, in order to find $\rho_q(\mathbf{x}^N; t)$ we restrict ourselves a priori to a certain set of the most slowly physical quantities $\{\hat{P}_\alpha\}$ which are thought to determine the nonequilibrium state. Of course in general case this is an approximation whose applicability depends on the choice of the dynamic variables and has to be judged afterwards. The problem of a choice of the dynamic variables is one of the most difficult in nonequilibrium theory and will be further discussed in more detail. Here, we assume that such variables are the extensive quantities $\{\hat{P}_\alpha\}$ and $\alpha = 1, 2, \dots, M$.

The quasi-equilibrium statistical operator $\rho_q(\mathbf{x}^N; t)$ can be found from the condition of the informational entropy extremum under the additional constraint that the mean values of the dynamic variables

$$\langle \hat{P}_\alpha \rangle_q^t = \text{Sp}\{\hat{P}_\alpha \rho_q(\mathbf{x}^N; t)\} \quad (2.6)$$

are fixed with preserving the normalization

$$\text{Sp} \rho_q(\mathbf{x}^N; t) = 1. \quad (2.7)$$

As a result, the quasi-equilibrium statistical operator $\rho_q(\mathbf{x}^N; t)$ can be written in Gibbs-like form

$$\rho_q(\mathbf{x}^N; t) = \exp\{-\Phi(t) - \sum_\alpha \hat{P}_\alpha F_\alpha(t)\} = \exp\{-\hat{S}(t)\}, \quad (2.8)$$

where

$$\Phi(t) = \ln \text{Sp} \exp\{-\sum_\alpha \hat{P}_\alpha F_\alpha(t)\} \quad (2.9)$$

is a corresponding quasi-equilibrium thermodynamic potential or, in other words, so-called Massieu-Planck functional. The operator $\hat{S}(t)$ is in fact the entropy operator, since its mean value $\langle \hat{S}(t) \rangle^t$ gives the nonequilibrium entropy of a system. We see in (2.8) and (2.9) that the dependence of $\rho_q(\mathbf{x}^N; t)$ on time is given only via the time-dependent quantities $\{F_\alpha(t)\}$ which are in fact the conjectured quantities to the set of dynamic variables $\{\hat{P}_\alpha\}$. The intensive quantities $\{F_\alpha(t)\}$ can be found from the conditions of self-consistency

$$\langle \hat{P}_\alpha \rangle_q^t = \langle \hat{P}_\alpha \rangle^t = \text{Sp} \{\hat{P}_\alpha \rho(\mathbf{x}^N; t)\}, \quad (2.10)$$

which follow immediately from the condition

$$\frac{\partial}{\partial t} \langle \mathcal{A} \rangle^t = \langle \frac{d\mathcal{A}}{dt} \rangle^t. \quad (2.11)$$

when $\mathcal{A} = \hat{P}_\alpha$. For arbitrary quantity \mathcal{A} , the equality (2.11) is satisfied in the sense of quasiaverages only when the limit $\epsilon \rightarrow 0$ is performed after the thermodynamic limit.

Since the dynamic variables have to describe the local properties of the system, they are depending on the space coordinate \mathbf{r} (or \mathbf{k} in the case of the corresponding Fourier transforms). Hence, in the above and following expressions an integration over space coordinates has to be made explicit together with a summation over index of dynamic variables α .

Introducing the deviation

$$\delta\rho(\mathbf{x}^N; t) = \rho(\mathbf{x}^N; t) - \rho_q(\mathbf{x}^N; t),$$

equation (2.5) can be rewritten in the form

$$\left\{ \frac{\partial}{\partial t} + i\hat{L} + \epsilon \right\} \delta\rho(\mathbf{x}^N; t) = -\left\{ \frac{\partial}{\partial t} + i\hat{L} \right\} \rho_q(\mathbf{x}^N; t). \quad (2.12)$$

Since the operator $\rho_q(\mathbf{x}^N; t)$ depends on time only via $F_\alpha(t)$ (or the conjectured quantities $\langle \hat{P}_\alpha \rangle^t$), one can introduce the projection operator $\mathcal{P}_q(t)$ according to the definition

$$\frac{\partial}{\partial t} \rho_q(\mathbf{x}^N; t) = -\mathcal{P}_q(t) i\hat{L} \rho_q(\mathbf{x}^N; t) \quad (2.13)$$

The operator $\mathcal{P}_q(t)$ is known as the Kawasaki-Gunton projection operator [21] and acts on the statistical operators or their combinations. For $\rho_q(\mathbf{x}^N; t)$ in the form (2.8), it has the following structure

$$\begin{aligned} \mathcal{P}_q(t)(\dots) = & \{ \rho_q(\mathbf{x}^N; t) - \sum_\alpha \frac{\delta \rho_q(t)}{\delta \langle \hat{P}_\alpha \rangle^t} \langle \hat{P}_\alpha \rangle^t \} \text{Sp}(\dots) + \\ & + \sum_\alpha \frac{\delta \rho_q(t)}{\delta \langle \hat{P}_\alpha \rangle^t} \text{Sp} \hat{P}_\alpha(\dots) \end{aligned} \quad (2.14)$$

and possesses the following properties

$$\mathcal{P}_q(t) \rho(t) = \rho_q(t), \quad \mathcal{P}_q(t) \rho_q(t) = \rho_q(t).$$

Finally, with the projector (2.14) the Liouville equation (2.12) takes the form

$$\left\{ \frac{\partial}{\partial t} + (1 - \mathcal{P}_q(t)) i\hat{L} + \epsilon \right\} \delta \rho(\mathbf{x}^N; t) = -(1 - \mathcal{P}_q(t)) i\hat{L} \rho_q(\mathbf{x}^N; t). \quad (2.15)$$

2.2. Nonequilibrium statistical operator

A formal solution of (2.15) for the nonequilibrium statistical operator $\rho(\mathbf{x}^N; t)$ can be written as follows

$$\begin{aligned} \rho(\mathbf{x}^N; t) = & \rho_q(\mathbf{x}^N; t) - \\ & - \int_{-\infty}^t e^{-\epsilon(t-t')} T(t, t') (1 - \mathcal{P}_q(t')) i\hat{L} \rho_q(\mathbf{x}^N; t') dt' \end{aligned} \quad (2.16)$$

where

$$T_q(t, t') = \exp_+ \left\{ - \int_{t'}^t d\tau (1 - \mathcal{P}_q(\tau)) i\hat{L} \right\} \quad (2.17)$$

is a generalized operator of time evolution with regard to projecting. Let us consider in (2.16) the action of \mathcal{P}_q and $i\hat{L}$ operators on the quasi-equilibrium statistical operator $\rho_q(\mathbf{x}^N; t)$. Taking into account the properties of the Liouville operator (2.3), the result of its action can be written

in the form

$$\begin{aligned} \mathcal{P}_q(t) i\hat{L} \rho_q(\mathbf{x}^N; t) = \\ = - \sum_\alpha F_\alpha(t) \int_0^1 d\tau [\rho_q(\mathbf{x}^N; t)]^\tau \mathcal{P}(t) \overset{\circ}{P}_\alpha [\rho_q(\mathbf{x}^N; t)]^{1-\tau}, \end{aligned} \quad (2.18)$$

where

$$\overset{\circ}{P}_\alpha \equiv i\hat{L} \hat{P}_\alpha. \quad (2.19)$$

The statistical operators are considered here as quantum ones for generality. For deriving of the equation (2.18) we used the equality

$$\begin{aligned} \mathcal{P}_q \int_0^1 d\tau (\rho_q(\mathbf{x}^N; t))^\tau \hat{\mathcal{X}}(\mathbf{x}^N) (\rho_q(\mathbf{x}^N; t))^{1-\tau} = \\ = \int_0^1 d\tau (\rho_q(\mathbf{x}^N; t))^\tau (\mathcal{P} \hat{\mathcal{X}}(\mathbf{x}^N)) (\rho_q(\mathbf{x}^N; t))^{1-\tau}, \end{aligned} \quad (2.20)$$

where $\hat{\mathcal{X}}(\mathbf{x}^N)$ is an arbitrary dynamic quantity depending on coordinates of phase space, and \mathcal{P} is the generalized Mori projection operator defined as follows

$$\mathcal{P}(t) \dots = \langle \dots \rangle_q^t + \sum_\alpha \frac{\delta \langle \dots \rangle_q^t}{\delta \langle \hat{P}_\alpha \rangle^t} \{ \hat{P}_\alpha - \langle \hat{P}_\alpha \rangle^t \} \quad (2.21)$$

with properties

$$\mathcal{P}(t) \mathcal{P}(t) = \mathcal{P}(t), \quad \mathcal{P}(t) (1 - \mathcal{P}(t)) = 0, \quad \mathcal{P}(t) \hat{P}_\alpha = \hat{P}_\alpha.$$

Contrary to the Kawasaki-Gunton projection operator, the Mori operator (2.21) acts only on the dynamic variables (or dynamic operators). Finally, taking into account (2.16) and (2.18), the nonequilibrium statistical operator can be written in the form

$$\begin{aligned} \rho(\mathbf{x}^N; t) = & \rho_q(\mathbf{x}^N; t) + \int_{-\infty}^t dt' e^{-\epsilon(t-t')} \sum_\alpha F_\alpha(t') \times \\ & \times \int_0^1 d\tau [\rho_q(\mathbf{x}^N; t')]^\tau T(t, t') \hat{I}_\alpha(t') [\rho_q(\mathbf{x}^N; t')]^{1-\tau}, \end{aligned} \quad (2.22)$$

where

$$\hat{I}_\alpha(t) = (1 - \mathcal{P}(t)) \overset{\circ}{P}_\alpha \quad (2.23)$$

are the generalized fluxes. The evolution operator $T(t, t')$ is now defined in terms of the Mori projection operator $\mathcal{P}(t)$, namely

$$T(t, t') = \exp_+ \left\{ - \int_{t'}^t d\tau (1 - \mathcal{P}(\tau)) i\hat{L} \right\}. \quad (2.24)$$

The nonequilibrium statistical operator (2.22) describes the nonequilibrium state of a liquid with the Hamiltonian (2.2) for the initial set of dynamic variables $\{\hat{P}_\alpha\}$. This operator is presented in the terms of the generalized dissipative fluxes (2.23) describing transport phenomena. In accordance with the hypothesis of an abbreviated description of nonequilibrium state, $\rho(\mathbf{x}^N; t)$ is a functional of the observed physical quantities varying in time (the mean values $\langle \hat{P}_\alpha \rangle^t$). Hence, using the solution (2.22) in order to obtain self-consistence description of nonequilibrium properties, one should derive equations for them, i.e. the generalized transport equations.

2.3. Nonlinear transport equations

To obtain transport equations for average values $\langle \hat{P}_\alpha \rangle^t$, the equalities

$$\frac{\partial}{\partial t} \langle \hat{P}_\alpha \rangle^t = \langle \overset{\circ}{P}_\alpha \rangle^t = \langle \overset{\circ}{P}_\alpha \rangle_q^t + \langle (1 - \mathcal{P}(t)) \overset{\circ}{P}_\alpha \rangle^t \quad (2.25)$$

may be used. The equalities (2.25) follow directly from the definition of the Mori operator definition (2.21). Performing the averaging in the right-hand side of (2.25) with the help of nonequilibrium statistical operator (2.22), the generalized transport equations can be found in the form

$$\frac{\partial}{\partial t} \langle \hat{P}_\alpha \rangle^t = \langle \overset{\circ}{P}_\alpha \rangle_q^t + \sum_{\beta} \int_{-\infty}^t dt' e^{-\epsilon(t-t')} \phi_{\alpha\beta}(t, t') F_\beta(t') dt', \quad (2.26)$$

where

$$\begin{aligned} \phi_{\alpha\beta}(t, t') &= \\ &= \int_0^1 d\tau \langle \hat{I}_\alpha(t), \rho_q^\tau(\mathbf{x}^N; t') T(t, t') \hat{I}_\beta(t') \rho_q^{-\tau}(\mathbf{x}^N; t') \rangle_q^t \end{aligned} \quad (2.27)$$

are so-called generalized memory functions of the system or, in other words, the generalized transport kernels.

Using the matrix notation the transport equations (2.26) can be rewritten as follows

$$\frac{\partial}{\partial t} \langle \hat{P} \rangle^t = \langle \overset{\circ}{P} \rangle_q^t + \int_{-\infty}^t dt' e^{-\epsilon(t-t')} \phi(t, t') (\hat{P}, \hat{P}^+) F(t') dt', \quad (2.28)$$

where $\phi = \|\phi_{\alpha\beta}\|$, and \hat{P} is a vector-column with elements $\{\hat{P}_\alpha\}$.

The transport equation system (2.26) for the chosen set of dynamic variables $\hat{P} = \{\hat{P}_\alpha\}$ corresponds of an abbreviated description of nonequilibrium behavior of a liquid and may be used to the study both strong and weak nonequilibrium states of the system. In general, this is a set of nonlinear equations. The intensive quantities $F_\alpha(t)$ entering in the quasi-equilibrium statistical operator $\rho_q(\mathbf{x}^N; t)$ depend on averages $\langle \hat{P}_\alpha \rangle^t$ through equations of self-consistency (2.8). The last ones are determined from the system (2.26). Besides that, as the generalized memory functions $\varphi_{\alpha\beta}(t; t')$ are unknown, the question about the solutions of the system (2.26) may be considered only under condition that the approximations for these functions should be based on analysis of the expression (2.27) and the corresponding equations for the higher-order memory functions. However, it is well-known the restriction to the linear case is a good approximation for transport phenomena in a fluid, and the nonlinear equations have to use only for special problems of nonequilibrium physics, for example for the description of dynamical behavior near phase transition, which are not a subject of our study.

For a weak non-equilibrium case, the transport equations (2.26) can be essentially simplified. Let us consider this case in more detail for deriving the linearized transport equations.

3. Weak nonequilibrium case

3.1. Linearized transport equations

The behavior of the system near the equilibrium may be described by set of the linear equations for deviations of macroscopic quantities $\langle \hat{P}_\alpha \rangle^t$ from the equilibrium values $\langle \hat{P}_\alpha \rangle_0 = \text{Sp} \{ \hat{P}_\alpha \rho_0(\mathbf{x}^N) \}$, where ρ_0 is the equilibrium statistical operator at temperature $1/\beta$. Assuming the deviations of intensive quantities $\delta F_n(t) = F_n(t) - F_n^0$ from their equilibrium values F_n^0 are small, the following expressions can be obtained

from (2.8)-(2.9):

$$\Phi(t) = \Phi_0 - \sum_{\alpha} \langle \hat{P}_{\alpha} \rangle_0 \delta F_{\alpha}(t), \quad (3.1)$$

$$\hat{S}(t) = \hat{S}_0 - \sum_{\alpha} \Delta \hat{P}_{\alpha} \delta F_{\alpha}(t), \quad (3.2)$$

where

$$\Phi_0 = \ln \text{Sp} \exp\{-\sum_{\alpha} \hat{P}_{\alpha} F_{\alpha}^0\}, \quad (3.3)$$

$$\hat{S}_0 = \Phi_0 + \sum_{\alpha} \hat{P}_{\alpha} F_{\alpha}^0, \quad (3.4)$$

and $\Delta \hat{P}_{\alpha} = \hat{P}_{\alpha} - \langle \hat{P}_{\alpha} \rangle_0$. The equilibrium statistical operator is

$$\rho_0 = \exp\{-\Phi_0 - \sum_{\alpha} \hat{P}_{\alpha} F_{\alpha}^0\} = \exp\{-\hat{S}_0\}. \quad (3.5)$$

From the definition of quasi-equilibrium statistical operator (2.8), in linear approximation we obtain

$$\rho_q(t) = \{1 - \sum_{\alpha} \int_0^1 d\tau \Delta \hat{P}_{\alpha}(\tau) \delta F_{\alpha}(t)\} \rho_0, \quad (3.6)$$

where

$$\Delta \hat{P}_{\alpha}(\tau) = \rho_0^{\tau} \Delta \hat{P}_{\alpha} \rho_0^{-\tau}. \quad (3.7)$$

Using the self-consistency conditions (2.10), the relationship between the deviations of intensive and extensive quantities can be found. In matrix form we have

$$\delta \langle \hat{P} \rangle^t = -(\Delta \hat{P}, \Delta \hat{P}^+) \delta F^+(t). \quad (3.8)$$

The static correlation function (A, B) in general case is defined as follows

$$(A, B) = \int_0^1 d\tau \langle A \rho_0^{\tau} B \rho_0^{-\tau} \rangle_0 = \int_0^1 d\tau \langle AB(t = i\hbar\beta\tau) \rangle_0. \quad (3.9)$$

In the case of classical treatment we obtain from (3.9) the usual definition of static correlation function in form $(A, B) = \langle AB \rangle_0$. As follows from (2.22), the linearized nonequilibrium statistical operator is

$$\begin{aligned} \delta \rho(t) = & \delta \rho_q(t) + \sum_{\alpha} \int_{-\infty}^t dt' e^{\epsilon(t'-t)} \delta F_{\alpha}(t') \times \\ & \times \int_0^1 d\tau \rho_0^{\tau} T_0(t-t') (1-\mathcal{P}) i \hat{L} \hat{P}_{\alpha} \rho_0^{1-\tau}. \end{aligned} \quad (3.10)$$

Using Fourier transformation for dependent on time functions

$$f(t) = \int d\omega \tilde{f}(\omega) \exp(i\omega t),$$

the expressions (3.6) and (3.10) can be rewritten in a matrix form as follows

$$\delta \tilde{\rho}_q(\omega) = - \int_0^1 d\tau \Delta \hat{P}^+(\tau) \delta \tilde{F}(\omega) \rho_0, \quad (3.11)$$

$$\delta \tilde{\rho}(\omega) = \delta \tilde{\rho}_q(\omega) +$$

$$+ \int_0^1 d\tau \rho_0^{\tau} \frac{1}{i\omega + \epsilon + (1-\mathcal{P}) i \hat{L}} (1-\mathcal{P}) \hat{P}^+ \rho_0^{1-\tau} \delta \tilde{F}(\omega), \quad (3.12)$$

where $\Delta \hat{P}^+$ is a transposed vector with the elements $\{\Delta \hat{P}_{\alpha}\}$ and $\delta \tilde{F}(\omega)$ is a vector-column with elements $\{\delta \tilde{F}_{\alpha}(\omega)\}$. The projection operator \mathcal{P} and the operator $T_0(t-t')$ are given by

$$\mathcal{P} \dots = (\dots, \Delta \hat{P}^+) (\Delta \hat{P}, \Delta \hat{P}^+)^{-1} \Delta \hat{P}, \quad (3.13)$$

$$T_0(t-t') = \exp\{-(t-t')(1-\mathcal{P}) i \hat{L}\}. \quad (3.14)$$

Using the linearized solution of the Liouville equation (3.12), it is easy to obtain the set of the linearized transport equations for the macroscopic quantities $\langle \Delta \hat{P} \rangle^t$

$$\{i\omega - i\Omega_0 + \tilde{\varphi}_{\epsilon}(\omega)\} \langle \Delta \hat{P} \rangle^{\omega} = 0 \quad (3.15)$$

where

$$i\Omega_0 = (\hat{P}, \Delta \hat{P}^+) (\Delta \hat{P}, \Delta \hat{P}^+)^{-1} \quad (3.16)$$

is a frequency matrix, and

$$\begin{aligned} \tilde{\varphi}_\epsilon(\omega) &\equiv \tilde{\varphi}(\epsilon + i\omega) = \\ &= ((1 - \mathcal{P}) \overset{\circ}{P}, \frac{1}{i\omega + \epsilon + (1 - \mathcal{P})i\hat{L}} (1 - \mathcal{P}) \overset{\circ}{P}^+) \times \\ &\times (\Delta\hat{P}, \Delta\hat{P}^+)^{-1} \end{aligned} \quad (3.17)$$

is a matrix of the memory functions. The matrix equation (3.15) is the set of the linearized transport equations or, in other words, the equations of the linear relaxation theory.

3.2. Equilibrium time correlation functions

It can be shown the equations for the equilibrium time correlation functions have the similar structure to (3.15). Really, a formal solution of the Liouville equation (2.12) can be also written in the form

$$\begin{aligned} \rho(\mathbf{x}^N; t) &= \rho_q(\mathbf{x}^N; t) - \\ &- \int_{-\infty}^t dt' e^{\epsilon(t'-t) - i\hat{L}(t-t')} \left\{ \frac{\partial}{\partial t'} + i\hat{L} \right\} \rho_q(\mathbf{x}^N; t'). \end{aligned} \quad (3.18)$$

For a weak nonequilibrium case, performing Fourier transformation for dependent on time functions, we obtain

$$\begin{aligned} \delta\tilde{\rho}(\omega) &= \delta\tilde{\rho}_q(\omega) + \\ &+ \int_0^1 d\tau \rho_0^\tau \frac{1}{i\omega + \epsilon + i\hat{L}} \{ \overset{\circ}{P}^+ + i\omega\Delta\hat{P}^+ \} \rho_0^{1-\tau} \delta\tilde{F}(\omega). \end{aligned} \quad (3.19)$$

From the equations of self-consistency (2.10)

$$\text{Sp} \{ \Delta\hat{P} [\delta\tilde{\rho}(\omega) - \delta\tilde{\rho}_q(\omega)] \} = 0,$$

using the solution in the form (3.19), one find

$$\begin{aligned} i\omega\delta\tilde{F}(\omega) &= - \frac{1}{(\Delta\hat{P}, \Delta\hat{P}^+)^z} (\Delta\hat{P}, \overset{\circ}{P}^+)^z \delta\tilde{F}(\omega) = \\ &= \left\{ - \frac{1}{(\Delta\hat{P}, \Delta\hat{P}^+)^z} (\Delta\hat{P}, \Delta\hat{P}^+) + z \right\} \delta\tilde{F}(\omega), \end{aligned} \quad (3.20)$$

where

$$(A, B^+)^z = \left(A, \frac{1}{z + iL} B^+ \right), \quad (3.21)$$

$A, B = \{ \hat{P}, \overset{\circ}{P} \}$ and $z = i\omega + \epsilon$, and $(A, B^+)^z$ is the matrix of Laplace transforms of the quantum-mechanical equilibrium time correlation functions $(A, B^+)(t)$ defined by

$$(A, B^+)(t) = \int_0^1 d\tau \langle A \rho_0^\tau \exp\{-i\hat{L}t\} B \rho_0^{-\tau} \rangle_0. \quad (3.22)$$

Comparing of (3.15) with (3.20), and using of (3.8), it is easy to obtain the matrix equation for Laplace transforms of the time correlation functions $(\Delta\hat{P}, \Delta\hat{P}^+)^z$

$$\{ z - i\Omega_0 + \tilde{\varphi}(z) \} (\Delta\hat{P}, \Delta\hat{P}^+)^z = (\Delta\hat{P}, \Delta\hat{P}^+). \quad (3.23)$$

Another result that follows immediately from such mathematical treatment and will be useful for subsequent calculations is the expression for the matrix of memory functions

$$\begin{aligned} \tilde{\varphi}(z) &= \left((\overset{\circ}{P}, \overset{\circ}{P}^+)^z - \right. \\ &\left. - (\overset{\circ}{P}, \Delta\hat{P}^+)^z \frac{1}{(\Delta\hat{P}, \Delta\hat{P}^+)^z} (\Delta\hat{P}, \overset{\circ}{P}^+)^z \right) \frac{1}{(\Delta\hat{P}, \Delta\hat{P}^+)^z}. \end{aligned} \quad (3.24)$$

It is important to note that as follows from the definition (3.21), the time correlation functions under consideration can be expressed in terms of the retarded correlation Green functions

$$G_{AB}^{(r)}(t - t') = -i\theta(t - t') \int_0^1 d\tau \langle A(t) \rho_0^\tau B(t') \rho_0^{-\tau} \rangle_0, \quad (3.25)$$

where $\theta(t) = 1$ or 0 according to whether $t > 0$ or $t < 0$. In the case of classical treatment the expression (3.25) gives the definition of classical correlation Green functions. The spectrum of collective modes can be determined from the equation

$$\text{Det} | z - i\Omega_0 + \tilde{\varphi}(z) | = 0, \quad (3.26)$$

which gives the poles of the regarded correlation Green functions constructed on the set of dynamic variables $\{\Delta\hat{P}_\alpha\}$. It should be also stressed that the matrix equation for the equilibrium time correlation functions (3.23) is in fact the exact equation. This statement can easily be proved using the expressions for the frequency matrix (3.16) and the matrix of memory functions (3.24).

The linearized transport equations (3.15), the equations for the equilibrium time correlation functions (3.23), and the equation for collective mode spectrum (3.26) will be used as the basis for the next our study of the dynamic behavior of simple fluids.

4. Extended set of dynamic variables

4.1. Introducing remarks

As it was noted before one of the most important problem of the nonequilibrium statistical theory of many-body systems is the correct choice of the dynamic (macroscopic) variables which have to be the most slowly ones. Frequently, the existence of slow variables can be traced back to conservation laws and, in the case of ordered systems, to continuous broken symmetries. The associated dynamic variables are then the densities of the conserved quantities as well as the order parameters. However the additional dynamic variables may be also included into the initial set $\{\hat{P}_\alpha\}$. Let us discuss this problem with some examples.

For the density-density time correlation function alone, many successful descriptions exist in the literature using the approximations for the memory functions defined on the one variable (the density of particles) [22], on the two variables (the densities of particles and momentum) [2,1], as well as on the three dynamic variables when the variable associated with the longitudinal component of the stress tensor is included in addition [1,23]. As the modification of such kind of theories the mode coupling theory of a fluid may be recalled. Mode-coupling approach was proposed by Kawasaki [24], and for a simple classical liquid it was later developed by Götze et.al. [12] In this approach the binary and higher-order combinations of the densities of particles and momentum are also considered. On the basis of the approximations for the memory functions in the form of two-mode decay integrals the closed equations for the time correlation functions have been derived. As it has been shown such approach is especially very useful for description of liquid–glass transition phenomena [25]. As will be seen later the methods noted above are in fact the results of the same theory if the explicit expressions for the corresponding memory functions are used. And the different results for the density-density time correlation function (or the dynamic structure factor) can be explained by the various approximations to have been used in the calculations. Moreover, such theory can not give the correct hydrodynamic equations in the limit $k \rightarrow 0$ because the fluctuations of the energy are neglected, and this means also that the generalized transport

coefficients can not be calculated correctly. Therefore, we may conclude that the all conserved variables should be considered from the very beginning if the consistent calculations of the time correlation functions as well as the generalized transport coefficients is the goal of the theory.

Nonequilibrium thermodynamics is based on the using of the conservation laws for the average values of physical quantities. Statistical thermodynamics of non-equilibrium processes proceeds from the conservation laws directly for the dynamic variables, i.e. it regards them from the microscopic point of view. There are three conserved variables for a single-component liquid, namely, the particle number density $\hat{n}(\mathbf{r})$, the momentum density $\hat{\mathbf{J}}(\mathbf{r})$, and the energy density $\hat{\varepsilon}(\mathbf{r})$. Their Fourier-transforms $\hat{n}_{\mathbf{k}}$, $\hat{\mathbf{J}}_{\mathbf{k}}$, $\hat{\varepsilon}_{\mathbf{k}}$ can be defined as follows

$$\hat{n}_{\mathbf{k}} = \frac{1}{\sqrt{N}} \sum_i e^{i\mathbf{k}\mathbf{r}_i}, \quad (4.1)$$

$$\hat{j}_{\mathbf{k}}^{L,T} = \frac{1}{\sqrt{N}} \sum_i p_i^{L,T} e^{i\mathbf{k}\mathbf{r}_i}, \quad (4.2)$$

$$\hat{\varepsilon}_{\mathbf{k}} = \frac{1}{\sqrt{N}} \sum_i \left(\frac{\mathbf{p}_i^2}{2m} + \frac{1}{2} \sum_{j(j \neq i)} V(|\mathbf{r}_{ij}|) \right) e^{i\mathbf{k}\mathbf{r}_i}. \quad (4.3)$$

In expression (4.2), as it usually is, the index L denotes longitudinal component, i.e. the component parallel to a vector of \mathbf{k} , which can be taken to be parallel to the Oz axis, and index T denotes transverse component, i.e. perpendicular to \mathbf{k} . In such case we can consider the variables (4.1)-(4.3) as functions of the wave-number k only.

It should be stressed that since the transverse component of the current density is independent of the other variables this variable may be treated separately from the longitudinal variables. From here on we consider the dynamic variables as classical ones, and this means that in our case $\langle A, B \rangle = \langle AB \rangle_0$.

For the longitudinal fluctuations the set of so-called orthogonal dynamic variables can be introduced instead of the variables (4.1), (4.2) and (4.3). These variables are connected with the initial ones by linear transformation and can be found with the help of Schmidt's orthogonalization procedure using the expression (3.9) as the definition of scalar product. Let us define the vector column $\hat{\mathcal{B}}^L(k)$ with the components $\hat{\mathcal{B}}_\alpha^L(k) = \{\hat{n}_k, \hat{J}_k^L, \hat{h}_k\}$, which are the orthogonal hydrodynamic variables, where

$$\hat{h}_k = \hat{\varepsilon}_k - \langle \hat{\varepsilon}_k \hat{n}_{-k} \rangle_0 \langle \hat{n}_k \hat{n}_{-k} \rangle_0^{-1} \hat{n}_k \quad (4.4)$$

is the density of generalized enthalpy. For the transverse fluctuations we have one hydrodynamic variable only, namely $\hat{\mathcal{B}}^T(k) = \hat{J}_k^L$. Now, using general results (3.15), (3.23) and (3.26), it is easy to derive the corresponding equations for $\hat{P} = \hat{\mathcal{B}}^{L,T}$ [27]. In this case the transport equations for $\langle \hat{\mathcal{B}}^{L,T} \rangle^t$ is known in the literature as the generalized hydrodynamic equations which give in the hydrodynamic limit the Navier-Stokes equations, but in general form they are much more complicated because of the k - and ω -dependent coefficients.

From comparison with results of the nonequilibrium thermodynamics the expressions for the generalized transport coefficients via the hydrodynamic memory functions can be found

$$\eta(k, z) + \frac{4}{3}\zeta(k, z) = \frac{mn}{k^2}\varphi_{JJ}^{H,L}(k, z), \quad (4.5)$$

$$\xi(k, z) = \frac{nC_V(k)T}{k^2}\varphi_{Jh}^H(k, z) = \frac{nm}{k^2}\varphi_{hJ}^H(k, z), \quad (4.6)$$

$$\lambda(k, z) = \frac{nC_V(k)}{k^2}\varphi_{hh}^H(k, z), \quad (4.7)$$

$$\zeta(k, z) = \frac{nm}{k^2}\varphi_{JJ}^{H,T}(k, z). \quad (4.8)$$

Here $\eta(k, z)$, $\zeta(k, z)$ and $\lambda(k, z)$ are generalized bulk and shear viscosities, and thermal conductivity coefficients, respectively. $\xi(k, z)$ is a generalized coefficient which describes the dynamical coupling between the stress and the heat current and vanishes in the limit $k \rightarrow 0$. The memory functions $\varphi_{\alpha,\beta}^H(k, z)$ are defined by (3.17) with $\hat{P} = \hat{\mathcal{B}}^{L,T}$.

From this point two main different ways in which the generalized hydrodynamic theory was developed for the description for intermediate range of k and ω may be separated. The first one may be called once again as the method of k - and ω -dependent memory functions in which the various approximations were used for the memory functions $\varphi_{\alpha,\beta}^H(k, z)$ (see, for instance, [2,3,1]). The main future of the second approach is an extension of the set of dynamic variables. As an example the microscopic viscoelastic theory of a liquid can be recalled. In early version of this theory [1] the flux of momentum was included into the initial set of dynamic variables, and the fluctuation of energy (4.3) was ignored. Later the variable of energy density $\varepsilon_{\mathbf{k}}$ was also considered in the scheme of the viscoelastic theory [6]. It is evident that the results of viscoelastic theory can be reproduced from (3.15), (3.23) and (3.26) if the corresponding variables are chosen. As the another example of such

kind of modifications the generalized mode approach can be noted [8–11]. In this approach using Markovian approximation for memory functions the general problem of dynamic theory reduced to the calculation of the generalized mode spectrum of a system, and the time correlation functions could be presented as a sum of partial terms related to the corresponding collective mode. In five-mode approximation the first results for Lennard-Jones liquid were obtained by de Schepper et. al. [9]. The seven-mode approximation for longitudinal fluctuations and the three-mode approximation for transverse fluctuations have been studied by Mryglod et. al. [10,11].

4.2. Relations for the memory functions

In order to compare between themselves the results of various approaches in which different sets of dynamic variables are used, let us consider more general case. We introduce the following notations

$$\begin{aligned} \hat{A}_0 &= \hat{\mathcal{B}}^{L,T}, \quad \hat{A}_1 = i\hat{L}\hat{\mathcal{B}}_0^{L,T}, \dots, \\ \hat{A}_l &= (i\hat{L})^l \hat{\mathcal{B}}_0^{L,T} = i\hat{L}\hat{A}_{l-1}, \dots, \quad \hat{A}_s = (i\hat{L})^s \hat{\mathcal{B}}_0^{L,T}, \end{aligned} \quad (4.9)$$

where $\hat{\mathcal{B}}^L = \{\hat{n}, \hat{\mathcal{B}}_0^L\}$, $\hat{\mathcal{B}}_0^L = \{\hat{J}^L, \hat{h}\}$ and $\hat{\mathcal{B}}^T = \hat{\mathcal{B}}_0^T = \hat{J}^T$. Contrary to Ref. [18] where the similar problem was considered we have a case that one from the variables, namely \hat{J}_k^L , has the special property of being both a flux and a conserved variable, so that $(i\hat{L})^l \hat{n} \sim (i\hat{L})^{l-1} \hat{J}^L$. The set of the dynamical variables $\{\hat{A}_0, \hat{A}_1, \dots, \hat{A}_s\}$ will be considered as initial one and includes $3 + 2s$ variables for the longitudinal fluctuations and $1 + s$ variables for the transverse fluctuations.

The orthogonalized dynamic variables constructed on (4.9) can be found with the help of the Schmidt's orthogonalization procedure with the definition of scalar product in the form (3.9). As a result we have

$$\begin{aligned} \hat{Y}_0 &= \hat{A}_0, \quad \hat{Y}_1 = (1 - \mathcal{P}_0)\hat{A}_1, \dots, \\ \hat{Y}_l &= (1 - \mathcal{P}_{l-1})i\hat{L}\hat{A}_{l-1} = (1 - \mathcal{P}_{l-1})\hat{A}_l, \dots, \\ \hat{Y}_s &= (1 - \mathcal{P}_{s-1})\hat{A}_s, \end{aligned} \quad (4.10)$$

where

$$\mathcal{P}_0 = \Delta\mathcal{P}_0 = \langle \dots \hat{A}_0^+ \rangle_0 \langle \hat{A}_0 \hat{A}_0^+ \rangle_0^{-1} \hat{A}_0, \quad (4.11)$$

$$\mathcal{P}_l = \sum_{m=0}^l \Delta\mathcal{P}_m, \quad (4.12)$$

$$\Delta\mathcal{P}_m = \langle \dots \hat{Y}_m^+ \rangle_0 \langle \hat{Y}_m \hat{Y}_m^+ \rangle_0^{-1} \hat{Y}_m, \quad (4.13)$$

are Mori-like projection operators. Now the variables $\{\hat{Y}_l\}$ possess the properties

$$\langle \hat{Y}_l^\alpha \hat{Y}_l^\beta \rangle_0 = \delta_{l\nu} \delta_{\alpha\beta} \langle \hat{Y}_l^\alpha \hat{Y}_l^\alpha \rangle_0, \quad (4.14)$$

where $\alpha, \beta = \{J, \varepsilon\}$ for the longitudinal fluctuations. It is easily seen that the $\langle \hat{Y}_l^\alpha \hat{Y}_l^\beta \rangle_0$ in (4.14) is proportional to $\delta_{\alpha\beta}$ because of the different symmetrical properties \hat{J}_k^L and $\hat{\varepsilon}_k$ under the time inversion.

Using the general expressions obtained above and the properties of Mori projection operators, the frequency matrix (3.16) and the matrix of memory functions (3.17) can be calculated with $\hat{P} = \{\hat{Y}_0, \hat{Y}_1, \dots, \hat{Y}_n\}$. For frequency matrix we have

$$i\Omega = \begin{pmatrix} i\Omega_0 & I & & & & \\ -\Gamma_0 & i\Omega_1 & I & & & 0 \\ & -\Gamma_1 & i\Omega_2 & I & & \\ & & \dots & \dots & \dots & \\ 0 & & & -\Gamma_{n-2} & i\Omega_{n-1} & I \\ & & & & -\Gamma_{n-1} & i\Omega_n \end{pmatrix}$$

where

$$i\Omega_l = i\Omega_{ll} = \langle iL\hat{Y}_l\hat{Y}_l^+ \rangle_0 \langle \hat{Y}_l\hat{Y}_l^+ \rangle_0^{-1}, \quad (4.15)$$

$$\begin{aligned} \Gamma_l &= i\Omega_{l+1,l} = \langle iL\hat{Y}_{l+1}\hat{Y}_l^+ \rangle_0 \langle \hat{Y}_l\hat{Y}_l^+ \rangle_0^{-1} = \\ &= -\langle \hat{Y}_{l+1}\hat{Y}_{l+1}^+ \rangle_0 \langle \hat{Y}_l\hat{Y}_l^+ \rangle_0^{-1}, \end{aligned} \quad (4.16)$$

$$i\Omega_{l-1,l} = \langle iL\hat{Y}_{l-1}\hat{Y}_l^+ \rangle_0 \langle \hat{Y}_l\hat{Y}_l^+ \rangle_0^{-1} = \langle \hat{Y}_l\hat{Y}_l^+ \rangle_0 \langle \hat{Y}_l\hat{Y}_l^+ \rangle_0^{-1} = I, \quad (4.17)$$

I is a unit matrix. All of other elements of the frequency matrix are equal zero. Moreover, as follows from the time reversal properties, for the longitudinal fluctuations $i\Omega_l$ and Γ_l are nondiagonal and diagonal matrices, respectively. For the transverse fluctuations we have $i\Omega_l = 0$.

From the definition of matrix of memory functions (3.17) and the equality

$$(1 - \mathcal{P}_s)i\hat{L}\hat{P} = (1 - \mathcal{P}_s) \begin{pmatrix} i\hat{L}\hat{Y}_0 \\ i\hat{L}\hat{Y}_1 \\ \dots \\ i\hat{L}\hat{Y}_s \end{pmatrix} = \begin{pmatrix} 0 \\ 0 \\ \dots \\ \hat{Y}_{s+1} \end{pmatrix}$$

it is seen that the matrix of memory functions has only one non-zero block, namely

$$\tilde{\varphi}^{(s)} = \begin{pmatrix} 0 & 0 & \dots & 0 & 0 \\ 0 & 0 & \dots & 0 & 0 \\ & & \dots & & \\ 0 & 0 & \dots & 0 & 0 \\ 0 & 0 & \dots & 0 & \tilde{\varphi}_s \end{pmatrix}$$

where

$$\tilde{\varphi}_s(z) = \langle \hat{Y}_{s+1} \frac{1}{z + (1 - \mathcal{P}_s)i\hat{L}} \hat{Y}_{s+1}^+ \rangle_0 \langle \hat{Y}_s \hat{Y}_s^+ \rangle_0^{-1}. \quad (4.18)$$

The set of equations for $\langle \hat{Y}_l \rangle^\omega$ follows from (2.28) when $\hat{P} = \{\hat{Y}_0, \hat{Y}_1, \dots, \hat{Y}_s\}$

$$\begin{aligned} (i\omega - i\Omega_0) \langle \Delta \hat{Y}_0 \rangle^\omega - \langle \Delta \hat{Y}_1 \rangle^\omega &= 0, \\ \Gamma_0 \langle \Delta \hat{Y}_0 \rangle^\omega + (i\omega - i\Omega_1) \langle \Delta \hat{Y}_1 \rangle^\omega - \langle \Delta \hat{Y}_2 \rangle^\omega &= 0, \\ &\dots\dots\dots \\ \Gamma_{s-2} \langle \Delta \hat{Y}_{s-2} \rangle^\omega + (i\omega - i\Omega_{s-1}) \langle \Delta \hat{Y}_{s-1} \rangle^\omega - \langle \Delta \hat{Y}_s \rangle^\omega &= 0, \\ \Gamma_{s-1} \langle \Delta \hat{Y}_{s-1} \rangle^\omega + (i\omega - i\Omega_s + \tilde{\varphi}_s(z)) \langle \Delta \hat{Y}_s \rangle^\omega &= 0. \end{aligned} \quad (4.19)$$

Solving the last equation with respect to $\langle \Delta \hat{Y}_s \rangle^\omega$ and excluding this variable from Eqs. (4.19) we obtain the transport equations for $\hat{P} = \{\hat{Y}_0, \hat{Y}_1, \dots, \hat{Y}_{s-1}\}$ with the memory functions $\tilde{\varphi}_{s-1}(z)$ in the form

$$\tilde{\varphi}_{s-1}(z) = \left(zI - i\Omega_s + \tilde{\varphi}_s(z) \right)^{-1} \Gamma_{s-1}. \quad (4.20)$$

The recurrent relations (4.20) give the connection between the lower- and higher-order memory functions and can be used for arbitrary choice of dynamic variables and any s . It is necessary to stress that the relation (4.20) is in fact exact. This statement could also be proved by the another way which was applied in Ref. [9] for the particular case $s = 1$, namely, using the known operator equality

$$(A + B)^{-1} = A^{-1} - A^{-1}B(A + B)^{-1}$$

in Eq. (4.18) with $A = z + (1 - \mathcal{P}_{s-1})i\hat{L}$ and $B = -\Delta\mathcal{P}_s i\hat{L}t$.

Let us consider some particular results which follows from (4.20). Using (4.20) the known result of Mori for dynamic structural factor in the form of an ordinary continued fraction [26] can be reproduced when

the set of the dynamic variables for longitudinal fluctuations includes the densities of particles and momentum as well as higher order their derivatives. Then, as it follows from (4.15), we have $i\Omega_l = 0$. Moreover, Γ_l and $\tilde{\varphi}_l(z)$ have not a matrix structure in this case and are usual functions, so that we have

$$\tilde{\varphi}_{s-1}(z) = \frac{\Gamma_{s-1}}{z + \tilde{\varphi}_s(z)}. \quad (4.21)$$

The similar result can be found in the case of transverse fluctuations.

For longitudinal fluctuations, if the density of energy as well as its derivatives are taken into account, $i\Omega_l$, Γ_l , and $\tilde{\varphi}_l(z)$ are 2×2 matrices. In this case the recurrent relation (4.20) for arbitrary s can be rewritten as follows

$$\tilde{\varphi}_{s-1}^{11}(z) = \frac{\Gamma_{s-1}^{11}}{z + \tilde{\varphi}_s^{11}(z) - \frac{(-i\Omega_s^{12} + \tilde{\varphi}_s^{12}(z))(-i\Omega_s^{21} + \tilde{\varphi}_s^{21}(z))}{z + \tilde{\varphi}_s^{22}(z)}}, \quad (4.22)$$

$$\tilde{\varphi}_{s-1}^{12}(z) = \frac{\Gamma_{s-1}^{22}}{-i\Omega_s^{21} + \tilde{\varphi}_s^{21}(z) - \frac{(z + \tilde{\varphi}_s^{11}(z))(z + \tilde{\varphi}_s^{22}(z))}{-i\Omega_s^{12} + \tilde{\varphi}_s^{12}(z)}}, \quad (4.23)$$

$$\tilde{\varphi}_{s-1}^{21}(z) = \frac{\Gamma_{s-1}^{11}}{-i\Omega_s^{12} + \tilde{\varphi}_s^{12}(z) - \frac{(z + \tilde{\varphi}_s^{11}(z))(z + \tilde{\varphi}_s^{22}(z))}{-i\Omega_s^{21} + \tilde{\varphi}_s^{21}(z)}}, \quad (4.24)$$

$$\tilde{\varphi}_{s-1}^{22}(z) = \frac{\Gamma_{s-1}^{22}}{z + \tilde{\varphi}_s^{22}(z) - \frac{(-i\Omega_s^{21} + \tilde{\varphi}_s^{21}(z))(-i\Omega_s^{12} + \tilde{\varphi}_s^{12}(z))}{z + \tilde{\varphi}_s^{11}(z)}}, \quad (4.25)$$

and the known relations [9,27] for the memory functions of three- and five-mode descriptions ($s = 1$) follow immediately from (4.22)-(4.25).

Using the equations for time correlation functions (3.23) and the expressions (4.22)-(4.25), the time correlation functions of the hydrodynamic variables can be written in the form of the "generalized continued fraction". As it was pointed out in Ref.[17] such expressions for the time correlation functions correctly conveys the analytic structure of the corresponding Green functions in the complex z -plane.

5. Concluding remarks

Zubarev's method of the nonequilibrium statistical operator has been applied to derive the general equations of the linear relaxation theory of a fluid for the set of dynamic variables which includes besides the hydrodynamic macrovariables their higher order derivatives. The recurrent relations between the lower- and higher-order memory functions are found. In fact these relations via Eqs. (4.5)-(4.5) give the connections between the generalized transport coefficients and the higher-order memory functions. The recurrent formulas (4.20) are exact when the explicit expressions (3.17) (or (3.17)) for the memory functions are used. Therefore, we may consider the set of the equations (3.15), (3.23) and (3.26) for the hydrodynamic macrovariables, which are complemented by the recurrent relations (4.20), as the basic equations of the generalized hydrodynamics of simple fluids. These equations describe correctly the hydrodynamic limit, make possible to calculate the generalized transport coefficients, and can be used in the intermediate range of wavenumbers k and frequencies ω .

It is also shown that the results of many known descriptions follow immediately from the basic equations when the dynamic variables and an approximation for the memory functions are specified. We will discuss this point in more detail in the next paper of a series. In general, it is important to stress especially, the choice of an approximation for the memory functions should be considered in combinations with the recurrent relations (4.20). In such a manner only the consistent description of a fluid can be obtained. From the physical point of view the choice of an approximation is closely connected to some dynamical model of a fluid to be considered and determines the range in which this model can be used (so-called "window conditions").

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