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ON NON-EQUILIBRIUM STATISTICAL THEORY OF FLUIDS: LINEAR RELAXATION THEORIES WITH DIFFERENT SETS OF DYNAMIC VARIABLES

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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 found. 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.

1. Introduction

In the past twenty years many 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 for 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 fluids the following ones should be noted: (i) approach of kinetic equations [4]; (ii) methods based on sum rules [5] or 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], generalized mode approach [8–11], etc.); (iv) mode-coupling theory [12]; (v) computer simulations [13,14]. The most part of these methods obtained 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 many questions even in the case of simple liquids remain to be

solved. Some of them can be formulated as follows: What is relation between various methods? What is the range of their applications? In which way the memory functions should be modified for description of intermediate range of wave-vector k and frequency ω? and so on. To recognize all these problems it is necessary to form the non-equilibrium statistical theory of a fluid from very beginning in such a way that this theory have to be enough general for subsequent consideration of the methods noted above, and rather simple to be studied in detail. the goal of our study is to develop such a view-point. In particular, the problem of choice of dynamic variables and the linear relaxation theories for them is fully considered here.

The outline of this paper is as follows. In Sec.2 the general ideas of nonequilibrium statistical operator method are formulated, and the solution of Liouville equation for arbitrary set of dynamic variables is found. A weak nonequilibrium case is considered in Sec.3, and the equations of linear relaxation theory in matrix form are derived. In the special cases the known results corresponding to so-called hydrodynamic models are obtained in Sec.4. The problem of choice of dynamic variables is discussed in Sec.5. One of the most general case when the set of dynamic variables besides the hydrodynamic variables includes their derivatives up to order s is also considered here. The recurrent relations for memory functions are found. A discussion and concluding remarks are given in Sec.6.

2. Method of nonequilibrium statistical operator. General description

2.1. Liouville equation with boundary condition

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

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

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

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

the Liouville operator is

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

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

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

where in the case of classical treatment of a fluid

$$\mathrm{Sp}\ (\ldots)=\int\ldots\int\frac{(d\mathbf{r}d\mathbf{p})^N}{N!(2\pi h)^{3N}}\ (\ldots).$$

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. It is possible to do this using two fundamental ideas of Bogolubov. First one is the conception of quasi-averages according to which one can introduce the infinitesimal source in the equation (2.1) which destroys its symmetry with respect of time inversion. The term with infinitesimal source has to 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 one 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 formulation of the nonequilibrium statistical operator method [15,16].

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 ($\epsilon \to +0$) which has the following form

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

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

According to general feature of the construction of $\rho_q(\mathbf{x}^N;t)$ 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 selection of the dynamic variables is one of the most difficult in nonequilibrium theory and will be further discussed in detail. Here we assume that such variables are the extensive quantities $\{\hat{P}_\alpha\}$, $\alpha = 1, 2, \ldots, M$.

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

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

are fixed with preserving the normalization

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

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

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

where

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

is a corresponding quasi-equilibrium thermodynamic potential or, in other words, so-called Massieu-Planck functional. 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 thermodynamically conjugated 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

$$\langle \hat{P}_\alpha \rangle_q^t = \langle \hat{P}_\alpha \rangle^t = \mathrm{Sp}\ \{\hat{P}_\alpha \rho(\mathbf{x}^N;t)\}. \tag{2.10}$$

Since the dynamic variables have to describe the local properties of the system they depend 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 explicitly 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

$$\{\frac{\partial}{\partial t} + i\hat{L} + \epsilon\}\delta\rho(\mathbf{x}^N;t) = -\{\frac{\partial}{\partial t} + i\hat{L}\}\ \rho_q(\mathbf{x}^N;t). \tag{2.11}$$

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

$$\frac{\partial}{\partial t}\rho_q(\mathbf{x}^N;t) = -\mathcal{P}_q(t)\ i\hat{L}\rho(\mathbf{x}^N;t) \tag{2.12}$$

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

$$\mathcal{P}_q(t)(\ldots) = \{\rho_q(\mathbf{x}^N;t) - \sum_\alpha \frac{\delta\rho_q(t)}{\delta\langle \hat{P}_\alpha \rangle^t}\langle \hat{P}_\alpha \rangle^t\}\ \mathrm{Sp}\ (\ldots) + \\ + \sum_\alpha \frac{\delta\rho_q(t)}{\delta\langle \hat{P}_\alpha \rangle^t}\ \mathrm{Sp}\ \hat{P}_\alpha(\ldots) \tag{2.13}$$

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 (2.13) the Liouville equation (2.11) takes the form

$$\{\frac{\partial}{\partial t} + (1-\mathcal{P}_q(t))i\hat{L} + \epsilon\}\ \delta\rho(\mathbf{x}^N;t) = -(1-\mathcal{P}_q(t))i\hat{L}\rho_q(\mathbf{x}^N;t). \tag{2.14}$$

2.2. Nonequilibrium statistical operator

A formal solution of (2.14) for the nonequilibrium statistical operator $\rho(\mathbf{x}^N;t)$ can be written as follows

$$\rho(\mathbf{x}^N;t) = \rho_q(\mathbf{x}^N;t) - \\ - \int_{-\infty}^{t} e^{-\epsilon(t-t')} T(t,t')(1-\mathcal{P}_q(t'))i\hat{L}\rho_q(\mathbf{x}^N;t')dt' , \tag{2.15}$$

where

$$T_q(t,t') = exp_+\{-\int_{t'}^{t} d\tau(1-\mathcal{P}_q(\tau))i\hat{L}\} \tag{2.16}$$

is a generalized operator of time evolution with regard to projecting. Let us consider in (2.15) 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

$$\mathcal{P}_q(t)i\hat{L}\rho_q(\mathbf{x}^N;t) = \\ = -\sum_\alpha F_\alpha(t)\int_0^1 d\tau\ [\rho_q(\mathbf{x}^N;t)]^\tau \mathcal{P}(t)\ \overset{\circ}{P}_\alpha\ [\rho_q(\mathbf{x}^N;t)]^{1-\tau}, \tag{2.17}$$

where

$$\overset{\circ}{P}_\alpha \equiv i\hat{L}\hat{P}_\alpha. \tag{2.18}$$

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

$$\mathcal{P}_q \int_0^1 d\tau(\rho_q(\mathbf{x}^N;t))^\tau\ \hat{\mathcal{X}}(\mathbf{x}^N)\ (\rho_q(\mathbf{x}^N;t))^{1-\tau} = \\ = \int_0^1 d\tau(\rho_q(\mathbf{x}^N;t))^\tau\ (\mathcal{P}\hat{\mathcal{X}}(\mathbf{x}^N))\ (\rho_q(\mathbf{x}^N;t))^{1-\tau}, \tag{2.19}$$

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

$$\mathcal{P}(t)... = \langle ...\rangle_q^t + \sum_\alpha \frac{\delta\langle ...\rangle_q^t}{\delta\langle\hat{P}_\alpha\rangle^t}\{\hat{P}_\alpha - \langle\hat{P}_\alpha\rangle^t\} \tag{2.20}$$

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 (2.20) acts only on the dynamic variables (or dynamic operators). Finally,

taking into account (2.15) and (2.17), the nonequilibrium statistical operator can be written in the form

$$\rho(\mathbf{x}^N;t) = \rho_q(\mathbf{x}^N;t) + \int_{-\infty}^{t} dt' e^{-\epsilon(t-t')} \sum_\alpha F_\alpha(t') \times$$

$$\times \int_0^1 d\tau\ [\rho_q(\mathbf{x}^N;t')]^\tau\ T(t,t')\hat{I}_\alpha(t')[\rho_q(\mathbf{x}^N;t')]^{1-\tau}, \tag{2.21}$$

where

$$\hat{I}_\alpha(t) = (1-\mathcal{P}(t))\ \overset{\circ}{P}_\alpha \tag{2.22}$$

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

$$T(t,t') = exp_+\{-\int_{t'}^{t} d\tau(1-\mathcal{P}(\tau))\ i\hat{L}\}. \tag{2.23}$$

The nonequilibrium statistical operator (2.21) 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 (2.22) 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 (2.21) in order to obtain self-consistent description of nonequilibrium properties, one should derive equations for them, i.e. the transport equations for a liquid.

2.3. Nonlinear transport equations

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

$$\frac{\partial}{\partial t}\langle\hat{P}_\alpha\rangle^t = \langle\overset{\circ}{P}_\alpha\rangle^t = \langle\overset{\circ}{P}_\alpha\rangle_q^t + \langle(1-\mathcal{P}(t))\ \overset{\circ}{P}_\alpha\rangle^t \tag{2.24}$$

can be used. The equalities (2.24) follow directly from the Mori operator definition (2.20). Performing the averaging in the right-hand part of (2.24) with the help of nonequilibrium statistical operator (2.21), the generalized transport equations for the system under consideration can be derived in the form

$$\frac{\partial}{\partial t}\langle\hat{P}_\alpha\rangle^t = \langle\overset{\circ}{P}_\alpha\rangle_q^t + \sum_\beta \int_{-\infty}^{t} dt' e^{-\epsilon(t-t')}(\hat{I}_\alpha(t),\ T(t,t')\hat{I}_\beta(t'))_q F_\beta(t')dt', \tag{2.25}$$

where

$$(\hat{I}_\alpha(t),\ T(t,t')\hat{I}_\beta(t'))_q =$$

$$= \int_0^1 d\tau\ \langle\hat{I}_\alpha(t),\ (\rho_q(\mathbf{x}^N;t'))^\tau\ T(t,t')\hat{I}_\beta(t')(\rho_q(\mathbf{x}^N;t'))^{-\tau}\rangle_q^t. \tag{2.26}$$

Let us use the matrix notation and introduce $\hat{P}$ as a vector-column with elements $\{\hat{P}_\alpha\}$. Then, for the matrix of static correlation functions $(\hat{P}, \hat{P}^+)$, where $\hat{P}^+$ is a transposed vector, the inverse matrix $(\hat{P}, \hat{P}^+)^{-1}$ can be defined

$$(\hat{P}, \hat{P}^+)^{-1}(\hat{P}, \hat{P}^+) = I, \tag{2.27}$$

where I is a unity matrix. Using (2.27), the transport equations (2.25) can be rewritten in the matrix form

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

where $\varphi = ||\varphi_{\alpha\beta}||$ and

$$\varphi_{\alpha\gamma}(t,t') = \sum_\delta (\hat{I}_\alpha(t),\ T(t,t')\hat{I}_\delta(t'))_q\ (\hat{P}, \hat{P}^+)^{-1}_{\delta\gamma} \tag{2.29}$$

are so-called generalized memory functions of the system or, in other words, the generalized transport kernels. As we see in (2.29), $\varphi_{\alpha\beta}(t,t')$ is defined in terms of quasi-equilibrium statistical operator $\rho_q(t)$. It is important to note that one can express the generalized transport coefficients of the system via the memory functions (2.29).

The transport equation system (2.25) for the chosen set of dynamic variables corresponds of an abbreviated description of nonequilibrium behavior of a liquid and may be utilized 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 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 (2.25). Besides that, as the generalized memory functions $\varphi_{\alpha\beta}(t;t')$ are unknown, the question about the solutions of the system (2.25) may be considered only under condition that the approximations for these functions should be based on analysis of the expression (2.29) and the corresponding equations for the higher-order memory functions. However, it is well-known that restriction to the linear case is a good approximation for transport phenomena in a fluid, and the nonlinear equations are to be used only for special problems of nonequilibrium physics, for example for the description of dynamical behavior near phase transition, which are not a subject of this paper.

For a weak non-equilibrium case, the transport equations (2.25) can be essentially simplified. Let us consider this case in more detail for deriving the linearized transport equations.

3. Weak nonequilibrium case

3.1. Generalized hydrodynamic 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 $\langle \hat{P}_\alpha\rangle_0 = \mathrm{Sp}\ \hat{P}_\alpha\rho_0(\mathbf{x}^N)$, 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 to be small, the following expressions can be obtained from (2.8)-(2.9):

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

$$S(t) = S_0 - \sum_\alpha \Delta \hat{P}_\alpha \, \delta F_\alpha(t), \tag{3.2}$$

where

$$\Phi_0 = \ln \, \mathrm{Sp} \, \exp\{-\sum_\alpha \hat{P}_\alpha \, F_\alpha^0\}, \tag{3.3}$$

$$S_0 = \Phi_0 + \sum_\alpha \hat{P}_\alpha \, F_\alpha^0, \tag{3.4}$$

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

$$\rho_0 = \exp\{-\Phi_0 - \sum_\alpha \hat{P}_\alpha \, F_\alpha^0\} = \exp\{-S_0\}. \tag{3.5}$$

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

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

where

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

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

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

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

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

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 (2.21), the linearized nonequilibrium statistical operator is

$$\delta \rho(t) = \delta \rho_q(t) + \sum_\alpha \int_{-\infty}^t dt' e^{\epsilon(t'-t)} \delta F_\alpha(t') \times$$

$$\times \int_0^1 d\tau \, \rho_0^\tau \, T_0(t - t') \, (1 - \mathcal{P}) i \hat{L} \hat{P}_\alpha \, \rho_0^{1-\tau}. \tag{3.10}$$

Using Fourier transformation for time dependent 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

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

$$\delta\tilde{\rho}(\omega) = \delta\tilde{\rho}_q(\omega) + \int_0^1 d\tau \; \rho_0^\tau \frac{1}{i\omega + \epsilon + (1-\mathcal{P})i\hat{L}}(1-\mathcal{P}) \; \overset{\circ}{P}{}^+ \; \rho_0^{1-\tau}\delta\tilde{F}(\omega), \tag{3.12}$$

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 linearized projection operator has the following structure

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

and, respectively, the evolution operator is

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

Using the linearized solution of the Liouville equation (3.12), it is easy to obtain the set of the generalized hydrodynamic equations or, in the other words, the linearized transport equations for the macroscopic quantities $\langle\Delta\hat{P}\rangle^t$. From the definition

$$\frac{\partial}{\partial t}\langle\Delta\hat{P}\rangle^t = \langle i\hat{L}\hat{P}\rangle^t,$$

we have

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

where

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

is a frequency matrix, and

$$\tilde{\varphi}_\epsilon(\omega) = ((1-\mathcal{P})\,\overset{\circ}{P}, \frac{1}{i\omega + \epsilon + (1-\mathcal{P})i\hat{L}}(1-\mathcal{P})\,\overset{\circ}{P}{}^+)\,(\Delta\hat{P}, \Delta\hat{P}^+)^{-1} \tag{3.17}$$

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

3.2. Equilibrium time correlation functions

It can be shown the equations for the equilibrium time correlation functions have the structure similar to (3.15). Really, a formal solution of the Liouville equation (2.11) can be also written in the form

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

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

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

From the equations of self-consistency (2.10)

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

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

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

where

$$(A,B^{+})^z = (A,\frac{1}{z+i\hat{L}}B^{+}) \quad (3.21)$$

is the matrix of Laplace transforms of the equilibrium time correlation functions, $A, B = \{\hat{P},\overset{\circ}{P}\ \}$ and $z = i\omega + \epsilon$.

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 $(\Delta\hat{P},\Delta\hat{P}^{+})^z$. We can find

$$\{\ z - i\Omega_0 + \tilde{\varphi}(z)\}(\Delta\hat{P},\Delta\hat{P}^{+})^z = (\Delta\hat{P},\Delta\hat{P}^{+}). \quad (3.22)$$

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

$$\tilde{\varphi}(z) = \Big((\overset{\circ}{P}\ ,\overset{\circ}{P}{}^{+})^z -$$
$$-(\overset{\circ}{P}\ ,\Delta\hat{P}^{+})^z\frac{1}{(\Delta\hat{P},\Delta\hat{P}^{+})^z}(\Delta\hat{P},\overset{\circ}{P}{}^{+})^z\Big)\frac{1}{(\Delta\hat{P},\Delta\hat{P}^{+})}. \quad (3.23)$$

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

$$G_{AB}^{(r)}(t-t') = -i\theta(t-t')\int_0^1 d\tau\ \langle A(t)\rho_0^{\tau}B(t')\rho_0^{-\tau}\rangle_0, \quad (3.24)$$

where $\theta(t) = 1$ or 0 according to whether $t > 0$ or $t < 0$ correspondingly. In the case of classical treatment the expression (3.24) gives the definition of classical correlation Green functions. The spectrum of collective modes can be determined from the equation

$$\mathrm{Det} \mid z - i\Omega_0 + \tilde{\phi}(z) \mid = 0, \tag{3.25}$$

which gives the poles of the retarded 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 (3.22) is in fact the exact equation. This is cosily shown using the expressions for the frequency matrix (3.16) and the matrix of memory functions in form (3.23).

As the result we have obtained a complete system of the generalized hydrodynamic equations that can be used for the subsequent investigation of dynamical behavior of the system. For the calculations of the time correlation functions, the generalized transport coefficients connected with the memory functions, and the collective modes spectrum, one has to find elements of the frequency matrix $i\Omega_0$ and the matrix of memory functions $\tilde{\varphi}(z)$.

4. Hydrodynamic models

Phenomenologic thermodynamics of non-circulating processes 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

$$\hat{n}(\mathbf{r}) = \sum_i \delta(\mathbf{r} - \mathbf{r}_i), \tag{4.1}$$

the momentum density

$$\hat{\mathbf{J}}(\mathbf{r}) = \sum_i \mathbf{p}_i \delta(\mathbf{r} - \mathbf{r}_i), \tag{4.2}$$

and the energy density

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

Here $\mathbf{r}_i$ and $\mathbf{p}_i$ are a position and momentum of i-th particle respectively.

For practical application it can be more convenient to use the collective variables which are Fourier-transforms of $\hat{n}(\mathbf{r})$, $\hat{\mathbf{J}}(\mathbf{r})$, and $\hat{\varepsilon}(\mathbf{r})$. The conservation equations relate their first time derivatives to the dissipative fluxes. We list below microscopic definition of $\hat{n}_\mathbf{k}$, $\hat{\mathbf{J}}_\mathbf{k}$, $\hat{\varepsilon}_\mathbf{k}$ and their associated fluxes. It should be stressed that since the transverse component of current density is independent of the other variables they may be treated separately.

The particle number density is given by

$$\hat{n}_\mathbf{k} = \frac{1}{\sqrt{N}} \sum_i e^{i\mathbf{k}\mathbf{r}_i}, \tag{4.4}$$

with

$$\overset{\circ}{n}_{\mathbf{k}} - \frac{ik}{m}\hat{j}^L_{\mathbf{k}} = 0. \tag{4.5}$$

Here index L denotes longitudinal component, i.e. the component parallel to a vector of $\mathbf{k}$, which is taken to be parallel to z axis, and

$$\hat{j}^{L,T}_{\mathbf{k}} = \frac{1}{\sqrt{N}}\sum_i p_i^{L,T} e^{i\mathbf{k}\mathbf{r}_i}, \tag{4.6}$$

where index T denotes transverse component (perpendicular to $\mathbf{k}$).

The momentum density $\hat{J}^{L,T}_{\mathbf{k}}$ satisfies the equation

$$\overset{\circ}{J}{}^{L,T}_{\mathbf{k}} + ik\sigma^{L,T}_{\mathbf{k}} = 0, \tag{4.7}$$

where $\sigma_{\mathbf{k}}$ is a stress tensor,

$$\sigma^{\alpha\beta}_{\mathbf{k}} = \frac{1}{\sqrt{N}}\sum_i \left(\frac{p_i^\alpha p_i^\beta}{m} - \frac{1}{2}\sum_{j(j\neq i)} \frac{r_{ij}^\alpha r_{ij}^\beta}{r_{ij}^2} P_{\mathbf{k}}(r_{ij})\right) e^{i\mathbf{k}\mathbf{r}_i} \tag{4.8}$$

with

$$P_{\mathbf{k}}(r) = r\frac{\partial V(r)}{\partial r}\frac{1-e^{-i\mathbf{k}\mathbf{r}}}{i\mathbf{k}\mathbf{r}} \tag{4.9}$$

and $\mathbf{r}_{ij} = \mathbf{r}_i - \mathbf{r}_j$. Here $\sigma^L_{\mathbf{k}}$ is the diagonal component, $\sigma^{zz}_{\mathbf{k}}$, while $\sigma^T_{\mathbf{k}}$ represents off-diagonal elements, for example $\sigma^{zx}_{\mathbf{k}}$. Note that $\hat{J}^L_{\mathbf{k}}$ has the special property of being both a flux and a conserved variable.

The energy density is defined by

$$\hat{\varepsilon}_{\mathbf{k}} = \frac{1}{\sqrt{N}}\sum_i \Big(\frac{\mathbf{p}_i^2}{2m} + \frac{1}{2}\sum_{j(j\neq i)} V(|\mathbf{r}_{ij}|)\Big) e^{i\mathbf{k}\mathbf{r}_i} \tag{4.10}$$

with its associated conservation equation

$$\overset{\circ}{\varepsilon}_{\mathbf{k}} + ikq_{\mathbf{k}} = 0, \tag{4.11}$$

where

$$q^\alpha_{\mathbf{k}} = \frac{1}{2m\sqrt{N}}\sum_i \Big[p_i^\alpha\big(\frac{\mathbf{p}_i^2}{2m} + \sum_{j(j\neq i)} V(|\mathbf{r}_{ij}|)\big) - \\ -\frac{1}{2}\sum_{j(j\neq i)} \frac{r_{ij}^\alpha}{r_{ij}^2}\mathbf{r}_{ij}(\mathbf{p}_i + \mathbf{p}_j)P_{\mathbf{k}}(r_{ij})\Big] e^{i\mathbf{k}\mathbf{r}_i} \tag{4.12}$$

is the energy flux.

The fluxes (4.6), (4.8), (4.12) govern the relaxations of the conserved variables, or more explicitly, the fluctuating components of fluxes determine the transport coefficients [1–3]. Before giving the expressions for transport coefficients let us consider so-called orthogonal dynamic variables $\{\hat{B}_\alpha\}$ which are connected with initial variables (4.4), (4.6) and (4.10) by linear

transformation and can be identified with thermodynamic quantities when $k \to 0$. Such variables can be found with the help of Schmidt's orthogonalization procedure using the expression (3.9) as the definition of scalar products. From here on we consider the dynamic variables as classical ones. This means that in our case $(A, B) = \langle AB \rangle_0$. For the longitudinal components we obtain $\{\hat{\mathcal{B}}^L_\alpha(\mathbf{k})\} = \{\hat{n}_\mathbf{k}, \hat{J}^L_\mathbf{k}, \hat{h}_\mathbf{k}\}$, where

$$\hat{h}_\mathbf{k} = (1 - \mathcal{P}_n)\hat{\varepsilon}_\mathbf{k} = \hat{\varepsilon}_\mathbf{k} - \langle \hat{\varepsilon}_\mathbf{k} \hat{n}_{-\mathbf{k}} \rangle_0 \langle \hat{n}_\mathbf{k} \hat{n}_{-\mathbf{k}} \rangle_0^{-1} \hat{n}_\mathbf{k} \tag{4.13}$$

is the density of generalized enthalpy, and

$$\mathcal{P}_n \ldots = \sum_\mathbf{k} \langle \ldots \hat{n}_{-\mathbf{k}} \rangle_0 \langle \hat{n}_\mathbf{k} \hat{n}_{-\mathbf{k}} \rangle_0^{-1} \hat{n}_\mathbf{k}, \tag{4.14}$$

is Mori-like projection operator. The variable $\hat{h}_\mathbf{k}$ is simply connected with the temperature variable as follows [2]

$$\hat{T}_\mathbf{k} = \frac{\hat{h}_\mathbf{k}}{nC_V(k)}, \tag{4.15}$$

where

$$C_V(k) = \frac{\langle \hat{h}_\mathbf{k} \hat{h}_{-\mathbf{k}} \rangle_0}{k_B T^2} \tag{4.16}$$

is generalized specific heat at constant volume.

For the transverse component in the hydrodynamic description we have only one variable, namely $\hat{\mathcal{B}}^T(\mathbf{k}) = \hat{J}^T_\mathbf{k}$. Now, using general results (3.15), (3.22) and (3.25), it is easy to derive the hydrodynamic equations for $\hat{\mathcal{B}}^{L,T}$. From comparison with results of phenomenological nonequilibrium thermodynamic the expressions for transport coefficients via the hydrodynamic memory functions can be obtained in the forms

$$\eta(k,z) + \frac{4}{3}\zeta(k,z) = \frac{mn}{k^2}\varphi^{H,L}_{JJ}(k,z), \tag{4.17}$$

$$\xi(k,z) = \frac{nC_V(k)T}{k^2}\varphi^H_{Jh}(k,z) = \frac{nm}{k^2}\varphi^H_{hJ}(k,z), \tag{4.18}$$

$$\lambda(k,z) = \frac{nC_V(k)}{k^2}\varphi^H_{hh}(k,z), \tag{4.19}$$

$$\zeta(k,z) = \frac{nm}{k^2}\varphi^{H,T}_{JJ}(k,z). \tag{4.20}$$

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 represents the dynamical coupling between the stress and the heat current, vanishing in the small k limit. The memory functions $\varphi^H_{\alpha,\beta}(k,z)$ are defined by (3.17) with $\hat{P} = \hat{\mathcal{B}}^{L,T}$. In this case for the frequency matrix we have

$$i\Omega_0^{H,L}(k) = \begin{pmatrix} 0 & i\Omega_{nJ}(k) & 0 \\ i\Omega_{Jn}(k) & 0 & i\Omega_{Jh}(k) \\ 0 & i\Omega_{hJ}(k) & 0 \end{pmatrix} \tag{4.21}$$

where

$$i\Omega_{nJ}(k) = \frac{ik}{m}, \quad i\Omega_{Jn}(k) = \frac{ik}{\beta S(k)},$$
$$i\Omega_{hJ}(k) = \frac{ikT\alpha(k)}{\beta m S(k)}, \; i\Omega_{Jh}(k) = \frac{ik\alpha(k)}{\beta S(k) C_V(k)}, \tag{4.22}$$

$S(k)$, $\alpha(k)$ are the static structure factor and the generalized thermal expansion coefficient, respectively. Then, as it follows from (3.22), the Laplace transform of the density-density time correlation function is given by

$$F_{nn}(k,z) = \langle \hat{n}_{\mathbf{k}} \hat{n}_{-\mathbf{k}} \rangle_0^z = \cfrac{S(k)}{z + \cfrac{k^2/(m\beta S(k))}{z + \Sigma_{JJ}(k,z)}} \tag{4.23}$$

where

$$\Sigma_{JJ}(k,z) = \Sigma_{JJ}^H(k,z) = \varphi_{JJ}^{H,L}(k,z) - \\ - \frac{(-i\Omega_{Jh}(k) + \varphi_{Jh}^{H,L}(k,z))(-i\Omega_{hJ}(k) + \varphi_{hJ}^{H,L}(k,z))}{z + \varphi_{hh}^{H,L}(k,z)}. \tag{4.24}$$

The similar results can be written for all other time correlation functions $\langle \hat{\mathcal{B}}^L(\mathbf{k}) \hat{\mathcal{B}}^L(-\mathbf{k}) \rangle_0^z$. In the limit $k \to 0$ the well-known result of hydrodynamic theory follows immediately from (4.23). In particular, the Landau-Placzek formula for the dynamic structure factor can be reproduced.

In viscoelastic theory when the current of momentum is included in the set of dynamic variables instead of $\hat{\varepsilon}_{\mathbf{k}}$, we obtain once again for $F_{nn}(k,z)$ the expression (4.23) but

$$\Sigma_{JJ}(k,z) = \Sigma_{JJ}^V(k,z) = -\frac{i\Omega_{J\pi}(k) i\Omega_{J\pi}(k)}{z + \varphi_{\pi\pi}^{V,L}(k,z)}, \tag{4.25}$$

where $\hat{\pi}_{\mathbf{k}} = (1 - \mathcal{P}_n - \mathcal{P}_J) i\hat{L} \hat{J}_{\mathbf{k}}^L$.

For the transverse fluctuations we have

$$F_{JJ}^T(k,z) = \frac{m/\beta}{z + \varphi_{JJ}^{H,T}(k,z)}. \tag{4.26}$$

Using certain approximation for the memory functions (Markovian, Gaussian, etc.), as it follows from (4.23) and (4.26), the problem of calculations of time correlation functions can be reduced to finding of some SCFs.

5. Extended set of dynamic variables

In order to develop the hydrodynamic description for intermediate range of k and ω many different modifications of hydrodynamic theory were proposed. One of the main future of these approaches is an extension of 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 is included into initial set of dynamic variables, but the fluctuation

of energy (4.10) is ignored. Later the variable of energy density $\varepsilon_{\mathbf{k}}$ was also included in the scheme of viscoelastic theory [6]. The results of viscoelastic theory can be reproduced from (3.15), (3.22) and (3.25) if the corresponding variables are considered.

Another way of theory modification is known as a mode coupling theory. Mode-coupling approach was proposed by Kawasaki [20], and later it was developed by Bosse et.al. [12] for simple classical liquid. In this approach the binary and other combinations of variables (4.4) and (4.6) have been included, besides of the particle number and momentum densities. Using the approximations for the memory functions in the form of two-mode decay integrals coupling longitudinal and transverse exitations the closed system of equations have been derived for time correlation functions. Such approach, as it has been shown is very fruitful especially for description of liquid–glass transition phenomena [17].

Below we consider a way of theory modification when the higher order derivatives of hydrodynamic variables are included into initial set of dynamic variables. As an example of such development the generalized mode approach can be noted [8–11]. In this approach using Markovian approximation for memory functions the general problem of dynamic theory is reduced to the calculation of generalized mode spectrum. The time correlation functions could be presented as a sum of partial terms connected with 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 were studied by Mryglod et. al. [10,11].

In order to compare the results of various approaches with different sets of dynamic variables [21] let us introduce the following notations

$$\begin{gathered}\hat{\mathcal{A}}_0 = \hat{\mathcal{B}}^{L,T}, \hat{\mathcal{A}}_1 = i\hat{L}\hat{\mathcal{B}}_0^{L,T}, \ldots, \\ \hat{\mathcal{A}}_l = (i\hat{L})^l \hat{\mathcal{B}}_0^{L,T} = i\hat{L}\hat{\mathcal{A}}_{l-1}, \ldots, \hat{\mathcal{A}}_s = (i\hat{L})^s \hat{\mathcal{B}}_0^{L,T},\end{gathered} \tag{5.1}$$

where $\hat{\mathcal{B}}^L = \{\hat{n}, \hat{\mathcal{B}}_0^L\}$, $\hat{\mathcal{B}}_0^L = \{\hat{J}^L, \hat{h}\}$ and $\hat{\mathcal{B}}^T = \hat{\mathcal{B}}_0^T = \hat{J}^T$. Here we took into account that $(i\hat{L})^l \hat{n} \sim (i\hat{L})^{l-1}\hat{J}^L$. The set of dynamical variables $\{\hat{\mathcal{A}}_0, \hat{\mathcal{A}}_1, \ldots, \hat{\mathcal{A}}_s\}$ can be considered as the initial one and includes $3 + 2s$ variables for longitudinal fluctuations and $1 + s$ variables for transverse fluctuations.

The set of orthogonalized variables constructed on (5.1) with the definition of scalar product in the form (3.9) can be found with the help of Schmidt's orthogonal procedure. As a result we have

$$\begin{aligned}\hat{Y}_0 &= \hat{\mathcal{A}}_0, \\ \hat{Y}_1 &= (1 - \mathcal{P}_0)\hat{\mathcal{A}}_1, \\ &\ldots, \\ \hat{Y}_l &= (1 - \mathcal{P}_{l-1})i\hat{L}\hat{\mathcal{A}}_{l-1} = (1 - \mathcal{P}_{l-1})\hat{\mathcal{A}}_l, \\ &\ldots, \\ \hat{Y}_s &= (1 - \mathcal{P}_{s-1})\hat{\mathcal{A}}_s,\end{aligned} \tag{5.2}$$

where

$$\mathcal{P}_0 = \Delta\mathcal{P}_0 = \langle \ldots \mathcal{A}_0^+ \rangle_0 \langle \mathcal{A}_0 \mathcal{A}_0^+ \rangle_0^{-1} \mathcal{A}_0, \tag{5.3}$$

$$\mathcal{P}_l = \sum_{m=0}^{l} \Delta\mathcal{P}_m, \tag{5.4}$$

$$\Delta\mathcal{P}_m = \langle \ldots \hat{Y}_m^+ \rangle_0 \langle \hat{Y}_m \hat{Y}_m^+ \rangle_0^{-1} \hat{Y}_m, \tag{5.5}$$

are Mori-like projection operators. For the variables $\{\hat{Y}_l\}$ we have

$$\langle \hat{Y}_l^\alpha \hat{Y}_{l'}^\beta \rangle_0 = \delta_{ll'} \delta_{\alpha\beta} \langle \hat{Y}_l^\alpha \hat{Y}_l^\alpha \rangle_0. \tag{5.6}$$

Using the general expressions obtained above and the properties of projection operator of Mori, the frequency matrix and matrix of memory functions for $\hat{P} = \{\hat{Y}_0, \hat{Y}_1, \ldots, \hat{Y}_n\}$ can be found. For frequency matrix we have

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

where

$$i\Omega_l = i\Omega_{ll} = \langle iL\hat{Y}_l \hat{Y}_l^+ \rangle_0 \langle \hat{Y}_l \hat{Y}_l^+ \rangle_0^{-1}, \tag{5.7}$$

$$\Gamma_l = i\Omega_{l+1,l} = \langle i\hat{L}\hat{Y}_{l+1} \hat{Y}_l^+ \rangle_0 \langle \hat{Y}_l \hat{Y}_l^+ \rangle_0^{-1} = -\langle \hat{Y}_{l+1} \hat{Y}_{l+1}^+ \rangle_0 \langle \hat{Y}_l \hat{Y}_l^+ \rangle_0^{-1}, \tag{5.8}$$

$$i\Omega_{l-1,l} = \langle i\hat{L}\hat{Y}_{l-1} \hat{Y}_l^+ \rangle_0 \langle \hat{Y}_l \hat{Y}_l^+ \rangle_0^{-1} = \langle \hat{Y}_l \hat{Y}_l^+ \rangle_0 \langle \hat{Y}_l \hat{Y}_l^+ \rangle_0^{-1} = I, \tag{5.9}$$

and in general case I is a unit matrix. All other elements of frequency matrix are equal zero. From the definition of matrix of memory functions (3.17) and the equality

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

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

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

where

$$\tilde{\varphi}_s(z) = \langle \hat{Y}_{s+1} \frac{1}{z + (1 - \mathcal{P}_s) i\hat{L}} \hat{Y}_{s+1}^+ \rangle_0 \langle \hat{Y}_s \hat{Y}_s^+ \rangle_0^{-1}. \tag{5.10}$$

The set of equations for $\langle \hat{Y}_l \rangle^\omega$ can be found from (2.28)

$$\begin{aligned} \left(i\omega - i\Omega_0\right) \langle \Delta\hat{Y}_0 \rangle^\omega - \langle \Delta\hat{Y}_1 \rangle^\omega &= 0, \\ \Gamma_0 \langle \Delta\hat{Y}_0 \rangle^\omega + \left(i\omega - i\Omega_1\right) \langle \Delta\hat{Y}_1 \rangle^\omega - \langle \Delta\hat{Y}_2 \rangle^\omega &= 0, \\ \ldots\ldots\ldots\ldots\ldots\ldots \\ \Gamma_{s-2} \langle \Delta\hat{Y}_{s-2} \rangle^\omega + \left(i\omega - i\Omega_{s-1}\right) \langle \Delta\hat{Y}_{s-1} \rangle^\omega - \langle \Delta\hat{Y}_s \rangle^\omega &= 0, \\ \Gamma_{s-1} \langle \Delta\hat{Y}_{s-1} \rangle^\omega + \left(i\omega - i\Omega_s + \tilde{\varphi}_s(z)\right) \langle \Delta\hat{Y}_n \rangle^\omega &= 0. \end{aligned} \tag{5.11}$$

Solving the last equation with respect to $\langle \Delta \hat{Y}_s \rangle^\omega$ and excluding this variable from the set (5.11) we obtain the set of equations for $\hat{P} = \{\hat{Y}_0, \hat{Y}_1, \ldots, \hat{Y}_{s-1}\}$ with the matrix of memory functions $\tilde{\varphi}_{s-1}(z)$ in the form

$$\tilde{\varphi}_{s-1}(z) = \Big(zI - i\Omega_s + \tilde{\varphi}_s(z)\Big)^{-1} \Gamma_{s-1}. \tag{5.12}$$

The recurrent relation (5.12) gives 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 (5.12) is in fact exact and can be used for finding the approximated expressions for the memory functions.

Let us consider some examples. Using (5.12) one can reproduce the well-known results of Mori for dynamic structural factor in the form of an ordinary continued fraction [18]. In this case for longitudinal fluctuations it is necessary to include in the initial set of dynamic variables the densities of particle number and momentum as well as derivatives of the momentun density only. Then, as it follows from (5.7), $i\Omega_l = 0$. Besides, Γ_l and $\tilde{\varphi}_l(z)$ are usual functions, and we have

$$\tilde{\varphi}_{s-1}(z) = \frac{\Gamma_{s-1}}{z + \tilde{\varphi}_s(z)}. \tag{5.13}$$

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 matrixes. In this case the recurrent relation (5.12) for arbitrary s can be rewritten as follows

$$\tilde{\varphi}^{11}_{s-1}(z) = \frac{\Gamma^{11}_{s-1}}{z + \tilde{\varphi}^{11}_s(z) - \dfrac{(-i\Omega^{12}_s + \tilde{\varphi}^{12}_s(z))(-i\Omega^{21}_s + \tilde{\varphi}^{21}_s(z))}{z + \tilde{\varphi}^{22}_s(z)}}, \tag{5.14}$$

$$\tilde{\varphi}^{12}_{s-1}(z) = \frac{\Gamma^{22}_{s-1}}{-i\Omega^{21}_s + \tilde{\varphi}^{21}_s(z) - \dfrac{(z + \tilde{\varphi}^{11}_s(z))(z + \tilde{\varphi}^{22}_s(z))}{-i\Omega^{12}_s + \tilde{\varphi}^{12}_s(z)}}, \tag{5.15}$$

$$\tilde{\varphi}^{21}_{s-1}(z) = \frac{\Gamma^{11}_{s-1}}{-i\Omega^{12}_s + \tilde{\varphi}^{12}_s(z) - \dfrac{(z + \tilde{\varphi}^{11}_s(z))(z + \tilde{\varphi}^{22}_s(z))}{-i\Omega^{21}_s + \tilde{\varphi}^{21}_s(z)}}, \tag{5.16}$$

$$\tilde{\varphi}^{22}_{s-1}(z) = \frac{\Gamma^{22}_{s-1}}{z + \tilde{\varphi}^{22}_s(z) - \dfrac{(-i\Omega^{21}_s + \tilde{\varphi}^{21}_s(z))(-i\Omega^{12}_s + \tilde{\varphi}^{12}_s(z))}{z + \tilde{\varphi}^{11}_s(z)}}. \tag{5.17}$$

The known relations [19,9] for the memory functions in three- and five-mode descriptions ($s = 1$) follow immediately from (5.14)-(5.17). Using the equations for time correlation functions (3.22) and the expressions (5.14)-(5.17), the representations for time correlation functions in the form of "generalized continued fraction" can be derived. As it was pointed out in [22] such expressions for the time correlation functions correctly conveys the analytic structure of the corresponding Green functions in the complex z-plane.

6. Discussion and concluding remarks

Let us discuss the results obtained above. Using a general formalism of Zubarev's method of the nonequilibrium statistical operator we derived the equations of linear relaxation theories for arbitrary set of dynamic variables. For the case when the initial set of dynamic variables includes besides the conservative quantities their derivatives we found the relations between lower- and higher-order memory functions. This means, as it follows from (4.17)-(4.20) and (5.12) that the generalized transport coefficients can be expressed via the static correlation functions and higher-order memory functions. It can be shown that relation (5.12) is in fact exact if any approximations are not used. This is evident from expression (3.17) that, if we choose for $\hat{P}$ the most slow variables, the elements of the matrix of memory functions constructed on projected variables have to fall faster with time than the time correlation functions. This means that Markovian approximation for the memory functions can be used as good zero-order approximation. Using Markovian approximation on the level of hydrodynamic description (see Sec.4) one can see that the transport coefficients are functions of wave-vector k only. The extended set of dynamic variables in Markovian approximation gives the following possibilities: (i) to calculate the generalized transport coefficients as functions of k and ω; (ii) to obtain more detail information about the time correlation functions, in particular, to find the dynamic structure factor for which the sum rules will be satisfied in the higher-orders; (iii) to calculate the spectrum and eigenvectors of generalized collective modes and to present the time correlation functions via these quantities. Moreover, it can be shown [10,11] that the memory functions for arbitrary set of dynamic variables can be written via the static correlation functions and the same set of so-called hydrodynamic correlation times. These problems will be considered in more detail elsewhere.

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ДО НЕРІВНОВАЖНОЇ СТАТИСТИЧНОЇ ТЕОРІЇ РІДИН: ТЕОРІЇ ЛІНІЙНОЇ РЕЛАКСАЦІЇ З РІЗНИМИ НАБОРАМИ ДИНАМІЧНИХ ЗМІННИХ

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Методом нерівноважного статистичного оператора Зубарева отримані загальні рівняння теорії лінійної релаксації для довільного набору динамічних змінних. Проведено порівняльний аналіз отриманих результатів та знайдені рекурентні зв'язки між відповідними функціями пам'яті. Показано, що рівняння лінійної релаксації для довільного набору динамічних змінних є точними, коли функції пам'яті задані точними виразами для них. Проведено порівняння з іншими роботами.