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ON THE STATISTICAL HYDRODYNAMICS FOR A BINARY
MIXTURE OF MAGNETIC AND NONMAGNETIC ATOMS

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ЛЬВІВ

До статистичної гідродинаміки бінарної суміші магнітних та немагнітних атомів

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Анотація. Методом нерівноважного статистичного оператора досліджуються динамічні властивості суміші магнітних та немагнітних атомів. Отримані і проаналізовані рівняння узагальненої гідродинаміки. На цій основі розраховано спектр гідродинамічних колективних мод. Знайдена швидкість поширення звуку та коефіцієнти загасання гідродинамічних мод. Пропонується схема розрахунку часових кореляційних функцій у гідродинамічній границі і обговорюються вирази для них, отримані у парамагнітному випадку.

On the statistical hydrodynamics for a binary mixture of magnetic and nonmagnetic atoms

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Abstract. Dynamic properties of a binary mixture of magnetic and nonmagnetic particles is considered with the help of the method of nonequilibrium statistical operator. The generalized hydrodynamic equations are derived and analyzed. On this basis hydrodynamic collective modes spectrum was calculated. The expressions for sound velocity and damping coefficients of collective hydrodynamic modes are found. We propose the consistent scheme for calculation of time correlation functions in the hydrodynamic limit and discuss the expressions found for them in paramagnetic case.

1. Introduction

Magnetic liquids, mixtures of magnetic and nonmagnetic atoms in the external fields of mechanical or electromagnetic origin, have already taken their significant place in chemical, electronic and other modern technologies. That is why the investigations of the thermodynamical, structural and dynamical properties of liquid magnets are very actual for more deep understanding and forecasting of their behavior [1–3].

The investigation of time-dependent correlation functions as well as generalized transport coefficients of a liquid mixture of magnetic and nonmagnetic atoms are very interesting and valuable. They give us possibility of deep insight into the processes in the systems with coupled classical and quantum peculiarities. From the theoretical point of view one of the most interesting problems is investigation of the behavior of hydrodynamic collective modes, which describe the properties of heat, sound, and mass fluctuations. Another important aspect of this problem is the derivation of expressions for dynamic structure factors. It is known, that these functions can be extract from scattering experiments. Such theoretical study should be based on the statistical approach, on the equations of generalized hydrodynamics, particularly. Similar approach was applied to one-component magnetic mixture [4–8]. The collective modes of Heisenberg ferrofluid were also considered in [8].

Statistical hydrodynamics for a mixture of magnetic and nonmagnetic atoms in an external nonhomogeneous magnetic field $\mathbf{B}(\mathbf{r}; t)$ was studied in [9]. There was formulated the problem of derivation of generalized hydrodynamic equations for magnetic and nonmagnetic subsystems with the help of nonequilibrium statistical operator method for description both strong and weak nonequilibrium states. Magnetic and nonmagnetic subsystems were characterized by individual nonequilibrium thermodynamics parameters. As a result nonequilibrium thermodynamical relations and generalized equations of hydrodynamics were derived.

This paper is dedicated to the study of a binary magnetic mixture consisting of magnetic and nonmagnetic liquids within the method of nonequilibrium statistical operator. On the base of a set consisting of five parameters of abbreviated description, for the weak nonequilibrium case the calculation of hydrodynamic collective modes are carried out with the help of the perturbation theory. In the chapter 7 the problem of calculation of time-correlation functions is considered. We propose here the scheme which allows to calculate the weight coefficients describing a partial contribution of each mode to the hydrodynamic time correlation functions. In some limiting cases (pure non-magnetic fluid, simple magnetic liquid, etc.) the obtained results are discussed in comparison with the results known in the literature.

2. Theoretical framework of the method of nonequilibrium statistical operator

Let us start with the Liouville equation:

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

where classical part of $\mathrm{i}\hat{L}$ is determined as a Poisson brackets of the function ρ with a classical part of the Hamiltonian of the system and as a commutator with its quantum part, ρ is a function of phase variables $x^N = \{\boldsymbol{r}, \mathbf{p}, \hat{\boldsymbol{s}}\}^N$, where N is a total number of particles.

Following Zubarev's method of nonequilibrium statistical operator [10] we can rewrite equation (2.1) in the form:

$$\left(\frac{\partial}{\partial t} + \mathrm{i}\hat{L}\right)\rho(x^N) = -\varepsilon\left(\rho(x^N) - \rho_q(x^N)\right), \tag{2.2}$$

where $\varepsilon \to 0$, ρ_q is so-called quasi-equilibrium statistical operator. Nonzero right-hand side of the equation (2.2) imposes the boundary conditions, which destroy the time reversal symmetry of the Liouville equation. To restrict our consideration to the set of slow physical quantities $\left\{\hat{P}_\alpha\right\}$ which are thought to determine nonequilibrium state (the set of conserved quantities), we can write ρ_q in the Gibbs-like form:

$$\rho_q = exp\left\{-\Phi(t) - \sum_\alpha \hat{P}_\alpha F_\alpha(t)\right\}, \tag{2.3}$$

with conditions of self-consistency:

$$\langle\hat{P}_\alpha\rangle^t = \langle\hat{P}_\alpha\rangle^t_q \quad \text{or} \quad \mathrm{Sp}\left(\hat{P}_\alpha\rho(x^N, t)\right) = \mathrm{Sp}\left(\hat{P}_\alpha\rho_q(x^N, t)\right), \tag{2.4}$$

which determine $\{F_\alpha(t)\}$. The index $\alpha = \{i, \boldsymbol{k}\}$ denotes a combination of discrete index i which numerates the variables and wave vector $\boldsymbol{k}$ so, that summation in (2.3) means:

$$\sum_\alpha \cdots = \sum_i \sum_{\boldsymbol{k}} \cdots,$$

Taking into account projecting, the formal solution of equation (2.2) can be written [10,11]:

$$\rho(t) = \rho_q(t) + \sum_\alpha \int_{-\infty}^{t} dt'\, e^{-\varepsilon(t-t')} F_\alpha(t') T(t,t')$$

$$\times \int_0^1 d\tau\, \rho_q^\tau(t') \hat{I}_\alpha(t') \rho_q^{1-\tau}(t'), \qquad (2.5)$$

where

$$\hat{I}_\alpha(t) = (1 - \mathcal{P}(t))\dot{\hat{P}}_\alpha \qquad (2.6)$$

are the generalized fluxes, $\dot{\hat{P}}_\alpha \equiv i\hat{L}\,\hat{P}_\alpha$.

$$T(t,t') = exp_+ \left\{ -\int_{t'}^{t} d\tau (1 - \mathcal{P}(\tau)) i\hat{L}(\tau) \right\} \qquad (2.7)$$

is the operator of time evolution with the Mori-like projection operator $\mathcal{P}(t)$:

$$\mathcal{P}(t)\ldots = \langle\ldots\rangle + \sum_\alpha \frac{\delta\langle\ldots\rangle_q^t}{\delta\langle\hat{P}_\alpha\rangle^t} \left\{ \hat{P}_\alpha - \langle\hat{P}_\alpha\rangle^t \right\} \qquad (2.8)$$

which have the following properties:

$$\mathcal{P}(t)\hat{P}_\alpha = \hat{P}_\alpha, \quad \mathcal{P}(t)\mathcal{P}(t') = \mathcal{P}(t), \quad \mathcal{P}(1 - \mathcal{P}(t')) = 0,$$

Statistical operator (2.5) determines generalized transport equations in the form:

$$\frac{\partial}{\partial t}\langle\hat{P}_\alpha\rangle^t = \langle\dot{\hat{P}}_\alpha\rangle_q^t + \sum_\beta \int_{-\infty}^{t} dt'\, e^{\varepsilon(t-t')}\phi_{\alpha\beta}(t,t')F_\beta(t')dt', \qquad (2.9)$$

where

$$\phi_{\alpha\beta}(t,t') = \int_0^1 d\tau\, \langle \hat{I}_\alpha(t),\, T(t,t')\, \rho_q^\tau\, \hat{I}_\beta(t')\, \rho_q^{-\tau}\rangle_q^t \qquad (2.10)$$

are the generalized memory functions, or generalized transport kernels. Equations (2.9), (2.10) and (2.4) make up a closed system of nonlinear equations which describe both strong and weak nonequilibrium.

Now we will consider weak nonequilibrium, which allows us to linearize system (2.4), (2.9), (2.10). For the small deviations –

$$\delta\hat{P}_\alpha(t) = \langle\hat{P}_\alpha\rangle^t - \langle\hat{P}_\alpha\rangle_0 \qquad (2.11)$$

of averages $\langle\hat{P}_\alpha\rangle^t = \text{Sp}\,\hat{P}_\alpha\rho(t)$ from the equilibrium values $\langle\hat{P}_\alpha\rangle_0 = \text{Sp}\,\hat{P}_\alpha\rho_0(x^N)$, where

$$\rho_0(x^N) = exp\left\{ -\Phi_0 - \sum_\alpha \hat{P}_\alpha F_\alpha^0 \right\}, \qquad (2.12)$$

is the equilibrium statistical operator, deviations of the intensive quantities $\delta F_n(t) = F_n(t) - F_n^0$ can be easily found from the self-consistency conditions (2.4). In matrix form:

$$\delta F(t) = -(\Delta\hat{P}, \Delta\hat{P}^+)^{-1}\Delta\hat{P} \qquad (2.13)$$

where

$$\|(\Delta\hat{P}, \Delta\hat{P}^+)\|_{ij} = (\Delta\hat{P}_i, \Delta\hat{P}_j), \qquad (2.14)$$
$$\Delta\hat{P}_i = \hat{P}_i - \langle\hat{P}_i\rangle,$$

and $(\hat{A}, \hat{B})$ means correlation function:

$$(\hat{A}, \hat{B}) = \int_0^1 \langle\hat{A}\rho_0^\tau\hat{B}\rho_0^{-\tau}\rangle\, d\tau, \qquad (2.15)$$

which transforms into simple average $\langle\hat{A}\hat{B}\rangle_0$ in classical case. Now we can rewrite (2.5) in the form

$$\delta\rho(t) = \delta\rho_q(t) + \sum_\alpha \int_{-\infty}^{t} dt'\, e^{-\varepsilon(t-t')}\, \delta F_\alpha(t')\, T_0(t-t')$$

$$\times \int_0^1 d\tau\, \rho_0^\tau\, (1 - \mathcal{P})\, \dot{\hat{P}}_\alpha\, \rho_0^{1-\tau}, \qquad (2.16)$$

where the projection operator is given by

$$\mathcal{P}\ldots = (\ldots, \Delta\hat{P}^+)(\Delta\hat{P}, \Delta\hat{P}^+)^{-1}\Delta\hat{P}. \qquad (2.17)$$

For the Laplace transforms of time-dependent functions ($f(\omega) = \int\limits_0^\infty dt\, f(t)e^{-\omega t}$), using the equality

$$\frac{\partial}{\partial t}\langle\Delta\hat{P}\rangle^t = \langle i\hat{L}\,\hat{P}\rangle^t, \tag{2.18}$$

we get the generalized hydrodynamic equations in the form

$$(i\omega - i\Omega + \phi_\varepsilon(\omega))\,\Delta\hat{P}(\omega) = 0, \tag{2.19}$$

where

$$i\Omega = (\dot{\hat{P}}, \Delta\hat{P}^+)(\Delta\hat{P}, \Delta\hat{P}^+)^{-1}, \tag{2.20}$$

$$\phi_\varepsilon(\omega) = \left((1-\mathcal{P})\dot{\hat{P}}\,,\, \frac{1}{i\omega + \varepsilon + (1-\mathcal{P})i\hat{L}}(1-\mathcal{P})\dot{\hat{P}}^+\right)$$
$$\times(\Delta\hat{P}, \Delta\hat{P}^+)^{-1} \tag{2.21}$$

are the matrices of frequencies and memory functions.

The matrix equation for the Laplace-transforms $(\Delta\hat{P}, \Delta\hat{P}^+)^z$ of time correlation functions

$$(\Delta\hat{P}, \Delta\hat{P}^+)^z = \left(\Delta\hat{P}, \frac{1}{z+i\hat{L}}\Delta\hat{P}^+\right), \tag{2.22}$$

has the structure, similar to that of (2.19):

$$\{z - i\Omega + \phi_\varepsilon(z)\}(\Delta\hat{P}, \Delta\hat{P}^+)^z = (\Delta\hat{P}, \Delta\hat{P}^+). \tag{2.23}$$

Retarded correlation Green functions can be expressed in terms of time correlation functions $(\Delta\hat{P}, \Delta\hat{P}^+)^t$ (which are connected with $(\Delta\hat{P}, \Delta\hat{P}^+)^z$ by Laplace transformation):

$$G^{(r)}_{ij}(t) = -i\Theta(t)(\Delta\hat{P}, \Delta\hat{P}^+)^t. \tag{2.24}$$

Hence, the poles of the retarded Green functions, which give the spectrum of collective modes are determined by matrix equation

$$det|z - i\Omega + \phi_\varepsilon(z)| = 0. \tag{2.25}$$

3. Dynamic variables

Let us consider the system consisting of N_1 nonmagnetic and N_2 magnetic particles posed in external magnetic field. Hamiltonian of such a system can be written as in [12,13], taking into account the interaction with nonmagnetic subsystem:

$$\hat{H}(t) = H_1 + \hat{H}_2 + H_{int} + \hat{H}_{ex}. \tag{3.1}$$

Here and further subscripts $_1$, $_2$ or superscripts in parentheses $^{(1)}$, $^{(2)}$ indicate nonmagnetic and magnetic subsystem, respectively. Thus H_1 and $\hat{H}_2$ are the Hamiltonians of nonmagnetic and magnetic subsystems separately, H_{int} describes their interaction and $\hat{H}_{ex}$ is the energy of spin interaction with external magnetic field.

The Hamiltonian H_1 of nonmagnetic subsystem can be taken in classical form

$$H_1 = \sum_{j=1}^{N_1}\frac{\boldsymbol{p}_j^{(1)\,2}}{2m_1} + \frac{1}{2}\sum_{j\neq l}^{N_1,N_1}V^{(11)}(\boldsymbol{r}_{jl}), \tag{3.2}$$

where $V^{(11)}(\boldsymbol{r}_{jl})$ is the potential of interaction between 2 nonmagnetic particles j and l, which can be chosen for calculations in any convenient form; and m_1 is a mass of nonmagnetic particles.

The term $\hat{H}_2$

$$\hat{H}_2 = H_{2L} - \frac{1}{2}\sum_{j\neq l}^{N_2,N_2}J(\boldsymbol{r}_{jl})(\hat{\boldsymbol{s}}_j, \hat{\boldsymbol{s}}_l) \tag{3.3}$$

consists of classical, "liquid" part H_{2L} which has the same form as H_1 and quantum part, which describes spin subsystem and can be taken in Heisenberg-like form.

Other terms in (3.1) could be written in the form

$$H_{int} = \sum_{i,j}^{N_1,N_2}V^{(12)}(\boldsymbol{r}_{ij}), \tag{3.4}$$

$$\hat{H}_{ex} = -\sum_{i}^{N_2}\hat{\boldsymbol{s}}_i\cdot\hat{\boldsymbol{B}}(\boldsymbol{r}_i, t), \tag{3.5}$$

where $V^{(12)}(\boldsymbol{r}_{ij})$ is the potential of interaction between i-th nonmagnetic and j-th magnetic particle, $\hat{\boldsymbol{B}}(\boldsymbol{r}_i, t)$ – external magnetic field.

Liouville operator which corresponds to Hamiltonian (3.1) can be written as follows:

$$\mathrm{i}\hat{L} = \mathrm{i}L_1 + \mathrm{i}\hat{L}_2 + \mathrm{i}L_{int} + \mathrm{i}\hat{L}_{ex} + \mathrm{i}\hat{L}_s, \qquad (3.6)$$

where

$$\mathrm{i}L_1 = \sum_i^{(1)} \frac{\boldsymbol{p}_i^{(1)}}{m_1} \frac{\partial}{\partial \boldsymbol{r}_i} - \frac{1}{2} \sum_{i\neq j}^{(1,1)} \frac{\partial}{\partial \boldsymbol{r}_i} V^{(11)}(\boldsymbol{r}_{ij}) \left(\frac{\partial}{\partial \boldsymbol{p}_i^{(1)}} - \frac{\partial}{\partial \boldsymbol{p}_j^{(1)}} \right), \quad (3.7)$$

$$\mathrm{i}\hat{L}_2 = \mathrm{i}L_{(1\to 2)} + \frac{1}{2} \sum_{i\neq j}^{(2,2)} \frac{\partial}{\partial \boldsymbol{r}_i} J(\boldsymbol{r}_{ij})(\hat{\boldsymbol{s}}_i, \hat{\boldsymbol{s}}_j) \left(\frac{\partial}{\partial \boldsymbol{p}_i^{(2)}} - \frac{\partial}{\partial \boldsymbol{p}_j^{(2)}} \right), \quad (3.8)$$

$$\mathrm{i}L_{int} = -\sum_{i,j}^{(1,2)} \frac{\partial}{\partial \boldsymbol{r}_i} V^{(12)}(\boldsymbol{r}_{ij}) \left(\frac{\partial}{\partial \boldsymbol{p}_i^{(1)}} - \frac{\partial}{\partial \boldsymbol{p}_j^{(2)}} \right), \qquad (3.9)$$

$$\mathrm{i}\hat{L}_{ex} = \sum_i^{(2)} \frac{\partial \left(\hat{\boldsymbol{B}}(\boldsymbol{r}_i, t)\cdot\hat{\boldsymbol{s}} \right)}{\partial \boldsymbol{r}_i} \cdot \frac{\partial}{\partial \boldsymbol{p}_i^{(2)}}, \qquad (3.10)$$

and $\mathrm{i}\hat{L}_s$ is a purely quantum part of Liouville operator, it is determined as a commutator

$$\mathrm{i}\hat{L}_s\cdot\hat{A} = \frac{\mathrm{i}}{\hbar} \left[-\frac{1}{2}\sum_{i\neq j}^{(2,2)} J(\boldsymbol{r}_{ij})(\hat{\boldsymbol{s}}_i, \hat{\boldsymbol{s}}_j) - \sum_i^{(2)} \left(\hat{\boldsymbol{B}}(\boldsymbol{r}_i, t)\cdot\hat{\boldsymbol{s}}_i \right), \ \hat{A} \right]. \quad (3.11)$$

To study the dynamics near the equilibrium, we have to consider all the conserved quantities and most slow ones associated with them. For our model five parameters of abbreviated description $\{\hat{P}_i\}$, $i = 1..5$ can be chosen, namely: partial densities of particle number $\hat{n}_1(\boldsymbol{r})$, $\hat{n}_2(\boldsymbol{r})$, densities of momentum $\hat{\boldsymbol{p}}(\boldsymbol{r})$, magnetization $\hat{\boldsymbol{m}}(\boldsymbol{r})$ and total energy $\hat{\varepsilon}(\boldsymbol{r})$.

After the Fourier transformation $(f(\boldsymbol{k}) = \int dr\, f(\boldsymbol{r})e^{\mathrm{i}\boldsymbol{k}\boldsymbol{r}})$, these values can be written as follows:

$$\hat{n}_1(\boldsymbol{k}) = \sum_i^{(1)} e^{\mathrm{i}\boldsymbol{k}\boldsymbol{r}_i}, \qquad \hat{n}_2(\boldsymbol{k}) = \sum_i^{(2)} e^{\mathrm{i}\boldsymbol{k}\boldsymbol{r}_i}, \qquad (3.12)$$

$$\hat{\boldsymbol{p}}^\alpha(\boldsymbol{k}) = \sum_i^{(1)} \boldsymbol{p}_i^{(1)\,\alpha} e^{\mathrm{i}\boldsymbol{k}\boldsymbol{r}_i} + \sum_i^{(2)} \boldsymbol{p}_i^{(2)\,\alpha} e^{\mathrm{i}\boldsymbol{k}\boldsymbol{r}_i}, \qquad (3.13)$$

$$\hat{\boldsymbol{m}}^\alpha(\boldsymbol{k}) = \sum_i^{(2)} \hat{\boldsymbol{s}}_i^\alpha e^{\mathrm{i}\boldsymbol{k}\boldsymbol{r}_i}, \qquad (3.14)$$

$$\hat{\varepsilon}(\boldsymbol{k}) = \sum_i^{(1)} \hat{\varepsilon}_i^{(1)} e^{\mathrm{i}\boldsymbol{k}\boldsymbol{r}_i} + \sum_i^{(2)} \hat{\varepsilon}_i^{(2)} e^{\mathrm{i}\boldsymbol{k}\boldsymbol{r}_i}, \qquad (3.15)$$

index α indicates spatial α-component of vector, and

$$\hat{\varepsilon}_i^{(1)} = \frac{\boldsymbol{p}_i^{(1)\,2}}{2m_1} + \frac{1}{2} \sum_{j(\neq i)}^{(1)} V^{(11)}(\boldsymbol{r}_{ij}) + \frac{1}{2} \sum_j^{(2)} V^{(12)}(\boldsymbol{r}_{ij}), \qquad (3.16)$$

$$\hat{\varepsilon}_i^{(2)} = \hat{\varepsilon}_i^{(1\to 2)} - \frac{1}{2} \sum_{i\neq j}^{(2,2)} J(\boldsymbol{r}_{ij})(\hat{\boldsymbol{s}}_i, \hat{\boldsymbol{s}}_j). \qquad (3.17)$$

For our set of variables $\hat{P}_i$, the quantum equations of motion have the following form (see Appendix)

$$\dot{\hat{P}}_i(\boldsymbol{k}) = \mathrm{i}\boldsymbol{k}^\alpha J_i^\alpha(\boldsymbol{k}) + R_i(\boldsymbol{k}), \qquad (3.18)$$

$$\dot{\hat{P}}_l^\alpha(\boldsymbol{k}) = \mathrm{i}\boldsymbol{k}^\beta J_l^{\alpha\beta}(\boldsymbol{k}) + R_l^\alpha(\boldsymbol{k}), \qquad (3.19)$$

for the scalar $\{\hat{n}_1(\boldsymbol{k}), \hat{n}_2(\boldsymbol{k}), \hat{\varepsilon}(\boldsymbol{k})\}$ and vector $\{\hat{\boldsymbol{p}}(\boldsymbol{k}), \hat{\boldsymbol{m}}(\boldsymbol{k})\}$ variables respectively. Terms $R_i(\boldsymbol{k})$, $R_l^\alpha(\boldsymbol{k})$ appeared due to nonhomogeneous external magnetic field. When we assume that $\hat{\boldsymbol{B}}(\boldsymbol{r}, t)$ is homogeneous, these terms disappear and variables $\{\hat{P}\}$ become conserved.

4. Static correlation functions

For analysis of the generalized hydrodynamic equations (2.19) one needs to calculate the static correlation functions constructed on the variables

$$\hat{P}(\boldsymbol{k}) = \{\hat{n}_1(\boldsymbol{k}), \hat{n}_2(\boldsymbol{k}), \hat{\boldsymbol{p}}(\boldsymbol{k}), \hat{\boldsymbol{m}}(\boldsymbol{k}), \hat{\varepsilon}(\boldsymbol{k})\} \qquad (4.1)$$

Let us define static correlation function $(\hat{a}, \hat{b})$ as an average of deviations:

$$(\hat{a}, \hat{b}) = \int_0^1 d\tau \langle \Delta\hat{a}\rho_0^\tau \Delta\hat{b}\rho_0^{-\tau} \rangle, \qquad (4.2)$$

contrary to (2.15), where it was defined as a simple average.

In order to give some thermodynamical interpretation of correlation functions we chose the equilibrium statistical operator (2.12) as a Gibbs distribution for the grand canonical ensemble $(\mu, V, T, \hat{\boldsymbol{b}})$, $\mu = \{\mu_1, \mu_2\}$:

$$\rho_0 = \exp\left[\beta(\Omega - \hat{\omega})\right], \qquad (4.3)$$

$$\hat{\omega} \equiv \hat{\varepsilon} - \mu_1 \hat{n}_1 - \mu_2 \hat{n}_2 - \hat{\boldsymbol{b}}\hat{\boldsymbol{m}}, \qquad (4.4)$$

$$\Omega = \Omega(\mu, \hat{\boldsymbol{b}}, T). \qquad (4.5)$$

where Ω is the thermodynamical potential, β is the inverse temperature, $\hat{n}_1, \ldots, \hat{\varepsilon}$ are the quantities (4.1), taken with $\boldsymbol{k} = 0$: $\hat{n}_1 = \hat{n}_1(\boldsymbol{k} = 0), \ldots, \hat{\varepsilon} = \hat{\varepsilon}(\boldsymbol{k} = 0)$; and $\hat{\boldsymbol{b}}$ is an internal magnetic field.

For an arbitrary operator $\hat{a}$ and parameter γ it is easy to prove the equation

$$\frac{\partial \langle \hat{a} \rangle}{\partial \gamma} = - \left(\hat{a}, \frac{\partial(\beta\hat{\omega})}{\partial \gamma} \right), \qquad (4.6)$$

where the average $\langle \ldots \rangle$ is performed with the distribution (4.3). So, for example, if $\hat{a} = \hat{n}_1$, $\gamma = \mu_1$, we get:

$$\frac{\partial N_1}{\partial \mu_1} = \beta(\hat{n}_1, \hat{n}_2). \qquad (4.7)$$

Here and further the quantity, written by a capital letter denotes the average value of corresponding operator written by a small letter, for instance, $N_1 = \langle \hat{n}_1 \rangle$. In the same manner with the help of (4.6) we can connect other correlation functions with thermodynamical quantities:

$$(\hat{n}_i, \hat{n}_j) = \frac{1}{\beta} \left(\frac{\partial N_i}{\partial \mu_j} \right)_{T,b} = \frac{1}{\beta} \left(\frac{\partial N_j}{\partial \mu_i} \right)_{T,b}, \quad i,j = 1,2 \quad , \qquad (4.8)$$

$$(\hat{n}_i, \hat{m}) = \frac{1}{\beta} \frac{\partial N_i}{\partial b} = \frac{1}{\beta} \left(\frac{\partial M}{\partial \mu_i} \right)_{T,b}, \qquad (4.9)$$

$$(\hat{m}, \hat{m}) = \frac{1}{\beta} \left(\frac{\partial M}{\partial b} \right)_{T,\mu}, \qquad (4.10)$$

$$(\hat{\varepsilon}, \hat{m}) = \frac{1}{\beta} \left(\frac{\partial E}{\partial b} \right)_{T,\mu}, \qquad (4.11)$$

where

$$b = \boldsymbol{b}^z, \quad \hat{m} = \hat{\boldsymbol{m}}^z, \quad M = \boldsymbol{M}^z,$$

and it is supposed that field $\boldsymbol{b}$ is directed along '$0z$' axis.

As we see in (4.8)–(4.11), the set of $\hat{P}_i(\boldsymbol{k} = 0)$ is not orthogonal, in sense that nondiagonal elements of matrix $(\hat{P}, \hat{P}^+)$ do not vanish. But often it is more convenient to work with orthogonalized set of dynamic variables. Here we use the procedure of orthogonalization, which orthogonalize all variables one by one except of first two $- \hat{n}_1$ and $\hat{n}_2$, so only the block (2×2) of matrix $(\hat{P}, \hat{P}^+)$ which includes correlation functions $(\hat{n}_i, \hat{n}_j)$ will be nondiagonal.

At first we introduce the projection operators:

$$\mathcal{P}_{\hat{n}_1} \ldots = \sum_{i=1}^2 (\ldots, \hat{n}_i)(\hat{n}, \hat{n})_{i,1}^{-1} \cdot \hat{n}_1, \qquad (4.12)$$

$$\mathcal{P}_{\hat{n}_2} \ldots = \sum_{i=1}^2 (\ldots, \hat{n}_i)(\hat{n}, \hat{n})_{i,2}^{-1} \cdot \hat{n}_2, \qquad (4.13)$$

And consider 'projected' magnetization defined by

$$\hat{s} = (1 - \mathcal{P}_{\hat{n}_1} - \mathcal{P}_{\hat{n}_2})\hat{m} \qquad (4.14)$$

It is obvious that $\hat{s}$ is orthogonal to $\hat{n}_1$ and $\hat{n}_2$ in the sense, that

$$(\hat{s}, \hat{n}_1) = (\hat{s}, \hat{n}_2) = 0. \qquad (4.15)$$

Introducing the projection operator

$$\mathcal{P}_{\hat{s}} \ldots = (\ldots, \hat{s})(\hat{s}, \hat{s})^{-1} \hat{s} \qquad (4.16)$$

we can construct so-called 'enthalpy' operator

$$\hat{h} = (1 - \mathcal{P}_{\hat{n}_1} - \mathcal{P}_{\hat{n}_2} - \mathcal{P}_{\hat{s}})\hat{\varepsilon} = (1 - \mathcal{P}_{\hat{n}_1} - \mathcal{P}_{\hat{n}_2} - \mathcal{P}_{\hat{s}})\hat{\omega}, \qquad (4.17)$$

which is orthogonal to all previous operators, i.e.

$$(\hat{h}, \hat{n}_1) = (\hat{h}, \hat{n}_2) = (\hat{h}, \hat{s}) = 0. \qquad (4.18)$$

The momentum operator $\hat{\boldsymbol{p}}(\boldsymbol{k})$ is orthogonal to all variables intrinsically.

We must note, that projection with the help of operators (4.12), (4.13), (4.16) means transition to another ensemble, for example, projection like (4.14) means transition from (μ, V, T, b) to (N, V, T, b) ensemble. Really, magnetic susceptibility in (N, V, T)-ensemble is defined on 'projected' variable $\hat{s}$:

$$\chi_{T,N} = \left(\frac{\partial M}{\partial b} \right)_{N,V,T} = \left(\frac{\partial M}{\partial b} \right)_{\mu,V,T} - \left(\frac{\partial M}{\partial \mu_1} \right) \left(\frac{\partial N_1}{\partial \mu_1} \right)^{-1} \left(\frac{\partial N_1}{\partial b} \right)$$

$$- \left(\frac{\partial M}{\partial \mu_2} \right) \left(\frac{\partial N_2}{\partial \mu_2} \right)^{-1} \left(\frac{\partial N_2}{\partial b} \right) = \qquad (4.19)$$

$$= \beta \Big\{ (\hat{m}, \hat{m}) - (\hat{m}, \hat{n}_1)(\hat{n}_1, \hat{n}_1)^{-1}(\hat{n}_1, \hat{m}) - (\hat{m}, \hat{n}_2)(\hat{n}_2, \hat{n}_2)^{-1}(\hat{n}_2, \hat{m}) \Big\}$$

$$= \beta \left((1 - \mathcal{P}_{\hat{n}_1} - \mathcal{P}_{\hat{n}_2})\hat{m}, \hat{m} \right) = \beta(\hat{s}, \hat{s}).$$

Using (4.6) we can prove one more equality for entropy and arbitrary parameter γ

$$\frac{\partial S}{\partial \gamma} = -\beta \left(\hat{\omega}, \frac{\partial(\beta\hat{\omega})}{\partial \gamma} \right), \qquad (4.20)$$

and for specific heat in (μ, V, T, b)-ensemble we will have:

$$C_{\mu,b} = T\left(\frac{\partial S}{\partial T}\right) = \beta^2(\hat{\omega}, \hat{\omega}) \ .$$

Fulfilling transition to (N, M, V, T)-ensemble like in (4.19), we obtain

$$C_{N,M} = \beta^2\left((1 - \mathcal{P}_{\hat{n}_1} - \mathcal{P}_{\hat{n}_2} - \mathcal{P}_{\hat{s}})\hat{\omega} \ , \ \hat{\omega}\right) = \beta^2(\hat{h}, \hat{h}). \qquad (4.21)$$

For the operator $\hat{\boldsymbol{p}}(\boldsymbol{k})$ we have the equalities

$$\left(\hat{\boldsymbol{p}}^\alpha(\boldsymbol{k}), \hat{\boldsymbol{p}}^\beta(-\boldsymbol{k})\right) = \frac{\delta_{\alpha\beta}}{\beta}(m_1 N_1 + m_2 N_2) = \frac{\delta_{\alpha\beta}}{\beta}\mathcal{M}, \qquad (4.22)$$

$$\left(\hat{\boldsymbol{p}}^\alpha(\boldsymbol{k}), \hat{P}_j(-\boldsymbol{k})\right) = 0 \ , \quad \text{if} \quad \hat{P}_j(\boldsymbol{k}) \neq \hat{\boldsymbol{p}}^\alpha(\boldsymbol{k}), \qquad (4.23)$$

where $\mathcal{M}$ is a mass of our mixture.

Taking into account the relations (4.14), (4.15), (4.17), (4.18), (4.21), (4.22), (4.23), one sees, that the set of variables $\{\hat{n}_1, \hat{n}_2, \hat{\boldsymbol{p}}, \hat{s}, \hat{\varepsilon}\}$ is orthogonalized in the sense discussed above. Generalizing obtained results, we can introduce new set of dynamic variables ($\boldsymbol{k}$-dependent)

$$\hat{Y}(\boldsymbol{k}) = \{\hat{n}_1(\boldsymbol{k}), \hat{n}_2(\boldsymbol{k}), \hat{\boldsymbol{p}}(\boldsymbol{k}), \hat{s}(\boldsymbol{k}), \hat{h}(\boldsymbol{k})\}, \qquad (4.24)$$

which are mutually orthogonal. One exception is for variables $\hat{n}_1$ and $\hat{n}_2$ which are not mutually orthogonal. For $\hat{s}(\boldsymbol{k})$ and $\hat{h}(\boldsymbol{k})$ one has

$$\hat{s}(\boldsymbol{k}) = (1 - \mathcal{P}_{\hat{n}_1}(\boldsymbol{k}) - \mathcal{P}_{\hat{n}_2}(\boldsymbol{k})) \cdot \hat{m}(\boldsymbol{k}), \qquad (4.25)$$

$$\hat{h}(\boldsymbol{k}) = (1 - \mathcal{P}_{\hat{n}_1}(\boldsymbol{k}) - \mathcal{P}_{\hat{n}_2}(\boldsymbol{k}) - \mathcal{P}_{\hat{s}}(\boldsymbol{k})) \cdot \hat{\varepsilon}(\boldsymbol{k})$$
$$= (1 - \mathcal{P}_{\hat{n}_1}(\boldsymbol{k}) - \mathcal{P}_{\hat{n}_2}(\boldsymbol{k}) - \mathcal{P}_{\hat{s}}(\boldsymbol{k})) \cdot \hat{\omega}(\boldsymbol{k}), \qquad (4.26)$$

where

$$\mathcal{P}_{\hat{n}_1(\boldsymbol{k})} \ldots = \sum_{i=1}^{2} \left(\ldots, \hat{n}_i(-\boldsymbol{k})\right) \left(\hat{n}(\boldsymbol{k}), \hat{n}(-\boldsymbol{k})\right)^{-1}_{i,1} \cdot \hat{n}_1(\boldsymbol{k}), \quad (4.27)$$

$$\mathcal{P}_{\hat{n}_2(\boldsymbol{k})} \ldots = \sum_{i=1}^{2} \left(\ldots, \hat{n}_i(-\boldsymbol{k})\right) \left(\hat{n}(\boldsymbol{k}), \hat{n}(-\boldsymbol{k})\right)^{-1}_{i,2} \cdot \hat{n}_2(\boldsymbol{k}), \quad (4.28)$$

$$\mathcal{P}_{\hat{s}(\boldsymbol{k})} \ldots = (\ldots, \hat{s}(-\boldsymbol{k})) \cdot (\hat{s}(\boldsymbol{k}), \hat{s}(-\boldsymbol{k}))^{-1} \cdot \hat{s}(\boldsymbol{k}), \qquad (4.29)$$

The correlation functions of these quantities can be considered as generalization of well-known thermodynamical derivatives (4.7) – (4.11),

(4.19), (4.21), (4.22), for nonzero values of $\boldsymbol{k}$. Hence,

$$(\hat{n}_i(\boldsymbol{k}), \hat{n}_j(-\boldsymbol{k})) = (N_i, N_j)^{\frac{1}{2}} S_{ij}(\boldsymbol{k}), \quad i, j = 1..2, \qquad (4.30)$$

$$(\hat{\boldsymbol{p}}^\alpha(\boldsymbol{k}), \hat{\boldsymbol{p}}^\beta(-\boldsymbol{k})) = \frac{\delta_{\alpha\beta}}{\beta}\mathcal{M}, \qquad (4.31)$$

$$(\hat{s}(\boldsymbol{k}), \hat{s}(-\boldsymbol{k})) = \frac{1}{\beta}\chi_{T,N}(\boldsymbol{k}), \qquad (4.32)$$

$$(\hat{h}(\boldsymbol{k}), \hat{h}(-\boldsymbol{k})) = \frac{1}{\beta^2}C_{N,M}(\boldsymbol{k}), \qquad (4.33)$$

where $S_{ij}(\boldsymbol{k})$, defined in (4.30) are so-called partial structure factors, $\chi_{T,N}(\boldsymbol{k})$ and $C_{N,M}(\boldsymbol{k})$ are the generalized susceptibility and specific heat, respectively.

5. Frequency matrix and matrix of memory functions

Let us mark linear functions of momentums $\{\hat{\boldsymbol{p}}_i, \ i = 1..N_1 + N_2\}$ of particles by symbol $\hat{\pi}^1$, quadratic functions by $\hat{\pi}^2$, etc. One can notice, that, for example, energy $\hat{\varepsilon}(\boldsymbol{k})$ (3.15) is a sum of some quadratic function $\hat{\pi}^2$ and some function of zero power over $\{\hat{\boldsymbol{p}}_i\}$, $\hat{\pi}^0$. So, for our variables we can write:

$$\hat{n}_1(\boldsymbol{k}) \sim \hat{\pi}^0, \quad \hat{n}_2 \sim \hat{\pi}^0, \quad \hat{\boldsymbol{p}}(\boldsymbol{k}) \sim \hat{\pi}^1,$$
$$\hat{s} \sim \hat{\pi}^0, \quad \hat{\varepsilon}(\boldsymbol{k}) \sim (\hat{\pi}^0 + \hat{\pi}^2). \qquad (5.1)$$

It is easy to show that for arbitrary $\hat{\pi}^\alpha$ and distribution (4.3) fulfills:

$$\langle \hat{\pi}^\alpha \rangle = 0, \qquad (5.2)$$

if α is an odd number.

Liouville operator increase the number of α at 1 unit

$$\dot{\hat{n}}_1(\boldsymbol{k}) \sim \hat{\pi}^1, \quad \dot{\hat{n}}_2 \sim \hat{\pi}^1, \quad \dot{\hat{\boldsymbol{p}}}(\boldsymbol{k}) \sim \hat{\pi}^2,$$
$$\dot{\hat{s}} \sim \hat{\pi}^1, \quad \dot{\hat{\varepsilon}}(\boldsymbol{k}) \sim (\hat{\pi}^1 + \hat{\pi}^3). \qquad (5.3)$$

Taking into account (5.1), (5.2), (5.3) one can prove, that:

$$\left(\dot{\hat{Y}}_i(\boldsymbol{k}), \hat{Y}_j(-\boldsymbol{k})\right) = 0, \text{ if } (\hat{Y}_i \neq \hat{\boldsymbol{p}} \text{ and } \hat{Y}_j \neq \hat{\boldsymbol{p}})$$
$$\text{or } (\hat{Y}_i = \hat{\boldsymbol{p}} \text{ and } \hat{Y}_j = \hat{\boldsymbol{p}}). \qquad (5.4)$$

Because of the symmetry conditions

$$\left(\dot{\hat{Y}}_i(\boldsymbol{k}), \hat{Y}_j(-\boldsymbol{k})\right)_{\boldsymbol{k}\to 0} = \left(\hat{Y}_i(\boldsymbol{k}), \dot{\hat{Y}}_j(-\boldsymbol{k})\right)_{\boldsymbol{k}\to 0},$$

for calculation of frequency matrix we must find only correlation functions, which involve momentum:

$$\left(\dot{\hat{\boldsymbol{p}}}^{\alpha}(\boldsymbol{k}), \hat{Y}_j(-\boldsymbol{k})\right) = \mathrm{i}\boldsymbol{k}^{\beta}\left(J_p^{\alpha\beta}(\boldsymbol{k}), \hat{Y}_j(-\boldsymbol{k})\right), \quad \hat{Y}_j \neq \hat{\boldsymbol{p}}. \tag{5.5}$$

For $\boldsymbol{k} \to 0$ one can prove that (see [8])

$$\langle J_p^{\alpha\beta}(\boldsymbol{k})\rangle_{\boldsymbol{k}\to 0} = \left\{ (N_1 + N_2)T - \frac{1}{3}\left\langle \sum_i^{N_1+N_2} \boldsymbol{r}_i \frac{\partial U(\boldsymbol{r}^N)}{\partial \boldsymbol{r}_i}\right\rangle \right\}, \tag{5.6}$$

where $U(\boldsymbol{r}^N)$ is the total potential energy:

$$U(\boldsymbol{r}^N) = \hat{\varepsilon} - \sum_i^{(1)} \frac{\boldsymbol{p}_i^{(1)\,2}}{2m_1} - \sum_i^{(2)} \frac{\boldsymbol{p}_i^{(2)\,2}}{2m_2}.$$

Let us find now pressure of the system, which follows from the equilibrium treatment

$$P = -\left(\frac{\partial \Omega}{\partial V}\right)_{N,V,T}, \tag{5.7}$$

where Ω is the thermodynamical potential (4.5). Imposing Ω to be V-dependent by substitution $\boldsymbol{r} = \boldsymbol{R}V^{\frac{1}{3}}$ in potentials $V^{(11)}$, $V^{(12)}$, $V^{(22)}$, J, we can get, after some simplifications

$$P = T\frac{N_1 + N_2}{V} - \frac{1}{3V}\left\langle \sum_i^{N_1+N_2} \boldsymbol{r}_i \frac{\partial U(\boldsymbol{r}^N)}{\partial \boldsymbol{r}_i}\right\rangle. \tag{5.8}$$

Comparing (5.6) and (5.8), we obtain

$$\langle J_p^{\alpha\beta}(\boldsymbol{k})\rangle = \delta_{\alpha\beta}PV = -\delta_{\alpha\beta}\Omega. \tag{5.9}$$

Results (5.9) show that in system with 'isotropic' potential energy (which is a function of $|\boldsymbol{r}|$, and does not depend on its direction) in homogeneous external field $\left(\frac{\partial \hat{\boldsymbol{B}}(\boldsymbol{r},t)}{\partial \boldsymbol{r}} = 0\right)$, the pressure, defined from equilibrium treatment can be expressed through the average of the stress tensor.

With the help of (4.6) now we get

$$(\dot{\hat{\boldsymbol{p}}}^{\alpha}(\boldsymbol{k}), \hat{n}_1(-\boldsymbol{k}))_{\boldsymbol{k}\to 0} = \mathrm{i}\boldsymbol{k}^{\beta}\left(J_p^{\alpha\beta}, \hat{n}_1\right)$$
$$= \frac{\mathrm{i}\boldsymbol{k}^{\beta}}{\beta}\frac{\partial \langle J_p^{\alpha\beta}\rangle}{\partial \mu_1} = \frac{\mathrm{i}\boldsymbol{k}^{\alpha}}{\beta}\frac{\partial \Omega}{\partial \mu_1} = \frac{\mathrm{i}\boldsymbol{k}^{\alpha}}{\beta}N_1. \tag{5.10}$$

Let us introduce now scalar longitudinal momentum $\hat{p}(\boldsymbol{k})$, as a component of vector momentum $\hat{\boldsymbol{p}}(\boldsymbol{k})$, oriented along the vector $\boldsymbol{k}$. From the equation (5.10) we conclude, that in isotropic system only longitudinal components make contribution to the matrix of memory functions, correlation functions of transverse components are equal to zero. In the same way as (5.10) was obtained we can obtain other correlation functions with the longitudinal current $\hat{p}\boldsymbol{k}$ in the limit $k \to 0$, $(k = |\boldsymbol{k}|)$

$$(\dot{\hat{p}}, \hat{n}_i) = \frac{\mathrm{i}k}{\beta}N_i, \tag{5.11}$$

$$(\dot{\hat{p}}, \hat{m}) = \frac{\mathrm{i}k}{\beta}V\left(\frac{\partial P}{\partial b}\right)_{\mu,V,T}, \quad (\dot{\hat{p}}, \hat{\omega}) = \frac{\mathrm{i}k}{\beta^2}V\left(\frac{\partial P}{\partial T}\right)_{\mu,V,b}, \tag{5.12}$$

or for our set of orthogonalized variables

$$(\dot{\hat{p}}, \hat{s}) = \frac{\mathrm{i}k}{\beta}V\left(\frac{\partial P}{\partial b}\right)_{N,V,T}, \quad (\dot{\hat{p}}, \hat{h}) = \frac{\mathrm{i}k}{\beta^2}V\left(\frac{\partial P}{\partial T}\right)_{N,V,M}. \tag{5.13}$$

Using Gibbs-Duhem equation

$$S\,dT + N_1\,d\mu_1 + N_2\,d\mu_2 + M\,db - V\,dp = 0 \tag{5.14}$$

one can write the right-hand sides of equation (5.13) as follows

$$\left(\frac{\partial P}{\partial b}\right)_{N,V,T} = \frac{\nu_p}{\kappa_T}, \tag{5.15}$$

$$\left(\frac{\partial P}{\partial T}\right)_{N,V,M} = \frac{\alpha_p}{-\frac{1}{V}\left(\frac{\partial V}{\partial P}\right)_{T,N,M}}, \tag{5.16}$$

where

$$\nu_p = \frac{1}{V}\left(\frac{\partial V}{\partial b}\right)_{P,N,T}, \tag{5.17}$$

$$\kappa_T = -\frac{1}{V}\left(\frac{\partial V}{\partial P}\right)_{T,N,b}, \tag{5.18}$$

$$\alpha_p = \frac{1}{V}\left(\frac{\partial V}{\partial T}\right)_{P,N,M} \tag{5.19}$$

are the coefficients of magnetostriction, isothermal compressibility and isobaric thermal expansion, respectively. The value $(\partial V/\partial P)$ in equation

(5.16) is defined at constant magnetization M. After some algebra with the help of Gibbs-Duhem and Maxwell relations, we get

$$\left(\frac{\partial V}{\partial P}\right)_{T,N,M} = -V\kappa_T + \frac{V^2\nu_p^2}{\chi + \frac{\nu_p^2 V}{\kappa_T}},$$

and

$$\left(\frac{\partial P}{\partial T}\right)_{V,N,M} = \frac{\alpha_p}{\kappa_T}\left(1 + \frac{V\nu_p^2}{\chi\,\kappa_T}\right). \tag{5.20}$$

Now making generalization for $\boldsymbol{k} \neq 0$ we can write some elements of matrix $\mathrm{i}\Omega$ as follows

$$\mathrm{i}\Omega_{p,s}(\boldsymbol{k}) = \mathrm{i}kV\frac{\nu_p(\boldsymbol{k})}{\kappa_T(\boldsymbol{k})\chi(\boldsymbol{k})}, \quad \mathrm{i}\Omega_{s,p}(\boldsymbol{k}) = \mathrm{i}kV\frac{\nu_p(\boldsymbol{k})}{\kappa_T(\boldsymbol{k})\mathcal{M}}, \tag{5.21}$$

$$\mathrm{i}\Omega_{p,h}(\boldsymbol{k}) = \mathrm{i}kV\frac{\alpha_p(\boldsymbol{k})}{C_{NMV}(\boldsymbol{k})\kappa_T(\boldsymbol{k})}\left(1 + \frac{V\nu_p^2(\boldsymbol{k})}{\chi(\boldsymbol{k})\kappa_T(\boldsymbol{k})}\right), \tag{5.22}$$

$$\mathrm{i}\Omega_{h,p}(\boldsymbol{k}) = \mathrm{i}k\frac{V}{\beta}\frac{\alpha_p(\boldsymbol{k})}{\mathcal{M}\kappa_T(\boldsymbol{k})}\left(1 + \frac{V\nu_p^2(\boldsymbol{k})}{\chi(\boldsymbol{k})\kappa_T(\boldsymbol{k})}\right). \tag{5.23}$$

Elements, involving densities of particle number, as follows from (5.11) and (2.20), read:

$$\mathrm{i}\Omega_{p,n_i} = \frac{\mathrm{i}k}{\beta}\sum_j N_j\left(\hat{n}(\boldsymbol{k}),\hat{n}^+(-\boldsymbol{k})\right)_{ij}^{-1}, \tag{5.24}$$

$$\mathrm{i}\Omega_{n_i,p} = \mathrm{i}k\frac{N_i}{\mathcal{M}}, \tag{5.25}$$

where $(\hat{n}_j(\boldsymbol{k}),\hat{n}_i(-\boldsymbol{k}))_{ij}^{-1}$ means $\{ij\}$-element of matrix (2×2), which is inverse to the block $(\hat{n},\hat{n}^+)$ of the matrix of static correlation functions $(\hat{P},\hat{P}^+)$. Matrix $(\hat{n},\hat{n}^+)$ in the limit $k \to 0$ consists of functions $\frac{1}{\beta}\frac{\partial N_i}{\partial \mu_j}$ (see (4.13)), therefore

$$\left(\hat{n}(\boldsymbol{k}),\hat{n}^+(-\boldsymbol{k})\right)_{ij}^{-1} = \beta\left(\frac{\partial \mu_i}{\partial N_j}\right)_{VTbN_{\bar{j}}}, \tag{5.26}$$

where $N_{\bar{j}}$ denotes set of all N_k, except of N_j.

For the right-hand sides of (5.26) we can make transition to the ensemble with constant pressure

$$\left(\frac{\partial \mu_i}{\partial N_j}\right)_{VTbN_{\bar{j}}} = \left(\frac{\partial \mu_i}{\partial N_j}\right)_{VTbN_{\bar{j}}} + \frac{v_i v_j}{V\kappa_T}, \tag{5.27}$$

where

$$v_i = \left(\frac{\partial V}{\partial N_i}\right)_{TPbN_{\bar{i}}} \tag{5.28}$$

is the partial molar volume per molecule of species 'i', which possesses the property

$$\sum_i N_i v_i = V. \tag{5.29}$$

Using Gibbs-Duhem equation, we find for isobaric processes

$$\sum_i N_i\left(\frac{\partial \mu_i}{\partial N_j}\right)_{VTbN_{\bar{j}}} = 0. \tag{5.30}$$

Now, with the help of (5.29), (5.30) we can rewrite the elements (5.24) of $\mathrm{i}\Omega$

$$\mathrm{i}\Omega_{p,n_1}(\boldsymbol{k}) = \mathrm{i}k\frac{v_1(\boldsymbol{k})}{\kappa_T(\boldsymbol{k})}, \qquad \mathrm{i}\Omega_{p,n_2}(\boldsymbol{k}) = \mathrm{i}k\frac{v_2(\boldsymbol{k})}{\kappa_T(\boldsymbol{k})}. \tag{5.31}$$

For our case of weak nonequilibrium, matrix of memory functions (2.10), which Laplace transformation is given in (2.21), reads:

$$\phi(\boldsymbol{k},t) = \left((1-\mathcal{P})\dot{\hat{Y}}_i(\boldsymbol{k}), e^{-(1-\mathcal{P})\mathrm{i}\hat{L}t}(1-\mathcal{P})\dot{\hat{Y}}_j(-\boldsymbol{k})\right)$$
$$\times \left(\hat{Y}(\boldsymbol{k}),\hat{Y}(-\boldsymbol{k})^+\right)^{-1}, \tag{5.32}$$

where

$$\mathcal{P}(\boldsymbol{k}) = \sum_{i,j}(\ldots,\hat{n}_i)(\hat{n},\hat{n})_{i,j}^{-1}\cdot\hat{n}_j$$
$$+ (\ldots,\hat{p})\frac{1}{(\hat{p},\hat{p})}\hat{p} + (\ldots,\hat{s})\frac{1}{(\hat{s},\hat{s})}\hat{s} + \left(\ldots,\hat{h}\right)\frac{1}{(\hat{h},\hat{h})}\hat{h}, \tag{5.33}$$

here dependence on $\boldsymbol{k}$ in the right-hand side is omitted for simplicity. For the generalized fluxes

$$I_i(\boldsymbol{k}) = (1-\mathcal{P})\dot{\hat{Y}}_i(\boldsymbol{k}) = \dot{\hat{Y}}_i(\boldsymbol{k}) - \sum_j \mathrm{i}\Omega_{ij}\hat{Y}_j, \tag{5.34}$$

in the limit $k \to 0$ one has

$$I_i(\boldsymbol{k}) \simeq \mathrm{i}k f_i, \tag{5.35}$$

where f_i are regular longitudinal parts of fluxes. The elements of matrix $\phi(\boldsymbol{k},t)$ (5.32) are proportional to k^2.

One can prove that due to time-reversal symmetry, the structure of matrix $\phi(\boldsymbol{k}, t)$ in the limit $\boldsymbol{k} \to 0$, $\omega \to 0$ is opposite to that of $\mathrm{i}\Omega$:

$$\mathrm{i}\Omega = \mathrm{i}k \begin{pmatrix} 0 & 0 & \omega_{n_1,p} & 0 & 0 \\ 0 & 0 & \omega_{n_2,p} & 0 & 0 \\ \omega_{p,n_1} & \omega_{p,n_2} & 0 & \omega_{p,s} & \omega_{p,\varepsilon} \\ 0 & 0 & \omega_{s,p} & 0 & 0 \\ 0 & 0 & \omega_{\varepsilon,p} & 0 & 0 \end{pmatrix}, \tag{5.36}$$

$$\phi = -k^2 \begin{pmatrix} \varphi_{n_1,n_1} & \varphi_{n_1,n_2} & 0 & \varphi_{n_1,s} & \varphi_{n_1,h} \\ \varphi_{n_2,n_1} & \varphi_{n_2,n_2} & 0 & \varphi_{n_2,s} & \varphi_{n_2,h} \\ 0 & 0 & \varphi_{p,p} & 0 & 0 \\ \varphi_{s,n_1} & \varphi_{s,n_2} & 0 & \varphi_{s,s} & \varphi_{s,h} \\ \varphi_{h,n_1} & \varphi_{h,n_2} & 0 & \varphi_{h,s} & \varphi_{h,h} \end{pmatrix}, \tag{5.37}$$

because the elements, which involve momentum (except of $\phi_{p,p}$) can be neglected because they are proportional to higher powers of k. Thus, in the hydrodynamic limit, the Laplace transforms can be written as follows

$$\phi_{ij} = -k^2 \frac{V}{\beta} \sum_j L_{ik} \cdot \left(\hat{Y}, \hat{Y}^+ \right)^{-1}_{kj}, \tag{5.38}$$

where L_{ij} are the kinetic coefficients, defined by Green-Kubo - like formulas

$$L_{ij} = \frac{\beta}{V} \left(f_i, \frac{1}{\varepsilon + \mathrm{i}\hat{L}} f_j \right) = \frac{\beta}{V} \int\limits_0^\infty \left(f_i, e^{-\mathrm{i}\hat{L}t} f_j \right) dt. \tag{5.39}$$

6. Hydrodynamic collective modes

Collective modes' spectrum can be found from the equation (2.25):

$$\det |z - \mathrm{i}\Omega + \phi| = 0, \tag{6.1}$$

as the spectrum of a matrix:

$$\hat{T} = \mathrm{i}\Omega - \phi. \tag{6.2}$$

As $\delta \equiv \mathrm{i}k$ is a small parameter, $\mathrm{i}\Omega \sim \delta$, $\phi \sim \delta^2$, in the first approximation we can neglect the matrix ϕ. Defining 2 matrices:

$$\hat{\omega} = \mathrm{i}\Omega/\delta, \quad \hat{\varphi} = \phi/\delta^2, \tag{6.3}$$

we can find in first approximation 2 nonzero propagation modes:

$$z_{1,2} = \pm \mathrm{i}k v_s, \tag{6.4}$$

where

$$v_s = \sqrt{\omega_{p,n_1}\omega_{n_1,p} + \omega_{p,n_2}\omega_{n_2,p} + \omega_{p,s}\omega_{s,p} + \omega_{p,h}\omega_{h,p}} \tag{6.5}$$

is the sound velocity. Three vanishing eigenvalues $z_{3,4,5} = 0$ determine dissipative modes. After simplification and transformation of (6.5) with the help of Maxwell relations we come to simple result:

$$v_s^2 = -\frac{V^2}{\mathcal{M}} \left(\frac{\partial P}{\partial V} \right)_{NSM} = \left(\frac{\partial P}{\partial \rho} \right)_{NSM}, \tag{6.6}$$

where the mass density $\rho = \frac{\mathcal{M}}{V}$ was introduced.

To find the next approximation we consider z in (6.1) as a series over δ:

$$z = \delta z^0 + \delta^2 D + \dots \quad . \tag{6.7}$$

Putting (6.7) into (6.1) and equating to zero coefficients near δ, give us desirable values D. In such a way we will have for propagating sound modes

$$z = \pm \mathrm{i}k v_s - k^2 D_s, \tag{6.8}$$

and for purely damping hydrodynamic modes

$$z_i = -k^2 D_i, \quad i = 3, 4, 5 , \tag{6.9}$$

where D_s, $D_{3,4,5}$ are corresponding damping coefficients. For sound modes one gets

$$D_s = \frac{1}{2v_s^2} \mathrm{Sp}(\hat{\varphi} \cdot \hat{\omega}^2) = \frac{\mathrm{Sp}(\hat{\varphi} \cdot \hat{\omega}^2)}{\mathrm{Sp}(\hat{\omega}^2)}. \tag{6.10}$$

Damping coefficients for the purely diffusive modes can be found as the roots of an algebraic equation of third order

$$D^3 p_3 + D^2 p_2 + D^1 p_1 + p_0 = 0, \tag{6.11}$$

with coefficients p_i are defined via the elements of matrix

$$\hat{æ} \equiv \hat{\omega} + \hat{\varphi}, \quad æ_{p,p} \equiv 0$$

as some minors, which can be presented with the help of the operator $\frac{\partial}{\partial æ_{n_1 n_1}} + \frac{\partial}{\partial æ_{n_2 n_2}} + \frac{\partial}{\partial æ_{ss}} + \frac{\partial}{\partial æ_{\varepsilon\varepsilon}}$ in the following form:

$$p_i = \left(\frac{\partial}{\partial æ_{n_1 n_1}} + \frac{\partial}{\partial æ_{n_2 n_2}} + \frac{\partial}{\partial æ_{ss}} + \frac{\partial}{\partial æ_{\varepsilon\varepsilon}} \right)^i \cdot \det \hat{æ} \tag{6.12}$$

7. Calculation of time correlation functions

Equation for time-dependent correlation functions (2.23) can be written as follows

$$\hat{A}(z)\cdot\hat{F}^z = \hat{F}^0, \qquad (7.1)$$

where $\hat{A}(z) = z\cdot\hat{1} - \hat{T}$, and $\hat{T}$ is defined in (6.2); $\hat{F}^z = (\hat{Y}, \hat{Y}^+)^z$ and $\hat{F}^0$ are the matrix of Laplace transforms of time correlation functions and a matrix of static correlation functions. The formal solution of (7.1) is given by

$$\hat{F}^z = \hat{A}^{-1}(z)\cdot\hat{F}^0, \qquad (7.2)$$

where $\hat{A}^{-1}(z) = (z\hat{1} - \hat{T})^{-1}$ is the matrix inverse to $\hat{A}$. Its the ij-th element $(\hat{A}^{-1})_{ij}$ can be written as an algebraical adjunct $(Ad(A_{ij}))$ of element A_{ij}, divided on determinant $\Delta(\hat{A})$

$$(\hat{A}^{-1})_{ij} = \frac{Ad(A_{ij})}{\Delta(\hat{A})}. \qquad (7.3)$$

For $Ad(A_{ij})$ one has

$$Ad(A_{ij}) = \frac{\partial\Delta(\hat{A})}{\partial A_{ij}}, \qquad (7.4)$$

Where we omit the dependence on z for the sake of simplicity. Determinant of matrix $\hat{T}$ can be presented as a product of its eigenvalues $\{z_i\}$

$$\Delta(\hat{T}) = \prod_{i=1}^{5} z_i,$$

that's why

$$\Delta(\hat{A}) = \prod_{i=1}^{5} (z - z_i), \qquad (7.5)$$

where z_i were found in the previous section up to the second power over the the small parameter δ (see $(6.7) - (6.9)$).

Matrices $\hat{\omega}$ and $\hat{\varphi}$ have the opposite structure (see (5.36)), so that we can write

$$\frac{\partial}{\partial A_{ij}} = -\frac{\partial}{\partial i\Omega_{ij}} - \frac{\partial}{\partial\phi_{ij}} = -\frac{1}{\delta}\frac{\partial}{\partial\omega_{ij}} - \frac{1}{\delta^2}\frac{\partial}{\partial\varphi_{ij}}. \qquad (7.6)$$

Therefor from (7.4) - (7.6) we have

$$Ad(A_{ij}) = -\sum_k \frac{\Delta}{z - z_k}\frac{\partial z_k}{\partial A_{ij}}$$

$$= \Delta\sum_k \frac{1}{z - z_k}\cdot\left(\frac{1}{\delta}\frac{\partial z_k}{\partial\omega_{ij}} + \frac{1}{\delta^2}\frac{\partial z_k}{\partial\varphi_{ij}}\right), \qquad (7.7)$$

and $(\hat{A}^{-1})_{ij}$ now read (7.3)

$$(\hat{A}^{-1}(z))_{ij} = \sum_k \frac{1}{z - z_k}G_{ij}^k, \qquad (7.8)$$

where

$$G_{ij}^k = \frac{1}{\delta}\frac{\partial z_k}{\partial\omega_{ij}} + \frac{1}{\delta^2}\frac{\partial z_k}{\partial\varphi_{ij}} \qquad (7.9)$$

are so-called weight coefficients describing the contribution from corresponding collective modes. Because of $\{z_k\}$ are series with respect to δ (see $(6.7) - (6.9)$), the first approximations of G_{ij}^k can be found as:

$$G_{ij}^{(0)k} = \frac{\partial z_k^0}{\partial\omega_{ij}} + \frac{\partial z_k^1}{\partial\varphi_{ij}}. \qquad (7.10)$$

We can also represent $\hat{G}^{(0)k}$ in another form, which is mostly used in literature. Let $\hat{X}$ be a matrix of eigenvectors of $\hat{T}$, which corresponds to eigenvalues $\{z_k\}$:

$$\hat{T}\cdot\hat{X} = \hat{X}\cdot\hat{Z},$$

$\hat{Z} = \|z_i\delta_{ij}\|$ is the diagonal matrix of eigenvalues. It is easy to show that matrix $\hat{A}(z)$ have the same eigenvectors $\hat{X}$, with eigenvalues $\{z - z_k\}$, therefore:

$$\hat{A}^{-1} = \left(\hat{A}\cdot\hat{X}\cdot\hat{X}^{-1}\right)^{-1} = \left(\hat{X}\cdot(z\hat{1} - \hat{Z})\cdot\hat{X}^{-1}\right)^{-1}$$
$$= \hat{X}\cdot(z\hat{1} - \hat{Z})^{-1}\cdot\hat{X}^{-1}. \qquad (7.11)$$

Because the matrix of eigenvalues $(z\hat{1} - \hat{Z})$ of $\hat{A}(z)$ is diagonal, we can rewrite (7.11) in the form:

$$(\hat{A}^{-1})_{ij} = \sum_k \frac{X_{ik}(\hat{X}^{-1})_{kj}}{z - z_k}. \qquad (7.12)$$

Comparing (7.7) and (7.12), we conclude, that

$$G_{ij}^{(0)k} = X_{ik}^0(\hat{X}^{0\,-1})_{kj}. \qquad (7.13)$$

For nondegenerated index k ($k = 1, 2$), for which (7.13) can be easily calculated, results coincide with that ones obtained in (7.10), but for $k = 3, 4, 5$ calculation of (7.13) is more complicate. Therefore the expression (7.10) can be more convenient for some applications.

8. Discussion

In this paper the spectrum of collective hydrodynamic modes of a binary ferromagnetic mixture has been studied on the base of rigorous statistical treatment. We have found two complex-conjugated collective modes, which are responsible for the propagating of sound. Other modes are purely diffusive and describe processes connected with temperature, mass and magnetization fluctuations. The obtained result are valid for an arbitrary mixture under the same external conditions (homogeneity of the external magnetic field, isotropic interactions) within the hydrodynamical region. The square of the sound velocity (6.6) appears to be inversely proportional to the adiabatic compressibility in ensemble with constant magnetization and particle number, that coincide with the results for one-component magnetic system [8]. Damping coefficients for sound modes has been found (see (6.10)), and damping coefficients of other diffusive modes can be obtained from equation (6.11).

For the paramagnetic case and $b = 0$ the damping coefficient of spin mode can be easily found from equation (6.11)

$$D_m = \varphi_{ss}. \tag{8.1}$$

It can be shown that for this case the expressions for time correlation functions are formally the same as they are for a binary system of two simple liquids [16], and the Laplace transforms of "spin density–spin density" time correlation function can be written as

$$(\hat{s}, \hat{s})^z = \frac{T \cdot \chi_{TNV}(\boldsymbol{k})}{(z + k^2 \varphi_{ss})}. \tag{8.2}$$

For $b \neq 0$ the expressions for time correlation functions have more complicated form because of additional coupling between spin and translational degrees of freedoms. This problem will be considered elsewhere.

Appendix

It is easy to show that

$$\dot{\hat{n}}_1(\boldsymbol{k}) = \mathrm{i}\boldsymbol{k} \cdot \frac{\boldsymbol{p}^{(1)}(\boldsymbol{k})}{m_1}, \quad \dot{\hat{n}}_2(\boldsymbol{k}) = \mathrm{i}\boldsymbol{k} \cdot \frac{\boldsymbol{p}^{(2)}(\boldsymbol{k})}{m_2} \tag{A.1}$$

where $\boldsymbol{p}^{(\nu)}(\boldsymbol{k}) = \sum_j^{N_\nu} \boldsymbol{p}_j^{(\nu)} e^{\mathrm{i}\boldsymbol{k}\boldsymbol{r}_j}$, so that

$$J_{n_1}^\alpha = \frac{\boldsymbol{p}^{(1)\,\alpha}(\boldsymbol{k})}{m_1}, \quad J_{n_2}^\alpha = \frac{\boldsymbol{p}^{(2)\,\alpha}(\boldsymbol{k})}{m_2}, \quad R_{n_1} = 0 \tag{A.2}$$

Let us introduce now vector function $\boldsymbol{\Gamma_k}(\boldsymbol{r})$, cartesian coordinates of which are given by:

$$\Gamma_{\boldsymbol{k}}^\alpha(\boldsymbol{r}) = r^\alpha \frac{1 - e^{-\mathrm{i}\boldsymbol{k}\boldsymbol{r}}}{\mathrm{i}\boldsymbol{k}\boldsymbol{r}}. \tag{A.3}$$

Having calculated $\dot{p}^\alpha = \mathrm{i}\hat{L}\, p^\alpha$, we may write the stress tensor $J_p^{\alpha\beta}$ and term $R_p^\alpha(\boldsymbol{k})$ as follows:

$$J_p^{\alpha\beta}(\boldsymbol{k}) = J_{p,1}^{\alpha\beta}(\boldsymbol{k}) + J_{p,2}^{\alpha\beta}(\boldsymbol{k}) + J_{p,s}^{\alpha\beta}(\boldsymbol{k}) + J_{p,int}^{\alpha\beta}(\boldsymbol{k}) \tag{A.4}$$

$$R_p^\alpha(\boldsymbol{k}) = \sum_i^{(2)} \frac{\partial \left(\hat{\boldsymbol{B}}(\boldsymbol{r}_i), \hat{s}_i \right)}{\partial r_i^\alpha} e^{\mathrm{i}\boldsymbol{k}\boldsymbol{r}_i} \tag{A.5}$$

where

$$J_{p,1}^{\alpha\beta}(\boldsymbol{k}) = \sum_i^{(1)} \frac{\boldsymbol{p}_i^{(1)\beta} \boldsymbol{p}_i^{(1)\,\alpha}}{m_1} e^{\mathrm{i}\boldsymbol{k}\boldsymbol{r}_i}$$

$$- \frac{1}{2} \sum_{i \neq j}^{(1,1)} V^{(11)\prime}(\boldsymbol{r}_{ij}) \frac{r_{ij}^\alpha}{|\boldsymbol{r}_{ij}|} \Gamma_{\boldsymbol{k}}^\beta(\boldsymbol{r}_{ij}) e^{\mathrm{i}\boldsymbol{k}\boldsymbol{r}_i}, \tag{A.6}$$

$V^{(11)\prime}(\boldsymbol{r}_{ij})$ is a derivative of $V^{(11)}$ over its argument $|\boldsymbol{r}_{ij}|$; $J_{p,2}^{\alpha\beta}$ is the same as $J_{p,1}^{\alpha\beta}$ (implying the substitution of indices '1'→'2'),

$$J_{p,s}^{\alpha\beta} = \frac{1}{2} \sum_{i \neq j}^{(1,1)} J'(\boldsymbol{r}_{ij})(\hat{s}_i, \hat{s}_j) \frac{r_{ij}^\alpha}{|\boldsymbol{r}_{ij}|} \Gamma_{\boldsymbol{k}}^\beta(\boldsymbol{r}_{ij}) e^{\mathrm{i}\boldsymbol{k}\boldsymbol{r}_i}, \tag{A.7}$$

and

$$J_{p,int}^{\alpha\beta} = - \sum_{i,j}^{(1,2)} V^{(12)\prime}(\boldsymbol{r}_{ij}) \frac{r_{ij}^\alpha}{|\boldsymbol{r}_{ij}|} \Gamma_{\boldsymbol{k}}^\beta(\boldsymbol{r}_{ij}) e^{\mathrm{i}\boldsymbol{k}\boldsymbol{r}_i}. \tag{A.8}$$

The magnetization currents:

$$J_m^{\alpha\beta} = \sum_i^{(1)} \frac{\boldsymbol{p}_i^{(1)\beta}}{m_2} s_i^\alpha e^{\mathrm{i}\boldsymbol{k}\boldsymbol{r}_i} + \frac{1}{2} \sum_{i \neq j}^{(1,2)} J(\boldsymbol{r}_{ij}) \left[\hat{s}_i \times \hat{s}_j \right]^\alpha \Gamma_{\boldsymbol{k}}^\beta(\boldsymbol{r}_{ij}) e^{\mathrm{i}\boldsymbol{k}\boldsymbol{r}_i} \tag{A.9}$$

$$R_m^\alpha = \sum_i^{(1)} \left[\hat{s}_i \times \hat{\boldsymbol{B}}(\boldsymbol{r}_i; t) \right]^\alpha e^{\mathrm{i}\boldsymbol{k}\boldsymbol{r}_j}. \tag{A.10}$$

The energy currents:

$$J_\varepsilon^\alpha = J_{\varepsilon,1}^\alpha + J_{\varepsilon,2}^\alpha + J_{\varepsilon,ss}^\alpha + J_{\varepsilon,int}^\alpha, \tag{A.11}$$

$$R_\varepsilon = \sum_j^{(2)} \frac{\partial \left(\hat{\boldsymbol{B}}(\boldsymbol{r}_j)\hat{s}_j \right)}{\partial \boldsymbol{r}_j} \frac{\boldsymbol{p}_j^{(2)}}{m_2} e^{i\boldsymbol{k}\boldsymbol{r}_j}$$

$$+ \frac{1}{2} \sum_{i \neq j}^{(2,2)} J(\boldsymbol{r}_{ij})(\hat{\boldsymbol{B}}_i, [\hat{s}_j, \hat{s}_i]) \left(e^{i\boldsymbol{k}\boldsymbol{r}_i} + e^{i\boldsymbol{k}\boldsymbol{r}_j} \right) \tag{A.12}$$

where

$$J_{\varepsilon,1}^\alpha = \sum_i^{(1)} \left\{ \frac{\boldsymbol{p}_i^{(1)\alpha}}{m_1} \hat{\varepsilon}_i^{(1)} - \frac{1}{2} \sum_{j(\neq i)}^{1} V^{(11)\prime}(\boldsymbol{r}_{ij}) \frac{(\boldsymbol{r}_{ij}\boldsymbol{p}_i^{(1)})}{|\boldsymbol{r}_{ij}|m_1} \Gamma_{\boldsymbol{k}}^\alpha(\boldsymbol{r}_{ij}) \right\} e^{i\boldsymbol{k}\boldsymbol{r}_i}, \tag{A.13}$$

$J_{\varepsilon,2}^\alpha$ is the same as $J_{\varepsilon,1}^\alpha$ after substitution '1'→'2' and implying difference between $\hat{\varepsilon}_i^{(1)}$ and $\hat{\varepsilon}_i^{(2)}$;

$$J_{\varepsilon,ss}^\alpha = \frac{1}{2} \sum_i^{1} \left\{ J'(\boldsymbol{r}_{ij})(\hat{s}_i\hat{s}_j) \frac{(\boldsymbol{r}_{ij}\boldsymbol{p}_i^{(2)})}{|\boldsymbol{r}_{ij}|m_2} \Gamma_{\boldsymbol{k}}^\alpha(\boldsymbol{r}_{ij}) - \tag{A.14} \right.$$

$$\left. - \sum_{j \neq l(\neq i)}^{(2,2)} J(\boldsymbol{r}_{ij}) J(\boldsymbol{r}_{il})(\hat{s}_i, [\hat{s}_j, \hat{s}_l]) \Gamma_{\boldsymbol{k}}^\alpha(\boldsymbol{r}_{ij}) \right\} e^{i\boldsymbol{k}\boldsymbol{r}_i}$$

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До статистичної гідродинаміки бінарної суміші магнітних та немагнітних атомів