Non-Linear Response Theory and Hydrodynamics of Charged and Neutral Media

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INTRODUCTION

The basis of the research in the first part of the book (Chapters I-IV) is the response theory, which studies the reaction of a charged or neutral medium with strong interparticle interaction (non-ideal continuous medium) to electric and electromagnetic fields, gradients of mass velocity, temperature, chemical potentials of chemical elements (concentrations of chemical elements or components) forming the medium, etc. This part of the book is mainly devoted to the quadratic version of nonlinear response theory. The need to develop a theory of nonlinear response for non-ideal media arose due to the obviously insufficient amount of research in this area. For the first time, the book systematically discusses approaches to the study of the nonlinear response of non-ideal media to various perturbations

For non-ideal media in which the potential energy of the interaction of particles is comparable to their kinetic energy, the application of the response theory (RT) is correct (in comparison with traditional kinetic equations, for example, the Boltzmann equation, etc., which are not applicable for describing non-ideal media) [1-3]. Limitations of RT are related to the intensity of disturbing influences, which should not significantly change the state of the medium. The RT theory can be formulated with respect to both mechanical and thermal disturbances. For a medium under the action of mechanical disturbances, for example, external electric or electromagnetic fields, the Hamiltonian of the system is the sum of the undisturbed Hamiltonian of the medium and the Hamiltonian of the interaction of the medium with the external fields. Thermal perturbations of the medium (gradients of mass velocity, temperature, etc.) do not allow such a description, and in this case, an approach using generalized Langevin equations is proposed to determine the reaction of a non-ideal medium [3, 4].

The linear RT is quite well developed (see, for example, [1- 3[a]]). On its basis, theoretical and computational studies of linear thermophysical properties (in particular, by computer modeling methods) have been carried out of non-ideal charged and neutral systems, and characteristics of linear interaction of electromagnetic waves with non-ideal charged media (see, for example, [3[a], 5, 6]). In addition, there is a significant array of experimental results on the linear characteristics of non-ideal media, including non-ideal plasma (see, for example, [3[a], 6]).

At the same time, the theory of nonlinear response (NRT) for charged and neutral non-ideal media is not sufficiently developed. There are a number of results on NRT for mechanical disturbances: the nonlinear fluctuation-dissipation theorem for response functions and corresponding correlation functions, as well as on the frequency moments of these functions for charged media [7-9]. Theoretical approaches to the description of nonlinear phenomena and thermophysical characteristics of non-ideal charged media based on NRT have been discussed in recent papers [3[b,c], 10]. NRT is applied there to describe the nonlinear interaction of longitudinal waves of an electric field in dense charge media, i.e. phenomena such as plasma echo and wave transformation [10[abc]]. To verify the theoretical results, it is of interest to conduct studies of the corresponding nonlinear characteristics of non-ideal Coulomb systems by computer modeling methods, since the necessary experimental data are not available. In this regard, there is a problem of studying the conditions of experimental realization of nonlinear phenomena in non-ideal charged media and methods of measuring nonlinear characteristics.

Let us explain the role of the response theory in the study of thermophysical properties (including linear) of non-ideal media (i.e. thermodynamic, transport and rheological, and optical characteristics). Traditionally, there is an inconsistency of approaches in the computational and theoretical study of the thermophysical characteristics of non-ideal media, especially when using various model approximations (see, for example, [6, 10[c]]). Therefore, there is a need for a unifying theory that can be the basis for studying of the thermophysical characteristics of non-ideal media. Such a theory is the response theory. With the help of RT, it is possible to analyze correctly the general relations between the thermophysical characteristics. The formulation of general relations is possible because this theory does not operate directly with the thermophysical characteristics of the medium, but with response functions or with space-time correlation functions. In turn, the response functions determine the thermophysical characteristics of the medium in various ways (for more information, see Chapters I-III). In connection with transport (and thermodynamic) properties, the study of (linear and nonlinear) response functions of non-ideal media to thermal disturbances is of particular interest. We also note that it is advisable to use phenomenological methods of nonequilibrium thermodynamics to analyze and verify the results on the thermophysical properties of non-ideal media (Chapter IV), which are used in problems of nonlinear hydrodynamics of dense media with sources (Chapter V, Appendix II).

The book offers approaches to the study of the problems formulated above. At the same time, we emphasize that these problems obviously still have no final solution, so the development of the response theory in relation to the study of linear and nonlinear characteristics of non-ideal media and nonlinear phenomena in these media is relevant. As in the linear case, we investigate phenomena and properties corresponding to non-ideal media, and compare them (if possible) with analogues previously studied for a rarefied medium (plasma).

The book includes five Chapters and three Appendixes. Chapter I discusses, the theory of nonlinear response formulated to mechanical disturbances (electric and electromagnetic fields). Various variants of the fluctuation-dissipation theorem and dispersion relations are considered. Frequency moments are discussed of nonlinear response functions to longitudinal electric, and electromagnetic fields.

Nonlinear phenomena in non-ideal charged media are investigated in Chapter II based on the NRT for mechanical disturbances [7-10]. Phenomena related to the quadratic reaction of a non-ideal charged medium to longitudinal electric and electromagnetic fields are considered. These include plasma echo, wave transformation, second harmonic generation and parametric generation of radiation [10, 11]. Note that these phenomena have been studied in numerous theoretical and experimental works for plasma and electron gas with weak interparticle interaction (see, for example, [12, 13]). In these cases, theoretical approaches were based on the kinetic equations of Vlasov, Landau, etc. or the corresponding quantum mechanical approximations, which are not applicable for non-ideal media (see, for example, [1, 2]). The generation of the second harmonic of radiation and parametric generation of radiation were earlier studied in detail for various crystalline media.

The theory of reaction to thermal disturbances, which determines the transport properties of non-ideal media, is presented in Chapter III. This theoretical variant is based on a comparison of the conservation equations for a continuous (charged or neutral) medium and the generalized Langevin equations [3, 4] for the corresponding dynamic variables. In this approach to determining the second-order transport coefficients (linearized and nonlinear Burnett coefficients), expressions for heat, mass, momentum and charge fluxes are used in the most general form [14, 15].

Chapter IV analyzes the phenomenological relations of nonequilibrium thermodynamics, useful for hydrodynamic applications, which differ from the traditional linear-gradient approach [3[a]]. These sets of relations between the data on the thermophysical characteristics of non-ideal media determine the properties of an array of parameters in systems of conservation equations corresponding to problems of high-temperature nonlinear hydrodynamics (properties of matrices with higher derivatives, etc.). The properties of a matrix with higher derivatives in a system of conservation equations are discussed outside the framework of Euler's theorem on homogeneous functions.

Chapter V discusses a typical problem of nonlinear hydrodynamics of a dense medium with a volumetric heat source, in which consideration of nonlinear transport processes determines the main effects. Various thermal regimes and their dependence on parameters are investigated for conditions, which are actual for gas-phase nuclear reactors.

In Appendix I, a fluctuation-dissipation theorem, frequency moments of dense plasma quadratic response functions to electromagnetic field and a model for determination of the quadratic response functions are discussed (compare with [10[e]]). In Appendix II, methodically related to Chapter V, the actual problem of the multiplicity of heat and mass transfer modes in the shock layer of a spacecraft in the atmosphere of Mars is considered. The search is carried out to determine the most unfavorable modes in a wide range of parameter changes. In Appendix III (compare with the previous section), a structure and processes of heat and mass transfer are simulated in the system of exhaust jets of space vehicle brake motors during descent in the atmosphere of Mars. It is taken into account that the jet system is a chemically reacting gas medium formed by exhaust gases and atmosphere of the planet at sufficiently high temperatures and is characterized by developed turbulence. The calculated fields of the complete set of parameters of the medium, formed by the system of interacting supersonic exhaust jets of brake motors of the descent vehicle in the atmosphere of the planet, are obtained for realistic conditions. The parameters of the flow field in the most characteristic areas of the system of interacting jets are presented.

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Chapter I
The theory of nonlinear response to electric and electromagnetic field

The nonlinear response theory, the formulation of which is devoted to the first Chapter, allows us to determine the functions of the nonlinear reaction of a non-ideal charged medium to electric and electromagnetic fields (§1). The fluctuation-dissipation theorem, the symmetry relations, and the Kramers-Kronig relations are formulated for nonlinear response functions (§2). In the following paragraphs, the asymptotics (§3) and frequency moments (§§ 4, 5) of quadratic response functions, defining nonlinear phenomena (see Chapter II) are discussed. The frequency moments of both longitudinal (scalar) response functions (§4) and response functions to a transverse electromagnetic field (§5) are considered. At the same time, it should be borne in mind the difference between "screened" and "external" (describing the reaction to external disturbances) response functions (see Chapters II, III for more details). When presenting the material (conducting the study), the results of [7-10, 16] were used.

§ 1. Nonlinear reaction of a system to mechanical disturbances

We formulate the theory of nonlinear response to external disturbances based on the quantum Liouville equation (see, for example, [2, 3])

$$(1.1)\qquad i\hbar\frac{\partial\rho}{\partial t} = [H + H^{ext}, \rho]; \qquad \rho\big|_{t\to-\infty} = \rho_e\,.$$

Here ρ is the density matrix of the system; $\hbar$, k_B are the Planck and the Boltzmann constants; ρ_e is the density matrix of a medium in the absence of perturbation; H is the Hamiltonian of the medium;

$\beta = 1/k_B T$; and T is the temperature. The sign H^{ext} is a small addition to the Hamiltonian H due to the interaction of the medium with an external perturbation included in an infinitely remote time in the past. The symbol [.,.] means the commutator. The formal solution of (1.1) is in the following form [2, 3]

$$(1.2) \quad \rho(t) = \rho_e + \int_{-\infty}^{t} e^{-iH(t-t')/\hbar} \frac{1}{i\hbar} [H^{ext}, \rho] e^{iH(t-t')/\hbar} dt' .$$

Or in iterations

$$\rho(t) = \rho_e + \sum_{n=1}^{\infty} \frac{1}{(i\hbar)^n} \int_{-\infty}^{t} dt_1 \ldots \int_{-\infty}^{t_{n-1}} dt_n \, e^{-iHt/\hbar} [H^{ext}(t_1) \times$$

$$(1.3) \quad \times [H^{ext}(t_2) \ldots [H^{ext}(t_n), \rho_e] \ldots] e^{iHt/\hbar} ,$$

$$H^{ext}(t) = e^{iHt/\hbar} H^{ext} e^{-iHt/\hbar}$$

In these expressions, if the reaction of a medium consisting of spin-less charged particles to an external electromagnetic field is considered, H^{ext} is in the form [1-3]

$$(1.4) \quad H^{ext} = \sum_i \int d\mathbf{r} \rho_i(\mathbf{r}) \varphi^{ext}(\mathbf{r}, t) e^{\eta t} - \frac{1}{c} \int d\mathbf{r} \mathbf{j}(\mathbf{r}) \mathbf{A}(\mathbf{r}, t) e^{\eta t} +$$
$$+ \frac{e^2}{mc^2} \int d\mathbf{r} \rho(\mathbf{r}) A^2(\mathbf{r}, t) e^{\eta t} .$$

Here φ, $\mathbf{A}$, $\mathbf{j}$, ρ_i are, respectively, the potential, the vector potential (external), the current density operators (in the absence of a field), and the densities of particles of different charge in the medium. The letter η means a small positive value that ensures the adiabatic inclusion of the disturbance (and causality [1, 2]).

Consider the case when H^{ext} is linear in the external disturbance. Let us write down the interaction Hamiltonian for generality in the form (cf. with (1.4))

(1.5) $$H^{ext} = -\sum_j \int d\mathbf{r} B_j(\mathbf{r}) b_j^{ext}(\mathbf{r},t)$$

Here $B(\boldsymbol{r})$ is some observable property of the system (for example, the volume charge density), $b^{ext}(\mathbf{r},t)$ is a generalized external force (for example, the potential created by third-party charges).

We write an expression for the response of some observable property of the medium B to an external disturbance

$$\langle B\rangle = \langle B\rangle_0 + \sum_{n=1}^{\infty}\frac{1}{(i\hbar)^n}\int_{-\infty}^{t} dt_1 \ldots \int_{-\infty}^{t_{n-1}} dt_n\, Sp\{B(r,t)[H^{ext}(t_1) \times$$

(1.6) $$\times\, [H^{ext}(t_2)\ldots [H^{ext}(t_n),\rho_e] \ldots]\}$$

Here $\langle\ldots\rangle$, $\langle\ldots\rangle_0$ mean averaging over ρ and ρ_e, respectively, and

$B(\boldsymbol{r},t)$ is an operator in the Heisenberg representation [2]. In this case, the quadratic response has the form

$$\langle B\rangle^{(2)} = \frac{1}{(i\hbar)^2}\int_{-\infty}^{t}\int_{-\infty}^{t_1} Sp\{B(\mathbf{r},t)[H^{ext}(t_1)[H^{ext}(t_2),\rho_e]]\}dt_1 dt_2;$$

(1.7) $$\langle B_i\rangle^{(2)} = \sum_{j,k}\int_{-\infty}^{t}\int_{-\infty}^{t_1}\hat{\chi}_{ijk}^{(2)}(t-t_1,t-t_2;\mathbf{r}-\mathbf{r}_1,\mathbf{r}-\mathbf{r}_2)b_j^{ext}(\mathbf{r}_1,t_1)\times$$
$$\times\, b_k^{ext}(\mathbf{r}_2,t_2)dt_1 dt_2 d\mathbf{r}_1 d\mathbf{r}_2;$$

$$\hat{\chi}_{ijk}^{(2)}(\mathbf{r}-\mathbf{r}_1,t-t_1;\mathbf{r}-\mathbf{r}_2,t-t_2) = -\frac{\theta(t-t_1)\theta(t-t_2)}{2\hbar^2 V}\{\theta(t_1-t_2)\times$$

(1.8) $$\times\langle\!\langle[[B_i(\mathbf{r},t),B_j(\mathbf{r}_1,t_1)],B_k(\mathbf{r}_2,t_2)]\rangle\!\rangle_0 +$$
$$+\,\theta(t_2-t_1)\langle\!\langle[[B_i(\mathbf{r},t),B_k(\mathbf{r}_2,t_2)],B_j(\mathbf{r}_1,t_1)]\rangle\!\rangle_0\}.$$

Such a definition of the response function allows in (1.7) to extend the limits of integration over time to infinity. Substitute

(1.8) into (1.7) and proceed to the Fourier representation

(1.9)
$$\langle B_i(\mathbf{k},\omega)\rangle^{(2)} = \frac{1}{V(2\pi)^2}\sum_{\mathbf{k}_1,\mathbf{k}_2}\int d\omega_1 d\omega_2 \times$$
$$\times\hat{\chi}^{(2)}_{ijk}(\mathbf{k}_1\omega_1;\mathbf{k}_2\omega_2)b_j^{ext}(\mathbf{k}_1\omega_1)b_k^{ext}(\mathbf{k}_2\omega_2);$$
$$\mathbf{k}=\mathbf{k}_1+\mathbf{k}_2;\quad \omega=\omega_1+\omega_2.$$

Let consider the properties of the second - order response function $\hat{\chi}^{(2)}_{ijk}$. The symmetry of this response function with respect to the last two indices is evident from definition (1.8). For the Fourier image of the response function $\hat{\chi}^{(2)}_{ijk}(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)$ we have

(1.10) $$\hat{\chi}^{(2)}_{ijk}(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)=\hat{\chi}^{(2)*}_{ijk}(-\mathbf{k}_1,-\omega_1;-\mathbf{k}_2,-\omega_2).$$

Because, $\hat{\chi}^{(2)}_{ijk}(\mathbf{r}_1,\tau_1;\mathbf{r}_2,\tau_2)$ is a real function, which follows from its phenomenological definition. Let us show formally the validity of the last statement. From the hermiticity of operators *B(**r**,t)* [17] it follows that $[B_i(\mathbf{r}_1,t_1),B_j(\mathbf{r}_2,t_2)]$ is the "anti-Hermite" operator, in addition ρ_e is the Hermite operator [17], therefore we will have

(1.11) $$[\hat{\chi}_{ijk}^{(2)}(\mathbf{r}_1,t_1;\mathbf{r}_2,t_2)]^*=\hat{\chi}^{(2)}_{ijk}(\mathbf{r}_1,t_1;\mathbf{r}_2,t_2).$$

It is of interest to determine the properties of correlators of some operators through which quadratic response functions can be expressed (see (1.8), §2), with respect to time reversal and coordinate inversion. We investigate the properties of correlators. The time reversal operator T is given as follows (see, for example, [3,18])

$$(T\Psi, T\Phi) = (\Phi, \Psi)$$

(1.12) $TB_i(\mathbf{r},t)T^{-1} = \varepsilon_i B_i(\mathbf{r},-t)$;

$\varepsilon_i = 1(-1)$, if the corresponding operator is even (odd) with respect to the time reversal. Considering (1.12), as well as the possibility of permutation of operators under the sign of the *Sp*, we obtain for the correlator of operators

(1.13) $f_{ijk}^{(2)}(\mathbf{r}_1,t_1;\mathbf{r}_2,t_2) = \langle B_i(0,0); B_j(\mathbf{r}_1,t_1); B_k(\mathbf{r}_2,t_2)\rangle_0$;

$$f_{ijk}^{(2)}(\mathbf{r}_1,-t_1;\mathbf{r}_2,-t_2) = Sp\langle T\Psi^*, \rho_e B_i(0,0)B_j(\mathbf{r}_1,-t_1)B_k(\mathbf{r}_2,-t_2)T\Psi\rangle =$$

$$= \varepsilon_i\varepsilon_j\varepsilon_k Sp\langle T\Psi^*, \rho_e TB_i(0,0)T^{-1}TB_j(\mathbf{r}_1,t_1)T^{-1}TB_k(\mathbf{r}_2,t_2)T^{-1}T\Psi\rangle =$$

$$= \varepsilon_i\varepsilon_j\varepsilon_k Sp\langle T\Psi^*, T\rho_e B_i\ (0,0)B_j\ (\mathbf{r}_1,t_1)B_k(\mathbf{r}_2,t_2)\Psi\rangle = \varepsilon_i\varepsilon_j\varepsilon_k \times$$

$$\times f_{ijk}^{(2)*}(\mathbf{r}_1,t_1;\mathbf{r}_2,t_2)$$

Consider the properties of correlators with respect to the inversion of coordinates; the inversion operator *I* looks like as follows [18]

$$(I\Psi, I\Phi) = (\Psi, \Phi)$$

(1.14) $IB_i(\mathbf{r},t)I^{-1} = \varepsilon_i^I B_i(-\mathbf{r},t)$;

$\varepsilon_i^I = 1(-1)$, if the corresponding operator is even (odd) with respect to the inversion of coordinates. Considering (1.14), as well as the possibility of permutation of operators under the sign of the *Sp*, we obtain for the correlator

$$f_{ijk}^{(2)}(-\mathbf{r}_1,t_1;-\mathbf{r}_2,t_2) = Sp\langle I\Psi^*,\rho_e B_i(0,0)B_j(-\mathbf{r}_1,t_1)B_k(-\mathbf{r}_2,t_2)I\Psi\rangle =$$

$$(1.15) = \varepsilon_i^I\varepsilon_j^I\varepsilon_k^I Sp\langle I\Psi^*,\rho_e IB_i\ (0,0)I^{-1}IB_j\ (\mathbf{r}_1,t_1)I^{-1}IB_k(\mathbf{r}_2,t_2)I^{-1}I\Psi\rangle$$

$$= \varepsilon_i^I\varepsilon_j^I\varepsilon_k^I\ f_{ijk}^{(2)}(\mathbf{r}_1,t_1;\mathbf{r}_2,t_2)\,.$$

The relations between the Fourier images of correlators have the form (see (1.10), (1.11))

$$\langle B_i(-\mathbf{k},-\omega)B_j(\mathbf{k}_1,\omega_1)B_k(\mathbf{k}_2,\omega_2)\rangle_0 =$$

$$= \langle B_i(-\mathbf{k},-\omega)B_j(\mathbf{k}_1,\omega_1)B_k(\mathbf{k}_2,\omega_2)\rangle_0^*$$

$$(1.16)\quad \langle B_i(\mathbf{k},\omega)B_j(-\mathbf{k}_1,-\omega_1)B_k(-\mathbf{k}_2,-\omega_2)\rangle_0 =$$

$$= \varepsilon_i\varepsilon_j\varepsilon_k\ \varepsilon_i^I\varepsilon_j^I\varepsilon_k^I \times \langle B_i(-\mathbf{k},-\omega)B_j(\mathbf{k}_1,\omega_1)B_k(\mathbf{k}_2,\omega_2)\rangle_0^*$$

We define the properties of correlators with respect to cyclic permutations of operators

$$f_{ijk}^{(2)}(\mathbf{r}_1,t_1;\mathbf{r}_2,t_2) = \langle B_i(0,0);B_j(\mathbf{r}_1,t_1);B_k(\mathbf{r}_2,t_2)\rangle_0 =$$

$$= \langle B_k(\mathbf{r}_2,t_2-i\hbar\beta);B_i(0,0);B_j(\mathbf{r}_1,t_1)\rangle_0\,.$$

Hence, for Fourier images of correlators, we get (N is the number of particles in the system)

$$\langle B_i(-\mathbf{k},-\omega)B_j(\mathbf{k}_1,\omega_1)B_k(\mathbf{k}_2,\omega_2)\rangle_0 =$$

$$(1.17)\quad = 2\pi N\delta(\mathbf{k}-\mathbf{k}_1-\mathbf{k}_2)\delta(\omega-\omega_1-\omega_2)S(012)\,;$$

$$e^{-\beta\hbar\omega_2}S(201) = S(012), \text{ etc.}$$

The first of the relations (1.17) is the definition of correlators *S*.

It is easy to find the properties of $\hat{\chi}_{ijk}^{(2)}(\mathbf{r}_1,t_1;\mathbf{r}_2,t_2)$ (1.8) with respect to time reversal and coordinate inversion, using

(1.13) and (1.15). The properties of correlators are used to determine and study nonlinear kinetic (transport) coefficients (see Chapter III).

§ 2. Nonlinear fluctuation-dissipation theorem. Dispersion relations and frequency moments of response functions

As is known, the linear fluctuation-dissipation theorem connects linear response functions and correlation functions of physical quantities at two different spatiotemporal points (see, for example, [3[a], 18]), while the correlation function is connected with the imaginary (dissipative) part of the response function. Nonlinear FDT has a more complex structure, which includes response functions and correlators of higher orders. Let us consider an approach that allows us to find the relationship between the response functions and the corresponding correlators, using the example of a quadratic FDT. Let write out the quadratic response function (1.8)

$$\hat{\chi}^{(2)}_{ijk}(\mathbf{r}-\mathbf{r}_1,t-t_1;\mathbf{r}-\mathbf{r}_2,t-t_2) = -\frac{\theta(t-t_1)\theta(t-t_2)}{2\hbar^2 V}\{\theta(t_1-t_2)\times$$
$$\times\langle[[B_i(\mathbf{r},t),B_j(\mathbf{r}_1,t_1)],B_k(\mathbf{r}_2,t_2)]\rangle_0 +$$
$$+\theta(t_2-t_1)\langle[[B_i(\mathbf{r},t),B_k(\mathbf{r}_2,t_2)],B_j(\mathbf{r}_1,t_1)]\rangle_0\}. \tag{2.1}$$

In this expression, we will describe the commutators in terms of correlators, the properties of which discussed in the previous paragraph, and perform the space-time Fourier transform

$$\hat{\chi}_{ijk}{}^{(2)}(\mathbf{r}-\mathbf{r}_1,t-t_1;\mathbf{r}-\mathbf{r}_2,t-t_2)=-\frac{\theta(t-t_1)\theta(t-t_2)}{2\hbar^2 V}\{\theta(t_1-t_2)\times$$
$$\langle B_i(\mathbf{r},t)B_j(\mathbf{r}_1,t_1)B_k(\mathbf{r}_2,t_2)-B_j(\mathbf{r}_1,t_1)B_i(\mathbf{r},t)B_k(\mathbf{r}_2,t_2)-$$
$$-B_k(\mathbf{r}_2,t_2)B_i(\mathbf{r},t)B_j(\mathbf{r}_1,t_1)+B_k(\mathbf{r}_2,t_2)B_j(\mathbf{r}_1,t_1)B_i(\mathbf{r},t)\rangle_0+$$
$$+\theta(t_2-t_1)\langle B_i(\mathbf{r},t)B_k(\mathbf{r}_2,t_2)B_j(\mathbf{r}_1,t_1)-B_k(\mathbf{r}_2,t_2)B_i(\mathbf{r},t)B_j(\mathbf{r}_1,t_1)\rangle_0$$
$$-\theta(t_2-t_1)\langle B_j(\mathbf{r}_1,t_1)B_i(\mathbf{r},t)B_k(\mathbf{r}_2,t_2)-B_j(\mathbf{r}_1,t_1)B_k(\mathbf{r}_2,t_2)B_i(\mathbf{r},t)\rangle_0\}$$

$$\hat{\chi}^{(2)}_{ijk}(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)=$$
$$=-\frac{N}{2\hbar^2 V}\{\int\frac{d\omega_1'}{2\pi}\int\frac{d\omega_2'}{2\pi}[1/i(\omega_1'-\omega_1)]/i(\omega_1'+\omega_2'-\omega_1-\omega_2)\times$$
$$\times[S(012)+S(210)-S(102)-S(201)]+$$
$$+\int\frac{d\omega_1'}{2\pi}\int\frac{d\omega_2'}{2\pi}[1/i(\omega_2'-\omega_2)][1/i(\omega_1'+\omega_2'-\omega_1-\omega_2)]\times \tag{2.2}$$
$$\times[S(021)+S(120)-S(102)-S(201)]\}\,.$$

The ratio, used here, is ((1.17) re-assigns to (2.3) for convenience)

$$\langle B_i(-\mathbf{k},-\omega)B_j(\mathbf{k}_1,\omega_1)B_k(\mathbf{k}_2,\omega_2)\rangle_0=2\pi N\delta(\omega-\omega_1-\omega_2)\delta(\mathbf{k}-\mathbf{k}_1-\mathbf{k}_2)\times$$
$$\times S(012) \tag{2.3}$$

Relation (2.2) is one of the forms of the fluctuation-dissipation theorem. Applying the Sokhotsky formulas [19], we obtain the relations between the real parts of $\{\hat{\chi}^{(2)}_{ijk}(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)\}$ ($\{\ldots\}$ means a set of $\hat{\chi}^{(2)}_{ijk}$ with a different order of arguments) and the correlator (see Appendix I for details). Note that in this form of NFDT, the real part of the quadratic response function is directly related to the correlators [7], in contrast to the linear FDT (see [1-3]), where the imaginary part of the linear response function is determined by the correlator.

Let us consider the consequences of the causality of response functions $\hat{\chi}_{ijk}^{(2)}(\mathbf{r}_1,t_1;\mathbf{r}_2,t_2)$. From (1.8) follows the analyticity of Fourier image of this function in the upper half-plane of complex variables ω_1,ω_2, since given function is equal to zero at negative times (it is a causal function). We obtain the Kramers-Kronig relations with respect to ω_1, and write out the integral

$$(2.4)\qquad \oint_{\Gamma} \frac{\hat{\chi}_{ijk}^{(2)}(\mathbf{k}_1,\omega_1';\mathbf{k}_2,\omega_2)}{\omega_1-\omega_1'}d\omega_1'$$

We calculate the integral along the contour *Γ,* passing along the real axis from -∞ to +∞, wrapping around a special point $\omega_1 = \omega_1'$ from above, and closing along a semicircle in the upper half-plane of the complex variable ω_1'. The integral (2.4) is zero due to the analyticity of the response function in the upper half-plane ω_1'. It is possible, therefore, to distinguish contributions from three sections of the contour of *Γ*. The integral over the large semicircle turns to zero, because at infinity the response function tends to zero. What remains is an integral over two segments of the real axis (which is the main value of the integral) and an integral over a semicircle around a point ω_1. We get

$$(2.5)\qquad P\int \frac{\hat{\chi}_{ijk}^{(2)}(\mathbf{k}_1,\omega_1';\mathbf{k}_2,\omega_2)}{\omega_1-\omega_1'}d\omega_1' + i\pi\hat{\chi}_{ijk}^{(2)}(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)=0\,.$$

We write out the real and imaginary parts of this expression

$$\operatorname{Re}\hat{\chi}^{(2)}_{ijk}(\mathbf{k}_1\omega_1;\mathbf{k}_2\omega_2) = -\frac{1}{\pi}P\int\frac{\operatorname{Im}\hat{\chi}^{(2)}_{ijk}(\mathbf{k}_1,\omega_1';\mathbf{k}_2,\omega_2)}{\omega_1-\omega_1'}d\omega_1'$$

(2.6)

$$\operatorname{Im}\hat{\chi}^{(2)}_{ijk}(\mathbf{k}_1\omega_1;\mathbf{k}_2\omega_2) = \frac{1}{\pi}P\int\frac{\operatorname{Re}\hat{\chi}^{(2)}_{ijk}(\mathbf{k}_1,\omega_1';\mathbf{k}_2,\omega_2)}{\omega_1-\omega_1'}d\omega_1'$$

Similarly, it is possible to obtain dispersion relations with respect to ω_2 and ω_1,ω_2

$$(2.7)\quad \hat{\chi}^{(2)}_{ijk}(\mathbf{k}_1\omega_1;\mathbf{k}_2\omega_2) = -\frac{1}{\pi^2}P\int P\int\frac{\hat{\chi}^{(2)}_{ijk}(\mathbf{k}_1,\omega_1';\mathbf{k}_2,\omega_2')}{(\omega_1-\omega_1')(\omega_2-\omega_2')}d\omega_1'd\omega_2'$$

Let consider the decomposition of (2.6) (similarly for (2.7)), when $\omega_1(\omega_2)\to\infty$

$$\operatorname{Re}\hat{\chi}^{(2)}_{ijk}(\mathbf{k}_1,\omega_1\to\infty;\mathbf{k}_2,\omega_2)\cong$$

$$\cong -\frac{1}{\omega_1}\int\frac{d\omega_1'}{\pi}[1+\frac{\omega_1'}{\omega_1}+(\frac{\omega_1'}{\omega_1})^2+\ldots]\operatorname{Im}\hat{\chi}^{(2)}_{ijk}(\mathbf{k}_1,\omega_1';\mathbf{k}_2,\omega_2);$$

(2.8)

$$\operatorname{Im}\hat{\chi}^{(2)}_{ijk}(\mathbf{k}_1,\omega_1\to\infty;\mathbf{k}_2,\omega_2)\cong$$

$$\cong \frac{1}{\omega_1}\int\frac{d\omega_1'}{\pi}[1+\frac{\omega_1'}{\omega_1}+(\frac{\omega_1'}{\omega_1})^2+\ldots]\operatorname{Re}\hat{\chi}^{(2)}_{ijk}(\mathbf{k}_1,\omega_1';\mathbf{k}_2,\omega_2).$$

The numerators in these expansions are determined by the frequency moments of the imaginary and real parts of the quadratic response function $\hat{\chi}^{(2)}_{ijk}(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)$. It is also possible to obtain decompositions in ω_2 and ω_1,ω_2, which are determined by the corresponding frequency moments.

High-frequency decompositions of response functions in the time representation determine their behavior at short times (see the next paragraph). Indeed, by integration in parts, we will have

$$\hat{\chi}_{ijk}^{(2)}(\mathbf{k}_1\omega_1;\mathbf{k}_2\omega_2) = \int_0^\infty \hat{\chi}_{ijk}{}^{(2)}(\mathbf{k}_1,t_1;\mathbf{k}_2,\omega_2)e^{i\omega_1 t_1}dt_1 =$$

$$= -\frac{1}{i\omega_1}\hat{\chi}_{ijk}^{(2)}(\mathbf{k}_1,t_1;\mathbf{k}_2\omega_2)\,\Big|_{t_1=0} +$$

$$+\frac{1}{(i\omega_1)^2}\frac{\partial}{\partial t_1}\hat{\chi}_{ijk}^{(2)}(\mathbf{k}_1,t_1;\mathbf{k}_2\omega_2)\,\Big|_{t_1=0} -$$

$$-\frac{1}{(i\omega_1)^3}\frac{\partial^2}{\partial t_1^2}\hat{\chi}_{ijk}^{(2)}(\mathbf{k}_1,t_1;\mathbf{k}_2\omega_2)\,\Big|_{t_1=0} + \dots$$

We obtain a chain of frequency moments for the real and imaginary parts of the quadratic response function by comparing the above relation with expressions of type (2.8). Note that the zero frequency moments are equal to zero.

(2.9) $$\int \frac{d\omega_1}{\pi}\omega_1\hat{\chi}_{ijk}^{(2)}(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2) = i\frac{\partial}{\partial t_1}\hat{\chi}_{ijk}^{(2)}(\mathbf{k}_1,t_1;\mathbf{k}_2\omega_2)\,\Big|_{t_1=0};$$

$$\int \frac{d\omega_1}{\pi}\omega_1^2\hat{\chi}_{ijk}^{(2)}(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2) = -\frac{\partial^2}{\partial t_1^2}\hat{\chi}_{ijk}^{(2)}(\mathbf{k}_1,t_1;\mathbf{k}_2\omega_2)\,\Big|_{t_1=0};\ etc.$$

It should be noted that these relations are convenient for practical calculation of the frequency moments of response functions (§4, compare with [3[a], 7]). Similar relations for the two frequencies can be obtained by comparing high-frequency decompositions (2.7) and two-time integration in parts of the response function.

§ 3. The asymptotics of response functions

We will discuss the properties of specific quadratic response functions in this and the following paragraphs of this Chapter. Let consider the asymptotics of the quadratic longitudinal charge-

charge response function $\hat{\chi}^{(2)}_{\rho\rho\rho}$ at $\omega_1, \omega_2 \to \infty$. Such a limit, as is known [16], corresponds to t_1, $t_2 \to 0$, when the system is considered as a system of non-interacting particles. For a classical system of non-interacting charged particles, the quadratic charge-charge response function asymptotics are found (see, for example, [7, 16]). Let define this function (see (1.8), (1.9) and compare with (8.1)).

$$\langle \rho(\mathbf{k},\omega)\rangle^{(2)} = \frac{1}{V(2\pi)^2} \sum_{\mathbf{k}_1,\mathbf{k}_2} \int d\omega_1 d\omega_2 \hat{\chi}^{(2)}_{\rho\rho\rho}(\mathbf{k}_1\omega_1;\mathbf{k}_2\omega_2)\varphi^{ext}(\mathbf{k}_1\omega_1)\times$$

$$(1.9)' \quad \times \varphi^{ext}(\mathbf{k}_2,\omega_2);$$

$$\mathbf{k} = \mathbf{k}_1 + \mathbf{k}_2; \qquad \omega = \omega_1 + \omega_2.$$

We have in the high frequency limit

$$\hat{\chi}^{(2)}_{\rho\rho\rho}(\omega_1 \to \infty, \mathbf{k}_1; \omega_2 \to \infty, \mathbf{k}_2) = \frac{e}{2m_e} \frac{\omega_p^2}{4\pi\omega\omega_1\omega_2} [\frac{k^2}{\omega}(\mathbf{k}_1 \cdot \mathbf{k}_2) +$$

$$(3.1) \quad + \frac{k_1^2}{\omega_1}(\mathbf{k} \cdot \mathbf{k}_2) + \frac{k_2^2}{\omega_2}(\mathbf{k} \cdot \mathbf{k}_1)]$$

In (3.1) e, m_e, ω_p are the charge of the electron, mass of the electron, and plasma frequency. This expression makes it possible to analyze frequency moments $\hat{\chi}^{(2)}_{\rho\rho\rho}$, taking into account the symmetry properties of a given response function (see, for example, (4.3)). The real part dominates in the high-frequency limit of $\hat{\chi}^{(2)}_{\rho\rho\rho}$. Therefore, from (3.1) should

$$\operatorname{Re}\hat{\chi}^{(2)}_{\rho\rho\rho}(\omega_1 \to \infty, \mathbf{k}_1; \omega_2 \to \infty, \mathbf{k}_2) = \frac{e}{2m_e} \frac{\omega_p^2}{4\pi\omega_1^2\omega_2^2} R(\mathbf{k}_1, \mathbf{k}_2)$$

(3.2)
$$R(\mathbf{k}_1, \mathbf{k}_2) = \begin{cases} k_1^2 (\mathbf{k} \cdot \mathbf{k}_2), & \dfrac{\omega_2}{\omega_1} \to \infty \\ k_2^2 (\mathbf{k} \cdot \mathbf{k}_1), & \dfrac{\omega_1}{\omega_2} \to \infty \end{cases}.$$

This is the main, electronic, contribution to these ratios.

Let us write out the consequences of (3.2). We have at $\omega_1 \to \infty$ (because, there are relations (4.1))

$$\lim_{\omega_1 \to \infty} P\int \frac{\operatorname{Im}\hat{\chi}^{(2)}_{ijk}(\mathbf{k}_1, \omega_1'; \mathbf{k}_2, \omega_2 \to \infty)}{\omega_1 - \omega_1'} d\omega_1' =$$
$$= \lim_{\omega_1 \to \infty} \frac{1}{\omega_1^2} \int_{-\infty}^{\infty} d\omega_1' \omega_1' \operatorname{Im}\hat{\chi}^{(2)}_{ijk}(\omega_1', \mathbf{k}_1; \omega_2 \to \infty, \mathbf{k}_2) + \ldots .$$

We will have (3.3) [7], using the above relation and (2.6), (3.1)

(3.3)
$$\int_{-\infty}^{\infty} d\omega_1 \omega_1 \operatorname{Im}\hat{\chi}^{(2)}_{\rho\rho\rho}(\omega_1, \mathbf{k}_1; \omega_2 \to \infty, \mathbf{k}_2) = -\frac{e}{8m_e} \frac{\omega_p^2}{\omega_2^2} k_1^2 \mathbf{k} \cdot \mathbf{k}_2$$
$$\int_{-\infty}^{\infty} d\omega_2 \omega_2 \operatorname{Im}\hat{\chi}^{(2)}_{\rho\rho\rho}(\omega_1 \to \infty, \mathbf{k}_1; \omega_2, \mathbf{k}_2) = -\frac{e}{8m_e} \frac{\omega_p^2}{\omega_1^2} k_2^2 \mathbf{k} \cdot \mathbf{k}_1$$

The relations (3.3) are considered as the simplest exact frequency moments of the quadratic charge-charge response function (not only for the classical system). Analogous asymptotics can be found for moments of tensor quadratic response functions (see §5), using the analogues of (3.1) [16] for the corresponding response functions.

We obtain, following the relation (2.7), a high-frequency decomposition for $\hat{\chi}^{(2)}_{\rho\rho\rho}$ (see notation (4.4))

$$(3.4)\quad \hat{\chi}^{(2)}_{\rho\rho\rho}(\mathbf{k}_1\omega_1 \to \infty; \mathbf{k}_2\omega_2 \to \infty) =$$

$$= -\int \frac{d\omega_2'}{\pi} \int \frac{d\omega_1'}{\pi} \frac{\hat{\chi}^{(2)}_{\rho\rho\rho}(\mathbf{k}_1, \omega_1'; \mathbf{k}_2, \omega_2')}{\omega_1\omega_2} \times [1 + \frac{\omega_1'}{\omega_1} + \ldots][1 + \frac{\omega_2'}{\omega_2} + \ldots] =$$

$$= -4\left(\frac{X_{1,1}}{\omega_1^2\omega_2^2} + \frac{X_{2,0}}{\omega_1^3\omega_2} + \frac{X_{0,2}}{\omega_1\omega_2^3} + \frac{X_{1,.3}}{\omega_1^2\omega_2^4} + \frac{X_{2,2}}{\omega_1^3\omega_2^3} + \ldots \right)$$

A concretization of expressions of the form (3.4) (calculation of $X_{a,b}$ (4.4)) makes it possible to better clarify the structure of high-frequency asymptotics of response functions, which are determined by their frequency moments. In addition, we can obtain the corresponding decomposition of $\mathrm{Re}\hat{\chi}^{(2)}_{\rho\rho\rho}$ in the form (3.4), if we take into account the dominant contribution of the real part to the quadratic response function $\hat{\chi}^{(2)}_{\rho\rho\rho}$ in the high-frequency limit. In this case, the consequences of quadratic FDT (see (2.2), (2.3) allow us to find the frequency moments of triple correlators of charge densities. A more detailed analysis of the frequency moments of the nonlinear response function $\hat{\chi}^{(2)}_{\rho\rho\rho}$ is carried out below (see also Appendix I).

§ 4. Frequency moments of quadratic response functions. Longitudinal charge-charge response function

The frequency moments of the quadratic response functions are frequency integrals of these functions multiplied by various degrees of frequency. Such integrals are essentially thermodynamic characteristics of the medium and, therefore, can be studied more fully than the actual response functions. In this paragraph, the known response function $\hat{\chi}^{(2)}_{\rho\rho\rho}$ to the longitudinal field is mainly analyzed [7-9]. The frequency moments of the response functions to the electromagnetic field (see (1.4)) discussed in the next paragraph.

A distinction should be made between the "screened" and "external" response functions [1-3], when analyzing the quadratic response functions to the longitudinal field. The relations linking these response functions are obtained in Chapter II.

Let us start with the zero moment of the quadratic response functions. Since the "external" response functions describe the reaction of the medium to external disturbances and are causal functions, they do not have features in the upper half-plane of complex variables ω_1, ω_2. Therefore, assuming that these functions decrease fast enough at infinity ω_1, ω_2, we can show

$$(4.1)\quad \int_{-\infty}^{\infty} d\omega_1 \hat{\chi}^{(2)}_{ijk}(\omega_1,\mathbf{k}_1;\omega_2,\mathbf{k}_2) = \int_{-\infty}^{\infty} d\omega_2 \hat{\chi}^{(2)}_{ijk}(\omega_1,\mathbf{k}_1;\omega_2,\mathbf{k}_2) = 0 .$$

It is convenient to use relations of the form (2.9) and similar two-frequency expressions, to calculate the frequency moments of the response functions.

It is possible to define the longitudinal response functions through arbitrary tensor response functions, bearing in mind the fact that “densities” and “generalized external forces” in (1.4) can be vectors (or quantities of another tensor dimension). Let us assume, based on the definitions of the quadratic response function (see (1.8), (1.9); cf. with §5)

$$(4.2)\quad \hat{\chi}^{(2)}_{B^\alpha B^\beta B^\gamma}(\omega_1,\boldsymbol{k}_1;\omega_2,\boldsymbol{k}_2) = k_i k_j^1 k_k^2 \hat{\chi}^{(2)}_{ijk}(\omega_1,\boldsymbol{k}_1;\omega_2,\boldsymbol{k}_2).$$

Here $B^\alpha = k_j B_j^\alpha$ and so on. We present the known frequency moments for the most studied longitudinal quadratic response function of the density-density (charge-charge, cf. with §§6-8,

Appendix I) $\hat{\chi}^{(2)}_{\rho\rho\rho}(\omega_1,\mathbf{k}_1;\omega_2,\mathbf{k}_2)$ [7]

$$(4.3)\qquad \int_{-\infty}^{\infty} d\omega_1 \omega_1 \hat{\chi}^{(2)}_{\rho\rho\rho}(\omega_1,\mathbf{k}_1;\omega_2,\mathbf{k}_2) = i2\pi e \frac{\mathbf{k}\mathbf{k}_1}{4m_e}\hat{\chi}_{\rho\rho}(\omega_2,\mathbf{k}_2)$$

The relation (4.3) is obtained, using (2.9) for the case of a quantum system and relate three-particle response functions to two-particle (linear) ones. This relation is also valid for classical systems. Moments for higher degrees of ω_1 (and moments relatively ω_2) can be considered similarly.

Let us move on to the definition of two-frequency moments of the form

$$(4.4)\qquad X_{m,n}[a,b] = \frac{1}{4\pi^2}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty} d\omega_a d\omega_b \omega_a^m \omega_b^n \hat{\chi}^{(2)}_{\rho\rho\rho}(\omega_a,\mathbf{k}_a;\omega_b,\mathbf{k}_b)\,.$$

Let's start with $X_{-1,-1}$, using the Kramers-Kronig relations (2.7)

$$(4.5)\qquad X_{-1,-1}[1,2] = -\frac{1}{4\pi^2}\int\int \frac{\hat{\chi}^{(2)}_{\rho\rho\rho}(\mathbf{k}_1,\omega_1';\mathbf{k}_2,\omega_2')}{\omega_1'\omega_2'} d\omega_1' d\omega_2' =$$
$$= -\hat{\chi}^{(2)}_{\rho\rho\rho}(\mathbf{k}_1,0;\mathbf{k}_2,0)/4\,.$$

The moment $X_{0,0}$ [1,2], obviously (see (4.1)), is equal to zero; for the same reason, $X_{0,-1}$[1,2] and $X_{-1,0}$[1,2] are equal to zero (taking into account the Kramers-Kronig relations). The moments $X_{0,1}$[1,2] and $X_{1,0}$[1,2] are also equal to zero due to the analyticity of the linear response function (it's causality) in the upper half-plane of the complex variable ω. The moment $X_{-1,1}$[1,2] (which is equal to the moment $X_{1,-1}$[2,1]) is easy to find using the equality (4.3) and the Kramers-Kronig relations for linear response functions (in (4.6) – (4.8) we put e =1)