

# Hydrodynamic asymptotic forms of three-point space-time correlation functions of fluctuations of conserved quantities in a simple liquid

I. M. Tkachenko and I. Z. Fisher

Odessa State University

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The nonlinear three-index generalized susceptibilities of the hydrodynamic quantities of a classical liquid are calculated. With the aid of these, and on the basis of the three-index fluctuation-dissipation theorem, the dynamical three-point correlation functions of the thermal fluctuations of the densities of the conserved quantities are found. Sum rules for the three-point correlation functions are found.

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## 1. INTRODUCTION

The clear understanding, achieved about ten years ago, of the analytic structure of two-point space-time correlation functions (or Green functions) in the limit of small values of  $\omega$  and  $\mathbf{k}$  for the different dynamical variables in a simple classical liquid has played an important role in the development of modern methods and ideas in different applications of the theory of thermal fluctuations to slow-neutron and light scattering processes, acoustic relaxation, etc. The progress achieved is associated with the wide use of the two-index fluctuation-dissipation theorem, and with the possibility of direct calculation of the dynamical generalized susceptibilities for small  $\omega$  and  $\mathbf{k}$  from hydrodynamic models<sup>[1,2]</sup>.

Of substantial interest for the problem of multiple scattering of light and slow neutrons, for nonlinear acoustics, etc., are the many-point space-time correlation functions of the dynamical variables of a simple liquid. The lowest of these—the three-point functions—can be investigated with the same completeness as the two-point correlations, thanks to the proof of Efremov Efremov<sup>[3,4]</sup> and Stratonovich<sup>[5]</sup> of a three-index fluctuation-dissipation theorem that makes it possible to establish the three-point dynamical correlation functions of the different quantities for small  $\omega$  and  $\mathbf{k}$  exactly from the generalized susceptibilities found from hydrodynamic models.

In the present paper this program is carried out for densities of conserved quantities—the particle-number density  $n(\mathbf{r}, t)$ , internal-energy density  $h(\mathbf{r}, t)$  and momentum density  $\mathbf{g}(\mathbf{r}, t)$ .

For simplicity we consider a classical Newtonian fluid satisfying the usual hydrodynamic equations with frequency-independent kinetic coefficients. This leads to the necessity of taking only two types of nonlinearity into account in the equations of motion: 1) ordinary kinematic nonlinearities and 2) the nonlinearities associated with the dependence of the kinetic coefficients and thermodynamic quantities on the temperature and density.

In the classical limiting case  $\hbar\omega \ll k_B T$  the Efremov-Stratonovich theorem leads to the following expression<sup>[3]</sup>:

$$G_{abc}(1, 2) = \langle \delta x_a(0) \delta x_b(1) \delta x_c(2) \rangle = -(2k_B T)^2 \operatorname{Re} \left\{ \frac{\chi_{abc}(1, 2)}{\omega_1 \omega_2} \theta_{a_0} \theta_{b_0} \theta_{c_0} + \frac{\chi_{bca}(2, 0)}{\omega_2 \omega_0} + \frac{\chi_{cab}(0, 1)}{\omega_0 \omega_1} \right\}, \quad (1.1)$$

connecting the equilibrium correlation function of the

thermal fluctuations  $\delta x_a$ ,  $\delta x_b$ ,  $\delta x_c$  of any three dynamical variables with the generalized quadratic susceptibilities  $\chi_{abc}$ ,  $\chi_{bca}$ ,  $\chi_{cab}$  conjugate to them. The values of  $\theta_a$  are +1 or -1, depending on whether the quantity  $\delta x_a$  is even or odd under time reversal;  $T$  is the temperature. The symbols 0, 1, 2 denote pairs of Fourier arguments:

$$0 = (\mathbf{k}_0, \omega_0), \quad 1 = (\mathbf{k}_1, \omega_1), \quad 2 = (\mathbf{k}_2, \omega_2), \quad (1.2)$$

for which the relations

$$\mathbf{k}_0 + \mathbf{k}_1 + \mathbf{k}_2 = 0, \quad \omega_0 + \omega_1 + \omega_2 = 0, \quad (1.3)$$

which stem from the uniformity and stationarity conditions for the random processes under consideration, are fulfilled. The frequencies  $\omega_j$  and wave-vectors  $\mathbf{k}_j$  are assumed throughout to be real.

Because non-Newtonian effects have not been taken into account, it turns out that all three-index susceptibilities can be expressed in terms of combinations of known two-index susceptibilities. All the latter are expressed in terms of four independent susceptibilities<sup>[1,2]</sup>:

$$\begin{aligned} \chi_{nn}(\mathbf{k}, \omega) &= \frac{\rho}{m^2} \left( \frac{\partial \rho}{\partial p} \right)_T \left\{ \frac{(1-1/\gamma)\lambda k^2}{-i\omega + \lambda k^2} - \frac{1}{\gamma} \frac{c^2 k^2 + i\omega(\gamma-1)\lambda k^2}{\omega^2 - c^2 k^2 + i\omega\Gamma k^2} \right\}, \\ \chi_{qn}(\mathbf{k}, \omega) &= \chi_{nq}(\mathbf{k}, \omega) = \frac{T}{m} \left( \frac{\partial \rho}{\partial T} \right)_p \\ &\times \left\{ \frac{\lambda k^2}{-i\omega + \lambda k^2} - \frac{i\omega\lambda k^2}{\omega^2 - c^2 k^2 + i\omega\Gamma k^2} \right\}, \\ \chi_{qv}(\mathbf{k}, \omega) &= \kappa T k^2 / (-i\omega + \lambda k^2), \\ \chi_{t_{\alpha\beta}}(\mathbf{k}, \omega) &= (\delta_{\alpha\beta} - k_\alpha k_\beta / k^2) \chi_t(\mathbf{k}, \omega), \\ \chi_t(\mathbf{k}, \omega) &= \eta k^2 / (-i\omega + \nu k^2), \end{aligned} \quad (1.4)$$

where we have used the usual notation for the thermodynamic parameters and kinetic coefficients (cf. (3.15)); the subscripts  $n$ ,  $t_\alpha$ , and  $q$  correspond to the particle-number density, the transverse component of the momentum density, and the thermal-energy density, defined by the relation

$$q(\mathbf{r}, t) = h(\mathbf{r}, t) - m(\epsilon + p)n(\mathbf{r}, t) / \rho, \quad (1.5)$$

where  $\epsilon$  and  $\rho$  are the equilibrium values of the internal-energy and mass densities;  $m$  is the mass of a molecule of the liquid,  $p$  is the pressure and  $c^2 = \gamma(\partial p / \partial \rho) T$ .

## 2. GENERAL PROPERTIES OF THREE-POINT DYNAMICAL CORRELATION FUNCTIONS

We assume that the system under consideration moves with constant velocity  $\mathbf{u}$ , and introduces the in-

tegral dynamical variables of the conserved quantities:

$$X_a = \int dr x_a(r, t), \quad a=n, \alpha, e, \quad (2.1)$$

i.e., the particle-number  $N$ , momentum  $P_\alpha$  and energy

$$E = \int dr e(r, t) = H_0 - \mu N + m N u^2 / 2 - u P, \quad (2.2)$$

where  $H_0$  is the Hamiltonian of the system and  $\mu$  is the chemical potential in the rest ensemble ( $u = 0$ ). In the following, an average over this  $w_0$ -ensemble will be denoted by  $\langle \dots \rangle_0$ :

$$e = \langle h \rangle_0, \quad \rho = m \langle n \rangle_0 = m \langle n \rangle.$$

The brackets  $\langle \dots \rangle$  denote an average over the  $w$ -ensemble with distribution function

$$w = \exp \{ (\Omega - E) / k_B T \},$$

where  $\Omega = -pV$  and  $V$  is the volume of the system.

From the definition of the  $w$ -ensemble average we obtain the following relation<sup>[6]</sup>:

$$\langle R \delta X_a \rangle = \partial \langle R \rangle / \partial y_a, \quad (2.3)$$

where  $R$  is an arbitrary dynamical variable;  $\delta X_a \equiv X_a - V \langle x_a \rangle$ ,  $\delta X_\alpha = P_\alpha - m N u_\alpha$ ;  $y_n = \mu / k_B T$ ,  $y_\alpha = u_\alpha / k_B T$ ,  $y_e = -1 / k_B T$ . The quantity  $x_e(r, t)$  or  $X_e$  conjugate to  $y_e$  is  $e(r, t)$  or  $E$ , respectively (see (2.2)).

We introduce the three-point structure factors

$$S_{abc}(k_1, k_2) = \int \int d\omega_1 d\omega_2 G_{abc}(1, 2). \quad (2.4)$$

Putting  $k_2 \rightarrow 0$  in the left-hand side of (1.1), because  $X_a$  is independent of  $t$  by (2.1), we obtain

$$\lim_{k_2 \rightarrow 0} G_{abc}(1, 2) = 2\pi \delta(\omega_2) \langle \delta x_a^*(1) \delta x_b(1) \delta X_c \rangle. \quad (2.5)$$

A symmetric relation is obtained in the limit  $k_1 \rightarrow 0$ . If in (2.5) we go over to the limit  $k_1 \rightarrow 0$ , we shall have

$$\lim_{k_1, k_2 \rightarrow 0} G_{abc}(1, 2) = S_{abc}(0, 0) \delta(\omega_1) \delta(\omega_2). \quad (2.6)$$

The limiting properties (2.5) and (2.6) hold only for  $x_a$ ,  $x_b$  and  $x_c$  that are densities of conserved quantities. The analogous property of "contraction" of the spectrum in the long-wavelength limit is also possessed by bilinear fluctuations of the densities of constants of the motion<sup>[1,2]</sup>. Using (2.3) with  $R = \delta x_a^*(1) \delta x_b(1)$  and  $R = \delta x_a(r, t) \delta X_b$ , respectively, we obtain another form of the relations (2.5), (2.6):

$$\lim_{k_2 \rightarrow 0} G_{abc}(1, 2) = 2\pi \frac{\partial G_{ab}(1)}{\partial y_c} \delta(\omega_2), \quad (2.7)$$

$$\lim_{k_1, k_2 \rightarrow 0} G_{abc}(1, 2) = (2\pi)^2 \frac{\partial^2 \langle x_a \rangle}{\partial y_b \partial y_c} \delta(\omega_1) \delta(\omega_2), \quad (2.8)$$

where  $G_{ab}(1) \equiv \langle \delta x_a^*(1) \delta x_b(1) \rangle$ . In deriving (2.8) we have used the relation

$$\lim_{k_1 \rightarrow 0} G_{ab}(1) = 2\pi \frac{\partial \langle x_a \rangle}{\partial y_b} \delta(\omega_1), \quad (2.9)$$

which follows from (2.3) with  $R = x_a(r, t)$ <sup>[2]</sup>.

If one or all three of the dynamical variables  $x_a$ ,  $x_b$ ,  $x_c$  change sign under time reversal ( $\theta_a \theta_b \theta_c = -1$ ), then, obviously,  $S_{abc}(0, 0) = 0$  and the corresponding long-wavelength limit is equal to zero. Thus, e.g.,

$$\lim_{k_1, k_2 \rightarrow 0} G_{n\alpha\alpha}(1, 2) = \lim_{k_1, k_2 \rightarrow 0} G_{\alpha\beta\gamma} = 0, \quad (2.10)$$

$$\lim_{k_1, k_2 \rightarrow 0} G_{\alpha\beta n}(1, 2) = (2\pi)^2 \delta_{\alpha\beta} (k_B T)^2 \left( \frac{\partial \rho}{\partial \mu} \right)_{r,v} \delta(\omega_1) \delta(\omega_2). \quad (2.11)$$

In a classical liquid the functions  $G_{abc}(1, 2)$  possess

regular limit values as  $k_1, k_2 \rightarrow 0$ , and it is therefore easy to go over from the  $(k, \omega)$ - to the  $(r, t)$ -representation and obtain the following sum rules for the dynamical correlation functions of fluctuations of constants of the motion:

$$\int dr_2 G_{abc}(r_1, t_1; r_2, t_2) = \frac{\partial G_{ab}(r_1, t_1)}{\partial y_c}, \quad (2.12)$$

$$\int \int dr_2 dr_1 G_{abc}(r_1, t_1; r_2, t_2) = \frac{\partial^2 \langle x_a \rangle}{\partial y_b \partial y_c}. \quad (2.13)$$

In particular,<sup>[6]</sup>

$$\int dr_2 G_{nnn}(r_1, t_1; r_2, t_2) = k_B T \left( \frac{\partial G_{nn}(r_1, t_1)}{\partial \mu} \right)_{r,v}; \quad (2.14)$$

$$\int \int dr_1 dr_2 G_{\alpha\beta\gamma}(r_1, t_1; r_2, t_2) = 0. \quad (2.15)$$

### 3. DETERMINATION OF THE THREE-INDEX SUSCEPTIBILITIES

Using the equations of motion for the dynamical variables and the symmetry property<sup>[7]</sup>

$$\chi_{abc}(1, 2) = \chi_{acb}(2, 1), \quad (3.1)$$

it is easy to show that, of all the susceptibilities  $\chi_{abc}$  corresponding to densities of constants of the motion, only 18 are independent. For the latter, it is convenient to choose susceptibilities with 18 independent combinations of the indices  $a, b, c = n, q, t_\alpha$ . From these it is easy to construct the susceptibilities corresponding to the densities of all the other conserved quantities. For example, for the longitudinal component of the momentum density (index  $l_\alpha$ ), we have

$$\chi_{l_\alpha l_\beta l_\gamma}(1, 2) = m^2 \omega_\alpha \omega_\beta \omega_\gamma \frac{k_{0\alpha} k_{1\beta} k_{2\gamma}}{k_\alpha^2 k_\beta^2 k_\gamma^2} \chi_{nnn}(1, 2). \quad (3.2)$$

If one index is equal to  $h$  and the others are arbitrary, then

$$\chi_{hbc}(1, 2) = \chi_{ghc}(1, 2) + m(\varepsilon + p) \chi_{nbc}(1, 2) / \rho. \quad (3.3)$$

In accordance with the above-indicated choice of basis susceptibilities, we choose the particle-number density  $n(r, t)$ , the thermal-energy density  $q(r, t)$  and the transverse component  $g_t(r, t)$  of the momentum density as the independent variables in the hydrodynamic model. The variables  $h(r, t)$ ,  $p(r, t)$  and  $T(r, t)$  will be assumed throughout to be expressed in terms of  $n(r, t)$  and  $q(r, t)$ .

The complete system of hydrodynamic equations in the presence of external forces has, in these variables, the form

$$\frac{\partial n}{\partial t} + \frac{1}{m} \operatorname{div} g = 0, \quad (3.4)$$

$$\frac{\partial g_\alpha}{\partial t} + \frac{\partial p}{\partial r_\alpha} + \frac{\partial}{\partial r_\beta} \left( \frac{g_\alpha g_\beta}{mn} \right) - \frac{\partial}{\partial r_\beta} \left[ \eta \frac{\partial}{\partial r_\alpha} \left( \frac{g_\beta}{mn} \right) \right] + \eta \frac{\partial}{\partial r_\beta} \left( \frac{g_\alpha}{mn} \right) - \frac{\partial}{\partial r_\alpha} \left[ \left( \zeta - \frac{2}{3} \eta \right) \frac{\partial}{\partial r_\beta} \left( \frac{g_\beta}{mn} \right) \right] = F_\alpha, \quad (3.5)$$

$$\frac{\partial q}{\partial t} + \frac{g_\alpha}{mn} \frac{\partial q}{\partial r_\alpha} - \frac{\partial}{\partial r_\alpha} \left[ \kappa \left( \frac{\partial T}{\partial \rho} \right) \frac{m \partial n}{\partial r_\alpha} + \kappa \left( \frac{\partial T}{\partial q} \right) \frac{\partial q}{\partial r_\alpha} \right] - \frac{\eta}{2} \left[ \frac{\partial}{\partial r_\alpha} \left( \frac{g_\beta}{mn} \right) + \frac{\partial}{\partial r_\beta} \left( \frac{g_\alpha}{mn} \right) \right]^2 - \left( \zeta - \frac{2}{3} \eta \right) \left[ \frac{\partial}{\partial r_\gamma} \left( \frac{g_\gamma}{mn} \right) \right]^2 = F_q. \quad (3.6)$$

Here  $s$  is the entropy density ( $ds = dq/T$ );  $\eta$  and  $\zeta$  are the shear- and bulk-viscosity coefficients and  $\kappa$  is the thermal conductivity coefficient. The forces  $F$  and  $F_q$  are represented in the form

$$F = \nabla \{ \psi_n f_n + \psi_q f_q \} + \psi_i f_i, \quad (3.7)$$

$$F_q = \varphi_n f_n + \varphi_q f_q,$$

$$f_a(\mathbf{r}, t) = f_a(\mathbf{k}, \omega) \exp(i\mathbf{k}\mathbf{r} - i\omega t), \quad a = n, q, t_a, \quad (3.8)$$

where the  $f$  are harmonic external forces, chosen such that the Hamiltonian of the perturbation is written in the form

$$H_p(t) = - \sum_a \int d\mathbf{r} x_a(\mathbf{r}, t) f_a(\mathbf{r}, t). \quad (3.9)$$

The operator coefficients in (3.7) have a simple form<sup>[1,2]</sup>, e.g.,  $\psi_n = \rho/m$ ,  $\psi_1 = -\eta \nabla^2$ , etc.

We shall assume all the quantities  $f_a$  to be small and of the same order of smallness. Taking  $\langle \mathbf{g} \rangle = 0$  and  $\langle \mathbf{n} \rangle_0, \langle \mathbf{q} \rangle_0$  to be constants in the initial approximation, we can then construct the solutions of Eqs. (3.4)–(3.6) in the form of a perturbation-theory series

$$x_a(\mathbf{r}, t) = \langle x_a(\mathbf{r}, t) \rangle_0 + x_a^{(1)}(\mathbf{r}, t) + x_a^{(2)}(\mathbf{r}, t) + \dots \quad (3.10)$$

in increasing powers of  $f_a$ . In the linear approximation we obtain

$$x_a^{(1)}(\mathbf{r}, t) = \sum_b \chi_{ab}(1) f_b(1) \exp(i\mathbf{k}_1 \mathbf{r} - i\omega_1 t) \quad (3.11)$$

with the well-studied susceptibilities  $\chi_{ab}$  (cf. Sec. 1). In the next approximation the role of the external forces will be fulfilled by the bilinear combinations of the solutions (3.11) that arise from the nonlinear terms in the original equations (3.4)–(3.6).

The cumbersome calculation is analogous to that specially performed for a dilute gas in<sup>[8]</sup>. After the appropriate symmetrization over the Fourier arguments the result can be represented in terms of the three-index susceptibilities:

$$x_a^{(2)}(\mathbf{r}, t) = \sum_{b,c} \chi_{abc}(1, 2) f_b(1) f_c(2) \exp\{i[(\mathbf{k}_1 + \mathbf{k}_2)\mathbf{r} - (\omega_1 + \omega_2)t]\}. \quad (3.12)$$

In this case, explicit expressions are obtained for all the  $\chi_{abc}$  in terms of the  $\chi_{ab}$  and the parameters of the starting equations.

The results for  $\chi_{abc}(1, 2)$  turn out to be very unwieldy, and we shall not give them all. The simplest expression is obtained for the triply-transverse susceptibility and has the form

$$\chi_{t_a t_b t_c}(1, 2) = \chi_{t_a}^*(0) \chi_{t_b}(1) \chi_{t_c}(2) (2i\rho\eta k_0)^{-1} \times \{(\delta_{ab} - k_{0a}k_{0b}/k_0^2) k_{0c}/k_0 + (\delta_{at} - k_{0a}k_{0t}/k_0^2) k_{0c}/k_0\}, \quad (3.13)$$

where  $\chi_t$  is given in (1.4). The character, reflected here, of the relationship between the three- and two-index susceptibilities is preserved in all the other cases.

Of most physical interest is the susceptibility  $\chi_{nnn}$ , which is found to be equal to

$$\chi_{nnn}(1, 2) = \chi_{nn}^*(0) \cdot [A_n(1, 2) \chi_{nn}(1) \chi_{nn}(2) + B_n(1, 2) \chi_{nn}(1) \chi_{nn}(2) + B_n(2, 1) \chi_{nn}(1) \chi_{nn}(2) + C_n(1, 2) \chi_{nn}(1) \chi_{nn}(2)], \quad (3.14a)$$

where  $\chi_{nn}$  and  $\chi_{qn}$  are defined in (1.4). The coefficients  $A_n, B_n$  and  $C_n$  describe the interaction of the simple modes and are equal to

$$A_n(1, 2) = -\frac{m^2}{2\rho} \left\{ \frac{2\varepsilon_{1,0}\varepsilon_{2,0}}{\rho k_0^2} \omega_1 \omega_2 - \frac{iD_s}{\rho} (\omega_2 \varepsilon_{2,0} + \omega_1 \varepsilon_{1,0}) + m \left( \frac{\partial^2 p}{\partial \rho^2} \right)_s - i \left[ \omega_2 \left( \frac{\partial \Lambda(2, 0)}{\partial \rho} \right)_s + \omega_1 \left( \frac{\partial \Lambda(1, 0)}{\partial \rho} \right)_s \right] - \frac{im\kappa}{\rho D^*(0)} \left( \frac{\partial T}{\partial \rho} \right)_s \left[ -\omega_1 \omega_2 \frac{k_1 k_2}{k_1^2 k_2^2} + (\omega_1 \varepsilon_{1,0} + \omega_2 \varepsilon_{2,0}) \left( \frac{\partial p}{\partial \rho} \right)_s \right] - \frac{m\kappa}{\rho D^*(0)} \left( \frac{\partial T}{\partial \rho} \right)_s \left[ \nu (\varepsilon_{1,0}\varepsilon_{2,1} + \varepsilon_{2,0}\varepsilon_{1,2} + \frac{k_0^2}{k_1^2 k_2^2} k_1 k_2) \right] \right\}$$

$$- \left( \frac{2}{3} \nu - \xi \right) (\varepsilon_{2,0} + \varepsilon_{1,0}) \left[ \omega_1 \omega_2 + \frac{m\kappa}{D^*(0)} \left( \frac{\partial T}{\partial \rho} \right)_s \left( \frac{\partial^2 T}{\partial \rho^2} \right)_s \right] k_0^2 - \frac{\kappa k_0^2}{D^*(0)} \left( \frac{\partial T}{\partial \rho} \right)_s (\varepsilon_{1,0} + \varepsilon_{2,0}) \left( \frac{\partial \kappa}{\partial \rho} \right)_s, \quad (3.14b)$$

$$B_n(1, 2) = -\frac{m^2}{2\rho} \left\{ \left( \frac{\partial^2 p}{\partial \rho \partial q} \right) - i\omega_1 \left( \frac{\partial \Lambda(1, 0)}{\partial q} \right)_p - \frac{im\kappa}{\rho D^*(0)} \omega_1 \varepsilon_{1,0} \left( \frac{\partial T}{\partial \rho} \right)_s \left[ 1 + \left( \frac{\partial p}{\partial q} \right)_p \right] + \frac{2m\kappa^2 k_0^2}{D^*(0)} \left( \frac{\partial T}{\partial \rho} \right)_s \left( \frac{\partial^2 T}{\partial \rho \partial q} \right) - \frac{m\kappa k_0^2}{D^*(0)} \left( \frac{\partial T}{\partial \rho} \right)_s \right\} \times \left[ \varepsilon_{1,0} \left( \frac{\partial T}{\partial \rho} \right)_s \left( \frac{\partial \kappa}{\partial q} \right)_p + \varepsilon_{0,2} \left( \frac{\partial T}{\partial q} \right)_p \left( \frac{\partial \kappa}{\partial \rho} \right)_s \right], \quad (3.14c)$$

$$C_n(1, 2) = -\frac{m}{2\rho} \left\{ \left( \frac{\partial^2 p}{\partial q^2} \right)_p - \frac{m\kappa k_0^2}{D^*(0)} (\varepsilon_{0,1} + \varepsilon_{0,2}) \left( \frac{\partial T}{\partial \rho} \right)_s \left( \frac{\partial T}{\partial q} \right)_p \left( \frac{\partial \kappa}{\partial q} \right)_p \right\}. \quad (3.14d)$$

Here we have used the notation

$$D(\mathbf{k}, \omega) = -i\omega + \gamma \lambda k^2, \quad \varepsilon_{i,j} = \mathbf{k}_i \mathbf{k}_j / k_i^2,$$

$$\Lambda(i, j) = 2\nu \varepsilon_{i,j} \varepsilon_{i-2} \nu + \xi, \quad i, j = 1, 2, 0,$$

and have introduced the kinetic coefficients  $\nu, \xi, \lambda, D_e$  and  $\Gamma$  by the relations

$$\eta = \rho \nu, \quad \xi = \rho \xi, \quad D_e = \nu^2 \nu + \xi, \quad \kappa = \gamma (\partial q / \partial T)_p \lambda, \quad (3.15)$$

$$\gamma = c_p / c_v, \quad \Gamma = D_e + (\gamma - 1) \lambda,$$

where  $c_p$  and  $c_v$  are the specific heats.

#### 4. THREE-POINT CORRELATIONS OF FLUCTUATIONS

A knowledge of the three-index hydrodynamic susceptibilities makes it possible to determine the dynamical three-point correlations of the fluctuations of hydrodynamic quantities for small  $\omega$  and  $\mathbf{k}$  from the Efremov-Stratonovich theorem (1.1). For the most important (the density-density-density) correlation function we obtain in this way:

$$G_{nnn}(1, 2) = -(2k_B T)^2 \mathcal{P}_C \operatorname{Re} \left\{ \frac{\chi_{nnn}(1, 2)}{\omega_1 \omega_2} \right\}, \quad (4.1)$$

where  $\mathcal{P}_C$  is the operator that sums the cyclic permutations of the symbols 0, 1 and 2, with  $\chi_{nnn}(1, 2)$  from (3.14). Hence we have, in the limit  $\mathbf{k}_1, \mathbf{k}_2 \rightarrow 0$ ,

$$\lim_{\mathbf{k}_1, \mathbf{k}_2 \rightarrow 0} G_{nnn}(1, 2) = 2\pi k_B T \left( \frac{\partial G_{nn}(1)}{\partial \mu} \right)_{T, \nu} \delta(\omega_2), \quad (4.2)$$

$$\lim_{\mathbf{k}_1, \mathbf{k}_2 \rightarrow 0} G_{nnn}(1, 2) = \frac{(2\pi k_B T)^2}{m} \left( \frac{\partial^2 \rho}{\partial \mu^2} \right)_{T, \nu} \delta(\omega_1) \delta(\omega_2) \quad (4.3)$$

in complete agreement with (2.7), (2.8). The expressions (4.2) and (4.3) and the results of integrating them over the frequencies demonstrate the non-Gaussian character of the density fluctuations. As the critical point is approached,  $G_{nnn}(1, 2)$ , like  $G_{nn}(1)$ , increases without limit and the described variant of the theory becomes inapplicable.

From (4.1) we can obtain hydrodynamic estimates of the lowest frequency moments of the function  $G_{nnn}(1, 2)$ :

$$M_{1,1}(\mathbf{k}_1, \mathbf{k}_2) = \iint_{-\infty}^{\infty} d\omega_1 d\omega_2 \omega_1^2 \omega_2^2 G_{nnn}(1, 2). \quad (4.4)$$

They turn out to be equal to

$$M_{00}(\mathbf{k}_1, \mathbf{k}_2) = \frac{(2\pi k_B T)^2}{m} \left( \frac{\partial^2 \rho}{\partial \mu^2} \right)_{T, \nu} [1 + O(k_1^2, k_2^2)], \quad (4.5)$$

$$M_{01}(\mathbf{k}_1, \mathbf{k}_2) = M_{10}(\mathbf{k}_1, \mathbf{k}_2) = 0, \quad (4.6)$$

$$M_{11}(\mathbf{k}_1, \mathbf{k}_2) = \frac{(2\pi k_B T)^2}{m^2} k_1 k_2 \left( \frac{\partial \rho}{\partial \mu} \right)_{T, \nu} [1 + O(k_1^2, k_2^2)], \quad (4.7)$$

$$M_{02}(\mathbf{k}_1, \mathbf{k}_2) = -M_{11}(\mathbf{k}_1, \mathbf{k}_2) - M_{11}(\mathbf{k}_0, \mathbf{k}_2), \quad (4.8)$$

with an analogous expression for  $M_{20}$ . In (4.5) and

(4.7) the hydrodynamically small terms have not been written out; a molecular approach to the problem is required for an exact estimate of these terms. The right-hand side of (4.5) is the hydrodynamic estimate for the three-point structure factor  $S_{nnn}(\mathbf{k}_1, \mathbf{k}_2)$  (cf. (2.4)).

An analogous simple analysis is also possible for the remaining correlation functions  $G_{abc}(1, 2)$  of fluctuations of conserved quantities.

The results obtained can be used directly in the theory of the spectra and intensity of double molecular scattering of light.

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