Multiparticle correlations in a nonequilibrium gas with pair collisions

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Pair collisions in the nonequilibrium state are known to lead to kinetic correlation of the occupation numbers. The classical diagram technique is used to investigate the correlations produced as a result of pair collisions between triads and tetrads of occupation numbers.

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1. INTRODUCTION

In 1969, in the course of development of the theory of fluctuations in a nonequilibrium stationary state, ¹⁻³ an additional or collisional correlation was observed between the occupation numbers of single-particle states; this correlation arises in a gas as a result of pair (binary) collisions and is significant just under nonequilibrium conditions.¹⁾ The theory of fluctuations in a nonequilibrium state, including these results, is expounded in reviews by Gantsevich Gurevich, and Katilyus⁶ and by Ernst and Cohen,⁷ as well as in the book by Lifshitz and Pitaevskiĭ.⁸ These results were used, in particular, to calculate the cross section for light scattering by a nonequilibrium electron gas^{3,9} (see also the review, ⁶ p. 41), and quite recently by an ordinary nonequilibrium gas.^{10,11}

As the next step in the development of a quantitative kinetic theory of fluctuations in a nonequilibrium state, it is natural to calculate the higher correlation functions and ascertain how the aforementioned collisional correlation manifests itself in the case of many particles. To derive the kinetic equations for the correlation functions we use a suitably generalized classical kinetic diagram technique, developed in Ref. 12, where the classical diagram technique, subject to satisfaction of the usual criteria for the applicability of the kinetic equation, was obtained from first principles, namely from the statistical diagram technique of Konstantinov and Perel'.¹³

For the problem posed in the present paper, namely the calculation of higher correlation functions, the classical technique developed in Ref. 12 is insufficient, since we encounter the problem of splitting the higher equal-time correlation functions into lower, but such objects—higher equal-time correlation functions—were not encountered in the problems considered in Ref. 12. We had therefore to supplement the technique of Ref. 12 with new elements corresponding to (large) diagonal terms of equal-time correlators.

We confine ourselves in this article to a detailed investigation of spatially homogeneous correlations. In particular, we take into account also the correlation that occurs between occupation number as a result of the constancy of the total number of particles in the system, a correlation first predicted for two-particle correlation functions in Ref. 14. In contrast to the correlation due to pair collisions, the additional correlation does not vanish in the equilibrium state. We regard the main result of the paper, however, to be the expressions for the nontrivial parts of equal-time correlators, viz., Eqs. (22) and (24).

We call attention to the fact that higher correlation functions lend themselves at present to measurement. Thus, the quadratic current correlator is determined by measuring the electromagnetic radiation from the sample. By measuring its fluctuations, however, rather than the average radiation flux, i.e., by directing the radiated flux to the input of the correlation meter, we determine the quaternary correlator of the currents in the sample.

2. FORMULATION OF PROBLEM

We consider for the sake of argument an electron gas in a semiconductor (or a weakly ionized gas), acted upon by a sufficiently strong electric field that causes noticeable deviations of the electron velocity distribution from equilibrium Maxwellian. The electrons collide with one another as well as with the thermostat (phonons or neutrals, assumed to be in the equilibrium state).

Our task is to investigate the manifestations of the correlation mentioned in the Introduction as applied to threeand four-particle correlation functions in a stationary nonequilibrium state of an electron gas.

Of greatest interest is the four-particle distribution function with pairwise coinciding times; the spatial Fourier transform of this function

$$F_{\mathbf{p}_{1}\mathbf{p}_{2},\mathbf{p}_{2}\mathbf{p}_{3}\mathbf{p}_{4}}^{\mathbf{q}_{1}\mathbf{q}_{3}\mathbf{q}_{3}}(t+\tau,t) = F_{12,34}^{123}(t+\tau,t)$$

$$= \int d^{3}r_{1}d^{3}r_{2}d^{3}r_{3}\exp\left(-i\mathbf{q}_{1}\mathbf{r}_{1}-i\mathbf{q}_{2}\mathbf{r}_{2}-i\mathbf{q}_{3}\mathbf{r}_{3}\right)$$

$$\times F(t+\tau,\mathbf{r}+\mathbf{r}_{1}+\mathbf{r}_{2}+\mathbf{r}_{3},\mathbf{p}_{1}|t+\tau,\mathbf{r}+\mathbf{r}_{2}+\mathbf{r}_{3},\mathbf{p}_{2}|t,\mathbf{r}+\mathbf{r}_{3},\mathbf{p}_{3}|t,\mathbf{r},\mathbf{p}_{4}),$$
(1)

is expressed in terms of the creation and annihilation operators $a_{\mathbf{p}}^+$ and $a_{\mathbf{p}}$ of electrons with momentum **p** in the following manner:

$$F_{12,34}^{123}(t+\tau,t) = (\operatorname{Sp}\rho_0)^{-1} \operatorname{Sp} \{ \rho_0 S^+(t+\tau) a_{\mathfrak{p}_1-\mathfrak{q}_1/2}^+ a_{\mathfrak{p}_1+\mathfrak{q}_1/2} a_{\mathfrak{p}_2-(\mathfrak{q}_1-\mathfrak{q}_2)/2}^+ a_{\mathfrak{p}_2+(\mathfrak{q}_1-\mathfrak{q}_2)/2} \\ \times S(t+\tau,t) a_{\mathfrak{p}_2+(\mathfrak{q}_2-\mathfrak{q}_3)/2}^+ a_{\mathfrak{p}_3-(\mathfrak{q}_2-\mathfrak{q}_3)/2} a_{\mathfrak{p}_4+\mathfrak{q}_3/2}^+ a_{\mathfrak{p}_4-\mathfrak{q}_3/2}^+ S(t) \}, \quad (2)$$

where S(t) is the evolution operator: $iS = (H_0 + \hat{V})S$,

1212 Sov. Phys. JETP 57 (6), June 1983

 $(H_0 + \hat{V})$ is the total Hamiltonian of the system, and $\hbar = 1$. Included in \hat{V} are both the (strong) electric field) and the interaction of the electrons with one another and with phonons (neutrals), the latter regarded as the thermostat; H_0 is the free-electron Hamiltonian in the absence of a field. Just as before,^{1,3} we assume that a stationary state independent of the state at t = 0 is established in the system after α sufficiently large time interval (formally, as $t \to \infty$). This allows us to choose ρ_0 in the form

$$\rho_0 = \exp\{-(H_0 - \mu N)/T\}$$
(3)

and put S(0) = 1. Here N is the particle-number operator, μ the chemical potential, and T the thermostat temperature in energy units. Since the state is stationary, the mean value in Eq. (2) is independent of t at large t, i.e.,

$$F_{12,34}^{123}(t+\tau,t) = F_{12,34}^{123}(\tau).$$
⁽⁴⁾

In (2) is used the circumstance that in a uniform electric field E the function F of (1) does not depend on r. We shall investigate the function $F_{12,34}^{123}(\tau)$, assuming satisfaction of the usual criteria of existence of a kinetic equation:

$$1/\varepsilon_{p}\tau_{e}\ll 1, \quad 1/\varepsilon_{p}\tau\ll 1, \quad q/p\ll 1, \quad eE/\varepsilon_{p}p\ll 1.$$
 (5)

Here ε_p and **p** are the characteristic energy and momentum of the electron, and τ_e is the characteristic electron relaxation time.

3. CLASSICAL DIAGRAM TECHNIQUE FOR CORRELATION FUNCTIONS

We assume the criteria (5) to be satisfied and that the kinetic equation for the single-particle distribution function of the electrons, $F_p \ll 1$ (Boltzmann statistics), has been derived:

$$(e\mathbf{E}\partial_{\mathbf{p}}+I_{\mathbf{p}}^{th})F_{\mathbf{p}}+I_{\mathbf{p}}\{F, F\}=0.$$
(6)

Here I_p^{th} is the operator of electron collisions with the thermostat; $I_p \{F,F\}$ is the electron-electron collision term. We can use next the classical diagram technique proposed in Ref. 12. It is then necessary to process additionally the equal-time correlation functions and separate in them the terms with equal momenta.

In the classical diagram technique, the double ordering typical of the quantum diagram technique,¹³ in time and "by contour," is replaced by ordering in time only. The two propagator lines representing the observable merge into a single classical propagator.²⁾ Figure 1 shows two such lines that merge into a single distribution-function line. We regard this line as saturated by the interaction with the field and by the collisions with the thermostat and with the remaining electrons. Diagrams representing higher correlation functions have several propagator lines. These lines are interconnected by binary electron-electron collisions. Such a connection is shown in Fig. 2a. The vertical line corresponds to instantan-



FIG. 1.



FIG. 2.

eous momentum exchange during the time of the collision, with the momenta \mathbf{p} and \mathbf{p}' going over into the momenta \mathbf{p}_1 and \mathbf{p}_2 . Connecting from the left the distribution functions $F_{\mathbf{p}}$ and $F_{\mathbf{p}'}$ and summing over \mathbf{p} and \mathbf{p}' (the summation is over all the inner lines) we obtain the diagram 2b. The vertical connection together with the lines to the left of it corresponds to a paired collision term without one summation: I_{12} {F,F}. This is precisely the term that produces the collisional (additional) correlation in the nonequilibrium state, observed in Refs. 1-3 (I_{12} {F,F} = 0 at equilibrium). Summing over \mathbf{p}_2 (or \mathbf{p}_1) we obtain a diagram with one output line, shown in Fig. 2c, namely the electron-electron collision term of the kinetic equation I_1 {F,F}.

Corresponding to the propagator lines (e.g., to the lines on the right of the connection on Fig. 2b) are propagator denominators which we shall take into account in accord with rule for "vertical section' through the diagram (cf. Ref. 13). The section to the right of the connection on Fig. 2b corresponds to the denominator $(J_1 + J_2)^{-1}$, where J is the linearized kinetic-equation operator:

$$J_{1} = e E \partial_{p_{1}} + I_{p_{1}} {}^{th} + I_{p_{1}} \{F\},$$

$$J_{p} \{F\} X = I_{p} \{F, X\} + I_{p} \{X, F\}.$$
 (7)

The analytic expression for diagram 2b is thus

$$\varphi_{12} = -(J_1 + J_2)^{-1} I_{12} \{F, F\}, \quad \varphi_{ii} = 0.$$
 (8)

Unequal-time distribution functions are represented by diagrams with lines that extend to the right to different distances i.e., that bear against "terminals" located at different points on the time axis. For example, the unequal-time twoparticle distribution function

$$F_{i,2}(\tau) = (\operatorname{Sp} \rho_0)^{-1} \operatorname{Sp} \{ \rho_0 S^+(t+\tau) a_1^+ a_1 S(t+\tau, t) a_2^+ a_2 S(t) \},$$
(9)

corresponds to the sum of diagrams of Fig. 5. In noncoupled diagrams, the vertical section should be drawn for each part



FIG. 3.



FIG. 4.

of the diagram separately. Corresponding to diagram a is $(-i\omega)^{-1}F_1F_2$. In the analytic expression for diagram b, the denominator corresponding to the section between t and $t + \tau$, i.e., $(-i\omega + J_1)^{-1}$, is followed by a two-particle correlator with equal times, which stems from the same diagram, when there are no points of interaction between t and $t + \tau$ (in the approximation (5) assumed by us). In the corresponding correlator

$$g_{12}(\tau=0) = (\operatorname{Sp} \rho_0)^{-1} \operatorname{Sp} \{ \rho_0 S^+(t) a_1^+ a_1 a_2^+ a_2 S(t) \} - F_1 F_2, \quad (10)$$

it is necessary to separate specially the term with the coinciding momenta, i.e., to commute a_1 and a_2^+ . The term $\delta_{21}F_1$ that appears after the commutation can be graphically represented as the result of merging of two classical lines into one. We emphasize that this merging should be carried out only if the terminals (right-hand end) one of the merging lines is below (above) the other line. This remark is important when more complicated diagrams are considered. Corresponding to the correlator (9) is the sum of diagrams in Fig. 4:

$$g_{12}(\tau=0) = \varphi_{12} + \delta_{21} F_1. \tag{11}$$

In the diagrams 5a and 4a, the momenta \mathbf{p}_1 and \mathbf{p}_2 are not equal (but this is immaterial in the framework of Boltzmann statistics).

The unequal-time two-particle correlator $g_{12}(\omega)$, i.e., the connected part of the diagram of Fig. 5, is thus expressed by the sum of diagrams in Fig. 6.

Figures 5 and 6 correspond to the analytic expression

$$\mathbf{F}_{i,2}(\omega) = (-i\omega)^{-i} F_i F_2 + (-i\omega + J_1)^{-i} (\varphi_{12} + \delta_{21} F_1), \quad (12)$$

where $F(\omega)$ is a one-sided Fourier transform with respect to τ :

$$F(\omega) = \int_0^\infty F(\tau) \exp(i\omega\tau) d\tau.$$
(13)

Figure 6 means that there exist two types of coupling: via pair collisions and as a result of commutation. In more complicated cases it is necessary to merge all the lines by all possible methods and thus write out all the obtained diagrams with couplings due to pair collisions and to commutation.

4. FOUR-PARTICLE SPATIALLY HOMOGENEOUS CORRELATOR WITH PAIRWISE COINCIDING TIMES

By way of example of the application of the foregoing rules for the calculation of higher propagators by using clas-



FIG. 5.



FIG. 6.

 $F_{12, 34}(\tau)$

sical diagram technique, we calculate the spatially homogeneous four-particle distribution function with pairwise coinciding times:

$$= (\operatorname{Sp} \rho_0)^{-1} \operatorname{Sp} \{ \rho_0 S^+(t+\tau) a_1^+ a_1 a_2^+ a_2 S(t+\tau, t) a_3^+ a_3 a_4^+ a_4 S(t) \}.$$
(14)

It corresponds to the sum of diagrams of Fig. 3. Diagrams b and c constitute each several diagrams of the same type. Different groups of diagrams correspond to terms of different order in 1/N, where N is the total number of electrons in the system. The diagram of Fig. 3a corresponds to a product of our single-particle functions

$$(-i\omega)^{-1}F_{4}F_{2}F_{3}F_{4}.$$
 (15)

Diagrams b correspond to terms of order 1/N compared with diagram a:

$$(-i\omega)^{-1} [F_1 F_2 g_{34} (\tau=0) + F_3 F_4 g_{12} (\tau=0)] + F_2 F_4 g_{1,3} (\omega) + F_2 F_3 g_{1,4} (\omega) + F_1 F_4 g_{2,3} (\omega) + F_1 F_3 g_{2,4} (\omega).$$
(16)

Diagrams c correspond to terms of order $1/N^2$:

$$F_{4}g_{12, 3}(\omega) + F_{3}g_{12, 4}(\omega) + F_{2}g_{1, 34}(\omega) + F_{1}g_{2, 34}(\omega), \quad (17)$$

$$(-i\omega)^{-1}g_{12}(\tau=0)g_{34}(\tau=0)+g_{1,3}(\omega)g_{2,4}(\omega)+g_{1,4}(\omega)g_{2,3}(\omega),$$
(18)

where $g_{ij,k}$ are three-particle correlators with two coinciding times:

$$g_{ij,k}(\tau) = F_{ij,k}(\tau) - F_i F_j F_k - F_k g_{ij}(\tau=0) - F_j g_{i,k}(\tau) - F_i g_{j,k}(\tau).$$
(19)

Finally, diagram d corresponds to a term of order $1/N^3$, namely the four-particle correlator $g_{12,34}$ with pairwise coinciding times:

$$g_{12, 34}(\tau) = F_{12, 34}(\tau) - [Eqs. (15)-(19)].$$
 (20)

We begin with calculation of the three-particle correlator $g_{1,23}(\omega)$ —the connected part of the diagram of Fig. 3c1. Figure 7 shows its constitutent diagrams. They correspond to the analytic expression³⁾

$$g_{1,23}(\omega) = (-i\omega + J_1)^{-1} [\delta_{21}\varphi_{13} + \delta_{32}\varphi_{13} + \delta_{31}\varphi_{12} + \delta_{32}\delta_{21}F_1 - I_1 \{g_{i,2}(\omega)\}g_{k,3}(\omega) + \varphi_{123}], \qquad (21)$$

where φ_{123} is the equal-time part of Fig. 7a and is shown in Fig. 8. We call attention to the appearance of diagrams 7c, 7d, and 8b. The first is the result of the merging of both lines 2 and 3 with line 1. Diagrams 7d and 8b result from multiplication of the electron lines in pair collision—cf. Fig. 2c. Altogether there are six each of diagrams 8a and 8b. The analytic expression⁴⁾ for φ_{123} is the following:

$$\varphi_{123} = -(J_1 + J_2 + J_3)^{-1} \sum_{P} (I_2 I_{12} \{F\} \varphi_{i3} + I_1 \{\varphi_{i2}, \varphi_{k3}\}), \quad (22)$$

1214 Sov. Phys. JETP 57 (6), June 1983



with $\varphi_{123} = \varphi_{231} = \varphi_{312} = \dots$ etc., and the summation is over the permutations of the indices 1, 2, and 3.

For diagram 3d, of the four-particle correlator $g_{12,34}(\omega)$ we have analogously the diagrams of Fig. 9 (two each of type b, four of d2, two each of e and three each of f). The corresponding analytic expressions are:

$$g_{12, 34}(\omega) = \delta_{21}g_{1, 34}(\omega)$$
 (a)

$$+(-i\omega+J_1+J_2)^{-1}\{-I_{12}\{F\}g_{i,34}(\omega)$$
 (b1)

$$-I_{12}\{g_{1,3}(\omega)\}g_{k,4}(\omega)$$
 (b2)

$$+\varphi_{1234}+$$
 (c)

$$+ (\delta_{31} + \delta_{32}) \varphi_{124} + (\delta_{41} + \delta_{42} + \delta_{43}) \varphi_{123} \tag{d}$$

$$+ \left(\delta_{41} + \delta_{42} \right) \left(\delta_{31} + \delta_{32} \right) \varphi_{12} \tag{e}$$

$$-(I_{i}\{\varphi_{2k}\}g_{i,\ 34}(\omega)+I_{i}\{g_{i,\ 3}(\omega)\}g_{k2,\ 4}(\omega)$$

$$+I_{1}\{g_{i,4}(\omega)\}g_{k2,3}(\omega)\}_{k\neq 2}-$$
 (f1)

$$-(I_2\{\varphi_{ik}\}g_{i,34}(\omega)$$

$$+I_{2}\{g_{i,3}(\omega)\}g_{k1,4}(\omega)+I_{2}\{g_{i,4}(\omega)\}g_{k1,3}(\omega)\}_{k\neq 1}\}, \quad (f2)$$

where φ_{1234} is that part of the equal-time four-particle correlator which is connected via pair collisions, i.e., the equaltime part of Fig. 9c is represented in Fig. 10. Its analytic expression is

$$\varphi_{1234} = -(J_1 + J_2 + J_3 + J_4)^{-4} \times \sum_{P} ({}^{1}/_{4}I_{12} \{F\} \varphi_{i34} + {}^{1}/_{2}I_{12} (\varphi_{i3}, \varphi_{k4}) + {}^{1}/_{2}i_1 \{\varphi_{i2}\} \varphi_{k34}).$$
(24)

Obviously, the same method can be used if necessary to calculate also correlators of higher order and/or with larger numbers of unequal times.





FIG. 9.

5. ALLOWANCE FOR THE CONSTANCY OF THE NUMBER OF PARTICLES

So far the averaging at the initial instant of time was over a grand canonical ensemble. Since the number of electrons in the system at the initial instant of time is assumed given and not altered by the interaction and by the action of the field, an appropriate condition must be imposed on the results, namely Eqs. (21) and (23). The absence of fluctuations of the total number of electrons calls for vanishing of the summation, over any of the momenta, of the expressions

$$\langle \delta F_1 \delta F_2 \delta F_3 \ldots \rangle$$

where

$$\delta F_1 = S^+(t) a_1^+ a_1 S(t) - (\operatorname{Sp} \rho_0)^{-1} \operatorname{Sp} \{ \rho_0 S^+(t) a_1^+ a_1 S(t) \}$$

Direct calculation yields

$$\langle \delta F_1 \delta F_2 \delta F_3 \rangle = \varphi_{123} + \delta_{21} \varphi_{13} + \delta_{31} \varphi_{12} + \delta_{32} \varphi_{12} + \delta_{32} \delta_{21} F_1 = g_{123} (\tau=0)$$

$$(25)$$

$$\langle \delta F \delta F \delta F \delta F_2 \rangle = g_{123} (\tau=0) + g_{12} (\tau=0) g_{21} (\tau=0)$$

$$+g_{13}(\tau=0)g_{24}(\tau=0)+g_{14}(\tau=0)g_{23}(\tau=0), \qquad (26)$$

where the equal-time four-particle correlator is



FIG. 10.

1215

$$g_{1234}(\tau=0) = \delta_{21}g_{134}(\tau=0) + (\delta_{31}+\delta_{32})\phi_{124} + (\delta_{41}+\delta_{42}+\delta_{43})\phi_{123} + (\delta_{41}+\delta_{42})(\delta_{31}+\delta_{32})\phi_{12} + \phi_{1234}.$$
(27)

As shown in Ref. 6 (see also Refs. 1 and 3), constancy of the number of particles in the system leads to replacement of φ_{12} by

$$\tilde{\varphi}_{12} = \varphi_{12} - N \partial_N F_1 \partial_N F_2, \qquad \sum_{\mathbf{z}} \tilde{\varphi}_{12} = -F_1.$$
(28)

In exactly the same manner, it is necessary to add to the solution φ of Eq. (22) a particular solution of the corresponding homogeneous equation. The constant in front of this term is determined precisely from the condition that there be no fluctuations of the total number of particles:

$$\tilde{\varphi}_{123} = -(J_1 + J_2 + J_3)^{-1} \times \sum_{\mathbf{p}} ({}^{4}/{}_{2}I_{12} \{F\} \tilde{\varphi}_{13} + I_1 \{\tilde{\varphi}_{12}, \tilde{\varphi}_{k3}\}) + A \partial_N F_1 \partial_N F_2 \partial_N F_3,$$

$$A = 2N.$$
(29)

We recall that the inverse regression operators were chosen such that 2,6

$$\sum_{i} J_{i}^{-i} X_{i} = 0, \qquad \sum_{i} X_{i} = 0;$$

$$\sum_{i} (J_{i} + J_{2})^{-i} X_{i2} = J_{i}^{-i} \sum_{i} X_{i2}, \qquad \sum_{i,2} X_{i2} = 0;$$

$$\sum_{i} (J_{i} + J_{2} + J_{3})^{-i} X_{i23} = (J_{1} + J_{2})^{-i} \sum_{3} X_{i23},$$

$$\sum_{i,2,3} X_{i23} = 0 \text{ etc.}$$
(30)

Using (30), we rewrite (29):

$$\tilde{\varphi}_{123} = \varphi_{123} - \frac{1}{2}N \sum_{P} (\partial_{N}F_{1}\partial_{N}\tilde{\varphi}_{23}) - N\partial_{N}(N\partial_{N}F_{1}\partial_{N}F_{2}\partial_{N}F_{3}),$$
(31)
$$\sum_{3} \tilde{\varphi}_{123} = -2\tilde{\varphi}_{12}.$$
(32)

In exactly the same manner

$$\widetilde{\varphi}_{1234} = \varphi_{1234} - N \sum_{P} \left(\frac{1}{6} \partial_{N} F_{1} \partial_{N} \widetilde{\varphi}_{234} + \frac{1}{8} \partial_{N} \widetilde{\varphi}_{12} \partial_{N} \widetilde{\varphi}_{34} \right. \\ \left. + \frac{1}{4} \partial_{N} \left(N \partial_{N} F_{1} \partial_{N} F_{2} \partial_{N} \widetilde{\varphi}_{34} \right) \right)$$

$$(33)$$

$$-N\partial_{N} [N\partial_{N} (\partial_{N}F_{1}\partial_{N}F_{2}\partial_{N}F_{3}\partial_{N}F_{4})],$$

$$\sum_{\mathbf{4}} \tilde{\varphi}_{1234} = -3\tilde{\varphi}_{123}.$$
(34)

Thus, at a constant number of particles in the system it is necessary to replace in (21)–(24) the φ by the $\tilde{\varphi}$ given by Eqs. (28), (31), and (33).

If the conditions of the problem are such that pair collisions can be neglected, the correlation between the occupation numbers of the states is produced only by the constancy of the total number of particles in the system. The equal-time correlators in (25) and (27) take the form

$$\tilde{g}_{123}(\tau=0) = \delta F_1 \delta F_2 \delta F_3|_{N=\text{const}} = \delta_{32} \delta_{21} F_1 - (\delta_{21} F_3 + \delta_{31} F_2 + \delta_{32} F_3) F_1 / N + 2F_1 F_2 F_3 / N^2,$$
(35)

$$\tilde{g}_{1234}(\tau=0)$$

$$= \left[\langle \delta F_1 \delta F_2 \delta F_3 \delta F_4 \rangle - \frac{1}{s} \sum_{P} \langle \delta F_1 \delta F_2 \rangle \langle \delta F_3 \delta F_4 \rangle \right]_{N=\text{const}}$$

$$= \delta_{43} \delta_{32} \delta_{21} F_1 + \delta_{21} \left[2F_1 F_3 F_4 / N^2 - F_1 F_4 \delta_{32} / N - (\delta_{42} + \delta_{43}) F_1 F_3 / N \right]$$

$$+ 2 \left[(\delta_{31} + \delta_{32}) F_1 F_2 F_4 + (\delta_{44} + \delta_{42} + \delta_{43}) F_1 F_2 F_3 \right] / N^2 \qquad (36)$$

$$- (\delta_{44} + \delta_{42}) (\delta_{31} + \delta_{32}) F_1 F_2 / N.$$

We recall that (see, e.g., Refs. 14 and 1)

$$\tilde{g}_{12}(\tau=0) = \langle \delta F_1 \delta F_2 \rangle_{N=\text{const}} = \delta_{21} F_1 - F_1 F_2 / N.$$
(37)

Equations (35)-(37) describe also correlations in a gas with pair collisions but at equilibrium.

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¹⁾The idea of the existence of a correlation in an equilibrium gas with pair collisions, at distances large compared with the radius of the intermolecular forces, was advanced even earlier by M. S. Green⁴ and G. Ludwig.⁵ ²⁾We mean here a diagram technique for classical particles. The diagram

technique for classical waves is differently organized. ³⁾A similar result for a gas of uncharged particles is contained in Ref. 7.

⁴No expression is given in Ref. 7 for the equal-time four-particle correlator, apparently because of the unwieldiness of the calculation of such quantities within the framework of the BBGKY method.