Contribution of One-Time Pair Correlation Function to Kinetic Phenomena in Nonequilibrium Gas

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Abstract
It has been established in nineteen seventies that in nonequilibrium case the pair collisions generate non-zero two-particle correlations which are non-diagonal in momentum space and give the essential contribution to the current fluctuations of hot electrons. It is shown here that this correlations give also a contribution to the collision integral, i.e., to kinetic properties of nonequilibrium gas. The expression for electron energy loss rate \( P \) via phonons is re-derived in detail from this point of view. The contribution of the non-diagonal part of the nonequilibrium pair correlator to phonon-electron collision integral and to \( P \) is obtained and explicitly calculated in the electron temperature approximation. It is shown that these results can be obtained from stochastic non-linear kinetic equation with Langevin fluctuation force. Such an approach allows to formulate the simple general conditions under that a contribution of two-particle correlations might be essential in kinetics. The contribution obtained does not contain the extra powers of small gas parameter unlike the equilibrium virial decompositions.

1 Introduction
The derivation of kinetic equation for one-particle distribution function \( F(p, r, t) \) from BBGKY chain leads to the collision integral expressed in terms of the two-particle correlation function. The latter is in turn expressed by \( F(p, r, t) \) using Boltzmann’s Stoßzahlansatz that implies the neglect of initial one-time correlation function \( g_2(p_1, p_2, r, t) \), which is non-diagonal in the momentum space. In nonequilibrium case, however, it has been established \[1\]-\[2\] that pair collisions generate the non-diagonal two-particle correlator that gives the essential contribution to the current fluctuations of hot electrons. Here we show that this correlator gives also a contribution to the collision integral, i.e., to kinetic properties of nonequilibrium electrons. The expression for electron energy loss rate \( P \) via phonons is re-derived in detail from this point of view.

In fact, the formula for \( P \) in terms of electron density-density correlation function \( \langle \rho_k \rho_k^+ \rangle_\omega \) was obtained by Kogan \[3\] using Fermi Golden Rule. However, to come to the well-known result for \( P \) in the electron temperature approximation the fluctuation-dissipation theorem was implicated to express \( \langle \rho_k \rho_k^+ \rangle_\omega \) in terms of the dielectric function of the electron gas. This step is defensible in the case of the true equilibrium electron gas only and it gives rise to results equivalent to the Stoßzahlansatz. In this report the expression for \( P \) in terms of \( \langle \rho_k \rho_k^+ \rangle_\omega \) is derived from quantum kinetic equation for phonons interacting with nonequilibrium electron gas. The contribution of the non-diagonal part of the nonequilibrium pair correlator to phonon-electron collision integral and to \( P \) is obtained and calculated explicitly in the electron temperature approximation.

It is shown that these results may be obtained from stochastic non-linear kinetic equation with Langevin fluctuation force. Such an approach allows to formulate the simple general conditions when the two-particle correlator contribution might be essential in kinetics. The derivation of this equation based on the procedure of partial averaging over microscopic-scale fluctuations similar to the procedure using by H. Mori \[4\] to derive the generalized classical Boltzmann equation for dense gases and liquids.

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However, in our case the contribution obtained does not contain the extra powers of small gas parameter of the kinetic theory.

2 Kinetic equation for phonons

2.1 Basic designations

Hamiltonian $H = H_e + H_{ph} + V_{eph}$

Electron part $H_e = \sum_k \hbar \Omega_k a_k^\dagger a_k + V_{ee}$ where $V_{ee}$ stands for the electron-electron interaction energy

Phonon part $H_{ph} = \sum_f \hbar \omega_f b_f^\dagger b_f$

Electron-phonon interaction $V_{eph} = \frac{1}{\sqrt{\Omega}} \sum_f \left( c_f \rho_f^+ b_f + c_f^\dagger \rho_f b_f^\dagger \right)$

Spatial Fourier components of the electron density operator

$\rho_f = \sum_k a_k^{+}\rho a_{k-\delta}$, \hspace{1cm} $\rho_f^+ = \rho - f$

Phonon distribution function $N_f(t) = \langle \frac{1}{2} b_f^\dagger b_f \rangle$

Electron distribution function $F_k(t) = \langle \frac{1}{2} a_k^{+} a_k \rangle$

The angle brackets denote the full averaging with the density matrix corresponding to the noninteracting electrons and phonons at an initial instant of time. The subscript $t$ indicates the Heisenberg representation for electron and phonon operators.

2.2 Phonon-electron collisional integral and the electron density-density correlation function

We obtain the kinetic equation for slow variables $N_f(t)$, smoothing Heisenberg equations of motion over microscopically-large time interval $\Delta t$. Such a technique was suggested by N.N. Bogolyubov and N.M. Krylov (see the book [5], pp. 5-76) and it has been used in [6] for the derivation of the linearized kinetic equation for the occupation number fluctuations with Langevin sources of fluctuations. With accuracy to the second-order terms in $e - ph$ interaction we have

$$\frac{\Delta N_f(t)}{\Delta t} = -\frac{1}{\hbar^2 \Delta t} \left( S^+(t) \int t' dt' \int dt'' \left[ V_{eph}^0(t''), \left[ V_{eph}^0(t'), (b_f^\dagger b_f)\right] \right] S(t) \right) \tag{1}$$

The superscript 0 denotes operators in the interaction representation with respect to the electron-phonon interaction $V_{eph}$.

Let us remind interrelations between $S$-matrix and the time evolution of any operator $\hat{O}$ in the Heisenberg and interaction representations:

$$U(t, t_0) = e^{-\frac{i}{\hbar} H(t-t_0)} = U^0(t, t_0)S(t, t_0), \hspace{1cm} U^0(t, t_0) = e^{-\frac{i}{\hbar}(H_e + H_{ph})(t-t_0)},$$

$$\hat{O}(t) = U^+(t, t_0)\hat{O}(t_0)U(t, t_0) = S^+(t, t_0)\hat{O}^0(t, t_0)S(t, t_0)$$

$$i\hbar \partial S/\partial t = V_{eph}^0(t)S, \hspace{1cm} S(t_0) = 1, \hspace{1cm} \hat{O}^0(t, t_0) = U^0+(t, t_0)\hat{O}(t_0)U^0(t, t_0)$$

Calculations of the internal commutators in Eq.(1) lead to the desired result involving the electron density-density correlation function:

$$\frac{dN_f}{dt} = \frac{|c_f|^2}{v_0 \hbar^2} \left\{ \left( \rho_f^+ \rho_f \right)^0_{\omega_f} (N_f + 1) - \left( \rho_f \rho_f^+ \right)^0_{-\omega_f} N_f \right\} \tag{3}$$

where the commutativity of free electron and phonon operators in Eq.(1) and the condition $\omega_f \Delta t \gg 1$ have been used.
The function

$$\langle \rho_f \rho_f^+ \rangle_\omega = \int_{-\infty}^{\infty} d\tau \langle \rho_f(t+\tau)\rho_f^+(t) \rangle \exp(i\omega\tau)$$

(4)

is the spectral density of the correlation function of the electron density fluctuations. It is interested to remark that the right hand side of Eq.(3) may be rather interpreted as the emission and the absorption of phonons by the electron fluid with the structure factor $\langle \rho_f \rho_f^+ \rangle_\omega$ than as individual electron-phonon collisions.

2.3 Spectral density of the electron fluctuations

The sought spectral density must be calculated without $V_{ep}$ taken into account. From equations of motion for the electron operators we obtain in the random phase approximation:

$$\langle \rho_f \rho_f^+ \rangle_\omega^0 = \frac{\langle \rho_f \rho_f^+ \rangle_{\omega_0}}{|\epsilon(\omega, f)|^2},$$

(5)

where $\epsilon(\omega, f)$ is the dielectric function of the electron gas and

$$\langle \rho_f \rho_f^+ \rangle_\omega^0 = \sum_{k,k_1} \langle \rho_f(k)\rho_f^+(k_1) \rangle_t$$

is the spectral density of the density-density correlation function for non-interacting electrons. The one-time correlation function is

$$\langle \rho_f(k)\rho_f^+(k_1) \rangle_t \equiv \left\langle \frac{1}{i \left(\frac{\omega_{k+\frac{\Omega}{2}} - \omega_k - \frac{\omega}{2}}{\eta} + \eta \right) + \frac{1}{i \left(\frac{\omega_{k-\frac{\Omega}{2}} - \omega_k + \frac{\omega}{2}}{\eta} + \eta \right)}} \right\rangle.$$

(6)

The one-time correlation function of the non-interacting electrons is independent of time (in steady state) under condition that the electron energies $\hbar\Omega_k = \hbar^2 k^2/2m$ obey the equality

$$\Omega_k - \frac{\Omega}{4} = \Omega_k + \frac{\Omega}{4} - \Omega_{k-\frac{\Omega}{4}} \equiv \frac{\hbar}{m} f (k_1 - k) = 0.$$

(7)

As long as $f \neq 0$ it is necessary to be $k_1 = k$. Therefore, this condition provides the one-time correlator to be the integral of motion during the time interval $\Delta t$ like the distribution function $F_k$.

2.4 Kinetic equation with account for the contribution of nonequilibrium pair correlations

The substitution of the explicit expression for correlators to the kinetic equation (8) gives rise to

$$\frac{dN_f}{dt} = \frac{2|C_f|^2}{\omega_0 \hbar^2 |\epsilon(\omega, f)|^2} \sum_k \left\langle \frac{\alpha_k^+ a_k^+ a_{k-\frac{\Omega}{4}} a_{k+\frac{\Omega}{4}} a_{k-\frac{\Omega}{4}} a_{k+\frac{\Omega}{4}}}{\Omega_k + \frac{\Omega}{4} - \Omega_k - \frac{\Omega}{4} - \omega_f} \right\rangle (N_f + 1) -$$

$$- \frac{2|C_f|^2}{\omega_0 \hbar^2 |\epsilon(\omega, f)|^2} \sum_k \left\langle \frac{\alpha_k^+ a_k^+ a_{k-\frac{\Omega}{4}} a_{k+\frac{\Omega}{4}} a_{k-\frac{\Omega}{4}} a_{k+\frac{\Omega}{4}}}{\Omega_k + \frac{\Omega}{4} - \Omega_k - \frac{\Omega}{4} - \omega_f} \right\rangle N_f.$$

Taking into account the commutation rules $a_k^+ a_{k}, a_k a_{k}^+ = 1$ it is easy to obtain

$$\langle a_k^+ a_{k_1} a_{k_1} a_k \rangle_t = F_k(t) - \langle a_k^+ a_{k_1} a_{k_1} a_k \rangle_t.$$
According to the results from Gantsevich et al. [8],

$$\langle a_{k_1}^+ a_{k_2} a_{k_3} a_{k_4} \rangle = F_k F_{k_1} + \phi(k, k_1)$$

where $\phi(k, k_1)$ is a nonequilibrium correction to the pair correlator.

Finally, the kinetic equation for phonons with corrections for the non-equilibrium part of the electron pair correlator takes the form:

$$\frac{dN_f}{dt} = \frac{2(C_f)^2(N_f+1)}{\tilde{v} \hbar^2 \epsilon(\omega_f, f)^2} \sum_{k} \left[ F_k \left(1 - F_{k-f} \right) - \phi(k, k - f) \right] \delta(\omega_k - \omega_{k-f} - \omega) - \frac{2(C_f)^2 N_f}{\tilde{v} \hbar^2 \epsilon(\omega_f, f)^2} \sum_{k} \left[ F_k \left(1 - F_{k+f} \right) - \phi(k, k + f) \right] \delta(\omega_k - \omega_{k+f} + \omega).$$

It should be stressed that for ordinary classical equilibrium electron gas there are no pair correlations between different states in momentum space, i.e., $\phi(k, k_1) \equiv 0$. This fact can be easily seen from the form of the Gibbs distribution with the usual kinetic energy part of the Hamilton’s function $K = \sum_i p_i^2/2m$ since there are no terms proportional to $p_i p_j$ ($i, j$ stand for the particle enumeration).

### 3 Interrelation between non-linear stochastic kinetic equation and correlation corrections

Following [3] and [7] and using partial averaging over fast and short-wave fluctuations under conditions that the slow variables like $N_f(t)$ and $F_k(t)$ are constant instead of the full averaging over all states of the system under consideration, we find the non-linear stochastic kinetic equation with Langevin sources of fluctuations in the form

$$\frac{d\tilde{N}_f}{dt} = J_f(t) + \frac{2(C_f)^2}{\tilde{v} \hbar^2 \epsilon(\omega_f, f)^2} \times \left\{ \left( \tilde{N}_f + 1 \right) \sum_k \left[ \tilde{F}_k \left(1 - \tilde{F}_{k-f} \right) \right] \delta(\omega_k - \omega_{k-f} - \omega) - \tilde{N}_f \sum_k \left[ \tilde{F}_k \left(1 - \tilde{F}_{k+f} \right) \right] \delta(\omega_k - \omega_{k+f} + \omega) \right\}$$

The similar stochastic kinetic equation can be derived for the fluctuating electron occupation numbers $\tilde{F}_k$ with accounting for $V_{ee}$ and $V_{eph}$ interactions [3]. Both equations are nonlinear in $\tilde{F}_k$ and bilinear in $\tilde{N}_f \tilde{F}_k$. Here tilde denotes that the slow fluctuations are still kept. Langevin source of fluctuations can be expressed as the difference between the exact time derivative of the phonon occupation number and its approximation

$$J_f(t) = \frac{1}{\hbar \Delta t} \sum_{k} S^+(t) \int_{t}^{t+\Delta t} dt' V_{eph}(t') N_f^0(t') S(t) - \frac{1}{\hbar \Delta t} \sum_{k} S^+(t) \int_{t}^{t+\Delta t} dt' \overleftarrow{V_{eph}(t')} N_f^0(t') S(t)$$

It is seen that the partial average of Langevin sources themselves is equal to zero $\tilde{J}_f(t) = 0$. The partial correlation function $\frac{1}{2}[J_f(t_1), J_f(t_2)]_+$ is expressed by means of the transition rates $\gamma_{ij}$ in the collision integral similarly to the case of the linear stochastic kinetic equation for fluctuations of the occupation numbers. [3]

Taking full average from stochastic kinetic equation and using the evident properties (see Eq. (10))

$$\langle \tilde{F}_k \rangle = F_k; \quad \langle \tilde{F}_k \tilde{F}_{k_1} \rangle = F_k F_{k_1} + \phi(k, k_1), k \neq k_1,$$

and also $\langle J_f(t) \rangle = 0$ we can easily obtain the same kinetic equation for phonon occupation numbers with correlation corrections that was derived above. Moreover, from this point of view we can immediately write the respective terms in the Boltzmann kinetic equation for neutral particles and bring up the question about a possible contribution of nonequilibrium particle–particle correlations to the equations describing phenomena like flow turbulence. The similar statement concerns to electron-phonon correlations described by expressions like $\langle \delta \tilde{F}_k \delta \tilde{N}_f \rangle_0$, in the case when a phonon nonequilibrium takes place.

In conclusion of these general considerations it needs to underline that the contribution of non-equilibrium correlations to kinetics should be expected in that cases when the excitations considered are scattered by nonequilibrium many-particle system.
4 Electron energy loss rate via phonons

The energy loss rate of electrons due to phonons per unit volume can be obtained using the relation:

$$ P = \frac{1}{v_0} \sum_j \hbar \omega_j \frac{dN_j}{dt}. \quad (14) $$

It is seen from the kinetic equation for phonons Eq.(11) that the loss rate can be broken up into two parts:

$$ P = P_1 + P_2, \quad (15) $$

where on the right hand side the first term is related to the one-particle electron distribution function and it is described by the known expressions. The second one is due to the correlation contributions determined by the function $\phi$. We assume during the calculations of $P$ that the phonons are in thermal equilibrium and the electron-electron collisions are sufficiently frequent in order to provide the use of the electron temperature approximation for hot electrons. As a result of these assumptions the following formulae may be derived:

$$ P_1(T) = \sum_f \frac{2 |C_f|^2}{(v_0 \hbar)^2} \kappa \omega_j \left[ N_0^j(T) - N_f \right] $$$$ \times \sum_k \left( F_k^0 - F_{k+f}^0 \right) \delta (\Omega_{k+f} - \Omega_k - \omega_f) \quad (16) $$

and

$$ P_2(T, T_0) = -\sum_f \frac{2 |C_f|^2}{(v_0 \hbar)^2} \kappa \omega_j \sum_k \phi(k + f, k) \delta (\Omega_{k+f} - \Omega_k - \omega_f). \quad (17) $$

Here $N_0^j(T) = \left[ \exp(\hbar \omega_j/T) - 1 \right]^{-1}$ is the equilibrium Bose-Einstein distribution with hot-electron temperature $T$ and $N_f$ is the true nonequilibrium phonon distribution function. Further, we will consider the equilibrium phonons, i.e., $N_f = N_0^j(T_0)$. In this case the expression (18) for $P_1$ coincides with well-known Kogan’s formula except for dielectric function in the denominator describing the dynamic screening of the electron-phonon interaction.

The function $\phi(k, k_1)$ was found in [3] under accepted assumptions and can be presented in the form:

$$ \phi(k, k_1) = Q(T, T_0) \frac{\partial F_k^0}{\partial T} \frac{\partial F_{k_1}^0}{\partial T}, \quad (18) $$

where $F_k^0(T)$ is the Maxwell distribution of non-degenerate hot electrons and

$$ Q(T, T_0) = \frac{T^2}{mc_0} \left[ \frac{P(T, T_0)}{(T - T_0)} \frac{dP}{dT} - 1 \right] \quad (19) $$

The expression for $\phi$ in [3] was obtained without accounting for the contribution of $\phi$ itself in $P$. Therefore, the quantity $P_1$ was only implied as the total energy loss rate $P$. Now evidently we have the equation for determination of $P_2(T, T_0)$ since this quantity is defined by the right hand side of the equality (17) where $\phi(k + f, k)$ depends itself on $P_2(T, T_0)$ accordingly to Eqs. (13), (18). The situation is very resembling the self-consistent field approach in the plasma theory. Now we have to solve simultaneously the set of kinetic equations for the one-particle distribution function and the pair correlator.

It needs to stress that the substitution of the total loss rate $P$ instead of $P_1$ in the expression (18) for $\phi(k, k_1)$ is justified because all symmetry properties of the electron-electron and electron-phonon integrals of collisions used in [3] during the derivation are not destroyed by introducing $\phi(k, k_1)$ in collision integrals for electrons and phonons.

It is of importance to note in conclusion that the expression (18) for $\phi(k, k_1)$ found in the limit of high frequency of the electron-electron collisions does contain coupling constants of neither electron-electron nor electron-phonon interactions. Therefore, the corresponding corrections to the integral of
phonon-electron collisions do not contain additional powers of the small parameters of the kinetic theory. At low heating it is proportional to $T - T_0$ like $P_1(T, T_0)$, as it can be obtained from the expression (13) for $Q(T, T_0)$. However, the solution (18) for $\phi(k, k_1)$ has been obtained in [3] in the case of the non-degenerate electron gas. Hence $F_0^0(T) \ll 1$ and the product of two distribution functions in Eq. (18) makes $P_2 \ll P_1$ in the case of scattering electrons by acoustic phonons since $\hbar \omega_f \ll T, T_0$ and $N_f \gg 1$ for phonons involved. The situation can change in the case of low-temperature scattering of electrons by optical phonons under condition $\hbar \omega_0 \gg T, T_0$. The $\phi(k, k_1)$ corrections should be more pronounced for the hot-electron phenomena in the degenerate electron gas of semiconductors or metals [9].

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