

# MANY-BODY CORRELATIONS IN THE KINETIC THEORY OF SOLIDS

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## Abstract

Multiquantum integral kinetic equations for the micropolarization and longitudinal dielectric function are derived with account of pair correlations. The Heisenberg equation of motion and fluctuation-dissipation theorem has been used to calculate a micropolarization. It has been shown that kinetic equation for a micropolarization may be represented as power series in fluctuations of the instantaneous plasma field near the mean field. Direct Coulomb pair correlations have been evaluated explicitly by means of fluctuation-dissipation theorem. The kinetic motion equation with regard for pair correlations has been used to derive a multi-quantum integral equation for the complex order parameter  $\Delta_k$  in a superconductor.

## 1. General considerations

Correlation effects influencing the kinetic properties of semiconductors [1-3] and metals [4-6] are intensively studied at the present time. Processes of photon emission and absorption with simultaneous creation of a few low-frequency optical plasmons as typical electronic eigenmodes induced by band charge carrier correlations were investigated experimentally and theoretically [1-3].

Physical properties of transition metals, correlation energy, lifetimes of electrons, non-Fermi-liquid behavior, and Cooper pair formation have been investigated in [4-6].

In spite of the fact that all these investigations [1-6] quite naturally deal, from the outset, with different correlation effects, the theoretical approaches derived have some common features. First of all, it is standard non-relativistic many-body Hamiltonian given by the expression

$$\hat{H} = \sum_{\alpha, n, m} \hat{h}_{nm}^{\alpha} \hat{a}_{n\alpha}^+ \hat{a}_{m\alpha} + \frac{1}{2} \sum_{q, \alpha} V_q^{\alpha\alpha} \sum_{m, n, l, p} e^{iqr_{\alpha}} e^{-iqr_{\alpha}} \hat{a}_{l\alpha}^+ \hat{a}_{p\alpha}^+ \hat{a}_{m\alpha} \hat{a}_{n\alpha}, \quad V_q^{\alpha\beta} = \frac{4\pi e_{\alpha} e_{\beta}}{V q^2}, \quad (1)$$

where  $\hat{a}_{i\alpha}^+$ ,  $\hat{a}_{f\alpha}$  are the creation and destruction operators,  $n$  is the collective index incorporated all the degrees of freedom. The  $l, p, m, n$  subscripts represent the quantum number sets, describing the system of one-particle eigen-states. The  $\alpha = e, n$  index distinguishes electrons and nuclei. The last term in Eq. (1) describes the repulsive Coulomb interaction among electrons  $\alpha = e$  and among nuclei  $\alpha = n$ . The matrix  $\hat{h}_{n,m}^{\alpha}$  parameterises operator

$$\hat{h}^{\alpha} = \frac{1}{2m_{\alpha}} \left( \hat{p}_{\alpha} - \frac{e_{\alpha}}{c} \hat{A}_{\alpha}(r_{\alpha}, t) \right)^2 + e_{\alpha} \varphi(r_{\alpha}, t) + \sum_{\beta} \sum_q V_q^{\alpha, \beta} e^{iqr_{\alpha}} \hat{\rho}_q^{\beta} \quad (2)$$

$$\hat{\rho}_q^{\beta} = \sum_{n, m} e^{-i q r_{\beta}} \hat{P}_{nm}^{\beta}, \quad \hat{P}_{nm}^{\alpha} = \hat{a}_{n\alpha}^+ \hat{a}_{m\alpha}.$$

Here  $\hat{p}_{\alpha}$  is the momentum operator,  $r_{\alpha}$  is the radius vector of particle with charge  $e_{\alpha}$ . The symbol  $\alpha$  at sum after  $\beta$  means that the term with  $\beta = \alpha$  has to be excluded. Other designations are standard.

The lagging response of many-body system with Hamiltonian (1) to the external longitudinal and transversal electromagnetic field perturbations, the charge density, and the macroscopic polarization are determined by the operator of micropolarization  $\hat{P}_{if}^\alpha$  time dependence [1-4]. Thus, the problem reduces to determination of expectation value

$$P_{if}^\alpha(t) = \langle \hat{P}_{if}^\alpha(t) \rangle = Sp\{\hat{\rho}_0 \hat{P}_{if}^\alpha(t)\}, \quad \hat{P}_{if}^\alpha(t) = \hat{U}^\dagger(t) \hat{P}_{if}^\alpha \hat{U}(t), \quad \hat{U}(t) = T \exp\left(-\frac{i}{\hbar} \int_0^t H(s) ds\right), \quad (3)$$

where  $\hat{U}(t)$  is the evolution operator. Angular brackets denote average over the density of states. The calculation of macroscopical response to the external perturbations includes the operation of quantum statistic averaging with using of the equilibrium density matrix  $\hat{\rho}_0$ . In terms of creation and destruction operators, according to formula (3), the time dependence of the micropolarization operator  $\hat{P}_{if}^\alpha$ , has to be *ab initio* derived from the Heisenberg equation of motion. Working out commutators, one obtains

$$\begin{aligned} \frac{\hbar}{i} \frac{\partial P_{if}^\alpha}{\partial t} &= \frac{\hbar}{i} \dot{P}_{if}^\alpha = \sum_n (h_{ni} P_{nf}^\alpha - P_{in}^\alpha h_{fn}) + \\ &+ \sum_q V_q^{\alpha\alpha} \sum_{m,n,l} (e_{ln}^{iqr_\alpha} e_{fm}^{-iqr_\alpha} \langle \hat{a}_{l\alpha}^+ \hat{a}_{i\alpha}^+ \hat{a}_{n\alpha} \hat{a}_{m\alpha} \rangle - e_{li}^{iqr_\alpha} e_{nm}^{-iqr_\alpha} \langle \hat{a}_{n\alpha}^+ \hat{a}_{l\alpha}^+ \hat{a}_{m\alpha} \hat{a}_{f\alpha} \rangle). \end{aligned} \quad (4)$$

The second part of Eq. (4) shows contributions due to correlated four-operator terms. The dynamical Hartree-Fock (HF) like approximation, according to which

$$\langle \hat{a}_l^+ \hat{a}_i^+ \hat{a}_n \hat{a}_m \rangle = \langle \hat{a}_l^+ \hat{a}_m \rangle \langle \hat{a}_i^+ \hat{a}_n \rangle - \langle \hat{a}_l^+ \hat{a}_n \rangle \langle \hat{a}_i^+ \hat{a}_m \rangle = P_{lm} P_{in} - P_{ln} P_{im} \quad (5)$$

provides what is believed to be a simplest approximation for the solution of the many-body problem with Hamiltonian (1). It is necessary to note that in all above mentioned different theories [1-6] the HF approximation is used as the starting point. HF factorization (5) yields quite a satisfactory result for micropolarization  $P_{if}(t)$

$$\begin{aligned} \frac{\hbar}{i} \dot{P}_{if}^{HF}(t) &= \sum_{n,m} \hat{h}_{nm}(t) \langle [\hat{P}_{nm}(t), \hat{P}_{if}(t)] \rangle = \hbar \omega_{if}^{HF}(t) P_{if}(t) + \sum_n \tilde{h}_{ni}^{if}(t) P_{nf}(t) - \sum_n \tilde{h}_{fn}^{if}(t) P_{in}(t), \\ \hbar \omega_{if}^{HF} &= \tilde{h}_{ii} - \tilde{h}_{ff} + \sum_n M_{in}^{ff} P_{in} - \sum_n M_{nf}^{ii} P_{nf}. \end{aligned} \quad (6)$$

This equation is mathematically identical to the Schrödinger equation with mean-field Hamiltonian

$$\begin{aligned} \hat{H}' &= \sum_{n,m} \tilde{h}_{nm} \hat{a}_n^+ \hat{a}_m, \quad \tilde{h}_{nm} = h_{nm} + \sum_{l,p} M_{nm}^{lp} P_{lp}, \\ M_{nm}^{lp} &= \sum_q V_q (e_{nm}^{iqr} e_{lp}^{-iqr} - e_{np}^{iqr} e_{lm}^{-iqr}), \quad \tilde{h}_{nm}^{if} = \tilde{h}_{nm} - M_{nm}^{if} P_{if}. \end{aligned} \quad (7)$$

Equation (6) has to be considered a definition of generalized transition frequency  $\hbar \omega_{if}^{HF}(t)$ .

In addition to the Hartree-Fock difference of proper energies  $\tilde{h}_{ii} - \tilde{h}_{ff}$  of initial  $i$  and final  $f$  states (Koopmans approximation), Eq. (6) takes into account the unscreened exciton Coulomb interaction between the quasi-particle in  $f$  state and quasi-hole in  $i$  state.

As it is known, the random phase approximation implies the replacement of instantaneous field created by plasma with mean field [4]. Micropolarization (6) calculated in the mean-field HF approximation can be used next for evaluation of the response function, the light absorption coefficient, the spontaneous recombination rate, and so on. It leads to the important consequences for kinetic properties of solid states. Equation (6) at the level of dynamical HF approximation allows including the generalized random phase approximation into the consideration.

There is no difficulty in deriving Eq. (6) with using of HF decoupling scheme (5), but it is much more difficult to establish a validity of such a procedure. This work is concerned with *ab initio* derivation of multiquantum kinetic equation with a special emphasision on the many-body correlations. In particular, we calculate the dielectric function  $\epsilon(q, \omega)$  with account of fluctuations near the mean field in Eq. (4).

## 2. Multiquantum kinetic equations

To study the correlation contributions to kinetic equation (4), one has to go beyond mean-field HF approximation (5). We start by considering the deviations of the exact terms from their corresponding HF factorized parts

$$\dot{P}_{if}(t) = \dot{P}_{if}^{HF}(t) + \dot{P}_{if}^C(t). \quad (8)$$

The quantity  $\dot{P}_{if}^C(t)$  is defined as a correlation part of the kinetic equation for micropolarization. There are some interesting properties of the correlation part. First of all, Hartree-Fock factorization scheme (6) is explicit in the quasi-particle picture. Thus, first remarkable feature of Eq. (8) is that in the quasi-particle picture ( $\hat{H} \rightarrow \hat{H}_q$ ) the correlation part is identical to zero

$$\dot{P}_{if}^{qC}(t) = 0, \quad \hat{P}_{if}^q(t) = e^{\frac{i}{\hbar}\hat{H}_q t} \hat{P}_{if} e^{-\frac{i}{\hbar}\hat{H}_q t}, \quad \hat{H}_q = \sum_{n\alpha} E_{n\alpha} \hat{P}_{n\alpha}^\alpha. \quad (9)$$

Another feature appears if one calculates the correlation part  $\dot{P}_{if}^C(t)$  (8) by means of the perturbation theory, in which the exact expression for operator of evolution

$$U(t) = \exp(-\frac{i}{\hbar}\hat{H}_q t) - \frac{i}{\hbar} U(t) \int_0^t \exp(\frac{i}{\hbar}\hat{V}(s_1) ds_1) \hat{V}(s) ds, \quad \hat{V} = \hat{H} - \hat{H}_q \quad (10)$$

may be expanded as power series in  $\hat{V} = \hat{H} - \hat{H}_q$ . Keeping the first order terms to develop the equation of motion for microscopic polarization with account of correlation part  $\dot{P}_{if}^C(t)$ , we use the approximation

$$\hat{P}(t) = \hat{P}_q(t) + \frac{i}{\hbar} \int_0^t \langle [\hat{V}(t_1), \hat{P}(t)] \rangle dt_1, \quad \hat{V}(t) = U^+(t) \hat{V} U(t). \quad (11)$$

The correlation part  $\dot{P}_{if}^C(t)$  in the first order in  $\hat{V}$  is

$$\begin{aligned} \dot{P}_{if}^C(t) = \frac{i}{\hbar} \int_0^t dt_1 \left\{ \langle [\hat{V}(t_1), \hat{P}_{if}(t)] \rangle - \right. \\ \left. - \frac{i}{\hbar} \sum_{n,m} \left( \langle [\hat{V}(t_1), \hat{h}_{nm}(t)] \rangle \langle [\hat{P}_{nm}(t), \hat{P}_{if}(t)] \rangle + \tilde{h}_{nm}(t) \langle [\hat{V}(t_1), [\hat{P}_{nm}(t), \hat{P}_{if}(t)]] \rangle \right) \right\}. \end{aligned} \quad (12)$$

Equation (12) for the correlation part  $\dot{P}_{if}^C(t)$  (8) can be further simplified by carrying out of partial decoupling. In terms of fluctuations near mean field, this approximate expression for correlation part may be written as follows

$$\dot{P}_{if}^C(t_1) = \frac{1}{\hbar^2} \int_0^{t_1} dt_2 \{ I(1,2) - I(2,1) \}, \quad (13)$$

$$I(1,2) = \sum_{n_1 m_1} \sum_{n_2 m_2} \langle \Delta \hat{h}_{n_1 m_1}(t_1) \Delta \hat{h}_{n_2 m_2}(t_2) \rangle \langle \Delta [\hat{P}_{n_1 m_1}(t_1), \hat{P}_{if}(t_1)] \Delta \hat{P}_{n_2 m_2}(t_2) \rangle, \quad \Delta \hat{P} = \hat{P} - \langle \hat{P} \rangle,$$

where fluctuations near mean field are determined by

$$\begin{aligned}\Delta\hat{h}_{nm}(t) &= \sum_q \sum_\beta V_q^{\alpha\beta} e_{nm}^{iqr_\alpha} (\hat{\rho}_q^\beta(t) - \langle \hat{\rho}_q^\beta(t) \rangle), \\ \Delta\hat{h}_{nm}(t) &= \Delta\hat{h}_{nm}(t) - \sum_q V_q^{\alpha\beta} e_{np}^{iqr} e_{lm}^{-iqr} (\hat{P}_{lp}(t) - \langle \hat{P}_{lp}(t) \rangle).\end{aligned}\quad (14)$$

It should be noted here that the deviation of the first order correction (12) and approximate equation (13) in the quasi-particle picture vanishes. Thus, further remarkable feature of the correlation part  $\hat{P}_{if}^C(t)$  is that it may be represented as power series in fluctuations  $\Delta\hat{h}_{nm}(t)$ .

Direct Coulomb interaction gives the contribution into the density-density correlator, which is proportional to  $V_q^2$  and diverges as  $q^{-4}$  at  $q \rightarrow 0$ , if we restrict the consideration with the second order correction of the perturbation theory. To avoid this difficulty, some authors replace the unscreened Coulomb potential with its screened value. But screening should result from the proper treatment of correlations. The expectation value of charge density operator multiplication  $\langle \hat{\rho}_q(t) \hat{\rho}_{-q}(s) \rangle$ , taken in different time points in Eq. (14), specifies memory effects. It can be precisely calculated within the fluctuation-dissipation theorem [7] and can be expressed in terms of structure factor or inverse dielectric function

$$\begin{aligned}\langle \Delta\hat{h}_1(t_1) \Delta\hat{h}_2(t_2) \rangle &= \frac{\hbar}{\pi} \sum_q V_q \sum_{1,2} e_1^{iqr} e_2^{-iqr} \int_{-\infty}^{\infty} \exp(-i\omega(t-s)) [n(\omega) + 1] \text{Im} \left\{ \frac{1}{\varepsilon^*(\kappa, \omega)} \right\} d\omega, \\ n(\omega) &= \left[ \exp \left( \frac{\hbar\omega}{k_0 T} \right) - 1 \right]^{-1},\end{aligned}\quad (15)$$

where the asterisk signifies the complex conjugate. Equations (8), (13)-(15) determine the micropolarization in the pair correlation approximation. Making use of equation (15), we obtain generalized transition frequency  $\hbar\omega_{if}(t)$  in the form

$$\begin{aligned}\hbar\omega_{if}(t) &= \hbar\omega_{if}^{HF}(t) + \Delta E_i^f(t) - \Delta E_f^{i*}(t) \\ \Delta E_i^f(t) &= \frac{i}{\pi} \sum_q V_q \int_{-\infty}^{\infty} d\omega [n(\omega) + 1] \text{Im} \left\{ \frac{1}{\varepsilon^*(q, \omega)} \right\} \times \\ &\times \int_0^t \left( \sum_n |e_{in}^{iqr}|^2 G_{ni}(t, t_1) \varphi_n^\omega(t - t_1) - e_{ii}^{iqr} e_{ff}^{-iqr} \varphi_i^\omega(t - t_1) \right) dt_1 \\ \varphi_n^\omega(t - t_1) &= n_n \exp(-i\omega(t - t_1)) + (1 - n_n) \exp(i\omega(t - t_1)).\end{aligned}\quad (16)$$

The last excitonic term from Eq. (16), which is proportional to  $e_{ii}^{iqr} e_{ff}^{-iqr}$ , can be reduced by carrying out the time and frequency integration by using the integral

$$\frac{1}{\pi} \int_{-\infty}^{\infty} d\omega [n(\omega) + 1] \text{Im} \left\{ \frac{1}{\varepsilon^*(q, \omega)} \right\} \left\{ \frac{1}{\omega - \omega_1} + \frac{1}{\omega + \omega_1} \right\} = 1 - \frac{1}{\varepsilon(q, \omega_1)}.\quad (17)$$

Equation (17) at  $\omega_1 = 0$  shows that direct excitonic Coulomb interaction of the electron and the hole is screened statically. On the other hand, the exchange interaction is exposed to dynamical screening. It can be easily shown by using the quasi-particle approximation  $G_{ni}(t, t_1) = \exp(i\omega_{ni}^q(t - t_1))$ ,  $n = f$  and Eq. (17).

The dynamical screening of correlation contribution in energy of quasi-particles (time independent contribution in Eq. (16)) is taken into account. The results of work [8] obtained by the Green function method can be evaluated from Eq. (16) if one takes random phase ap-

proximation for dielectric function. More than that, the Coulomb interaction leads to the multi-quantum collision processes [1].

Using the results obtained, we can calculate the longitudinal dielectric function with regard for pair correlations (15)

$$\varepsilon(q, \omega) = 1 - \frac{i}{\hbar} \sum_{\alpha} V_q^{\alpha\alpha} \sum_{i,f} G_{if}(\omega) \{ |e_{if}^{iqr}|^2 (n_f - n_i) - \frac{i}{\hbar} \sum_{q_1} V_{q_1} \sum_{i,f,l,p} e_{il}^{iqr} e_{lp}^{iq_1r} e_{pf}^{-iqr} e_{fl}^{-iq_1r} (n_l - n_p) G_{pl}(\omega) \left[ \frac{n_f}{\varepsilon(q_1, \omega + \omega_{pf})} - \frac{n_i}{\varepsilon(q_1, \omega + \omega_{il})} \right] \}. \quad (18)$$

Here, at the level of the quasi-particle approximation

$$G_{if}(\omega) = \int_0^{\infty} \exp[i(\omega + i\delta)t] G_{if}^q(t) dt = \frac{i}{\omega + \omega_{if}^q + i\delta}, \quad G_{if}^q(t) = \exp(i\omega_{if}^q t), \quad \hbar\omega_{if}^q = E_i - E_f, \quad (19)$$

where  $\delta$  is the infinitesimal positive quantity. We have included here the Coulomb-exchange contribution, Fock-field to the polarization field, and its fluctuations near mean field. The results of work [9] can be obtained from Eqs. (18) and (19) if we calculate the transition frequency  $\omega_{k,k_1}$  at the level of the first order approximation of exchange-interacting electrons

$\hbar\omega_{k,k_1}^{HF} = e_k - e_{k_1} - \sum_q V_q (n_{k+q} - n_{k_1+q})$ ,  $e_k = \hbar^2 k^2 / 2m$  and set  $\varepsilon(q_1, \omega + \omega_1) = 1$ . Mean-field approximation yields only expression (18) with  $\varepsilon(q_1, \omega + \omega_1) = 1$ .

Equation (16) for generalized transition frequency is an integral equation, because there is the integral dependence between the transition frequency  $\hbar\omega_{if}(t)$  (16) and dielectric function  $\varepsilon(q, \omega)$  (18), which has to be calculated selfconsistently by iteration method.

At this stage, we already have obtained a closed set of integral equations (13)-(18). The density-density correlator from Eq. (15) depends on the longitudinal dielectric function Eq. (18). On the other hand, dielectric function  $\varepsilon(q, \omega)$  depends on the micro-polarization, in its turn. Hence, equations (13)-(19) constitute a self-consistent set of multi-quantum integral kinetic equations.

If one wants to calculate superconductive properties of solids, one has to take into account anomalous average quantities  $\langle a_n a_m \rangle$  and  $\langle a_n^+ a_m^+ \rangle$ . This leads to the mean-field Hamiltonian in the form

$$\hat{H}' = \sum_{n,m} \tilde{h}_{nm} \hat{a}_n^+ \hat{a}_m + \frac{1}{2} \sum_{n,m} \{ \eta_{nm} \hat{Q}_{nm} + (\eta_{nm} \hat{Q}_{nm})^+ \}, \quad (20)$$

$$\eta_{nm} = \sum_q V_q e_{\alpha}^2 e_{lm}^{iqr} e_{pn}^{-iqr} \langle \hat{Q}_{pl}^+ \rangle, \quad \hat{Q}_{nm} = \hat{a}_n \hat{a}_m.$$

Function  $I(1,2)$  in equation (13) has to be changed by  $\tilde{I}(1,2) = I(1,2) + \delta I(1,2)$ , where

$$\delta I(1,2) = 1/2 \sum_{n_1 m_1} \sum_{n_2 m_2} \{ \langle \Delta \hat{h}_{n_1 m_1}(t_1) \Delta \hat{Q}_{m_2 n_2}(t_2) \rangle + \langle \Delta [\hat{P}_{n_1 m_1}(t_1), \hat{P}_{if}(t_1)] \Delta \hat{\eta}_{m_2 n_2}(t_2) \rangle + \langle \Delta \hat{h}_{n_1 m_1}(t_1) \Delta \hat{\eta}_{m_2 n_2}(t_2) \rangle + \langle \Delta [\hat{P}_{n_1 m_1}(t_1), \hat{P}_{if}(t_1)] \Delta \hat{Q}_{m_2 n_2}(t_2) \rangle \}. \quad (21)$$

It is now straightforward to use equations (20, 21) and to develop the motion equation for the operators  $\hat{a}_k^+$  and  $\hat{a}_{-k}$  for Cooper pairs in the form of the Bogoliubov equations with the order parameter

$$\Delta_k = - \sum_q V_q \frac{\Delta_{k+q}}{2E_{k+q}} (1 - 2n_{k+q}) \sum_{n=-\infty}^{\infty} \frac{I_n(z) \exp[-N_p - ny]}{\varepsilon^*(q, n\hbar\omega_p + E_k + E_{k+q})}, \quad (22)$$

where

$$z = N_p [1 - (1 - n_k^c - n_{k+q}^c)^2]^{1/2}, N_p = \frac{2}{\pi} (a_B k_F)^{-1} = 0.33 r_s, \exp(-y) = \left[ \frac{n_k^c + n_{k+q}^c}{2 - n_k^c - n_{k+q}^c} \right]^{1/2}, n_k^c = \int \frac{d\Omega}{4\pi} \int_0^{q_c} n_{k+q} \frac{dq}{q_c}. \quad (23)$$

The magnitude of the mean number of high-frequency plasmons  $N_p$ , generated in every collision process, is determined by the expression  $N_p = 2e^2 q_c / \pi \hbar \omega_p = 0.33 r_s$  [1]. For typical values of metal parameters, the numerical value of coupling constant is more than one ( $N_p \geq 1$ ). Hence, the influence of electron inelastic scattering associated with the excitation of several high-frequency plasmons is considerable.

Equation (23) for the complex order parameter  $\Delta_k$  involves, in a self-consistent fashion, the multiplasmon transitions. The kernel of the equation for  $\Delta_k$  is strongly modified. Low frequency contribution at  $n=0$  differs from the standard one [10, 11]. Kernel (23) includes the interaction between electrons due to the exchange of plasmons. The resonant processes in which  $n$  plasmons are emitted and as many absorbed simultaneously are defined by high-frequency properties of the dielectric function  $\varepsilon(q, \omega)$ . In a simple plasmon-pole approximation [1, 7], we get  $n=1$ . The scattering process includes two plasmons, one of which is emitted and the other one is absorbed.

Several results, which stem from this formulation, will be discussed later in the next publications.

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