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Multiquantum scattering processes and transmission electron energy loss spectra

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Abstract. Microscopic many-body theory for electronic properties of solid states is developed with an emphasis on the role of correlation memory effects. Heisenberg equation of motion, fluctuation-dissipation theorem and operators of commutation have been used to calculate multiplasmon transmission electron energy loss spectra. Multiquantum integral kinetic equation for the longitudinal complex dielectric function is derived. Strong interaction between high-energy probe beam electrons penetrating the solid state and plasma of valence electrons is taken into account. It is shown that average number of high-frequency plasmons generated in every collision process is more than one for typical values of metal parameters. It is obtained that excitation of a good few plasmons is simultaneous event. Calculated multiplasmon structure of electron energy loss spectra coincides with experimental.

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1 Introduction

For the correlation effects influencing the optical properties of semiconductors the long-range Coulomb interactions between the free carriers play an important role [1–9]. They give rise different collective and coherence effects which are intensively investigated at the present time [1–9]. Among them there are effects that have been interpreted in terms of strong coupling of e-h pairs with low-frequency optical plasmons [1–6]. Thus, the modification of carrier dynamics by absorption of one plasmon was shown in the work [10]. The consecutive absorption and emission of more then one plasmon was considered in the paper [11]. A spectroscopic study of free-to-free, free-to-bound and bound-to-bound e-h multi-plasmon recombination was fulfilled using photo and cathodo-luminescence techniques [1–6]. Processes of photon emission and absorption with simultaneous creation of a few low-frequency optical plasmons ($\hbar\omega_p \cong 10$ meV) as a typical electronic eigenmodes induced by band charge carriers correlations were investigated experimentally and theoretically in detail [1–6]. It was found that a new composite wide radiation band including about ten plasmon replicas appears at high excitation levels in the region of electron-hole plasma light emission. Multiplasmon optical transitions are important both for their emission band fine structure origin manifestations and for understanding of many key features of II–VI semiconductor luminescence spectra [1–6].

As a line-width enhancement factor multiplasmon optical transitions [1–6] satisfy energy conservation law which one can state by equation $\omega = \omega_g \pm n\omega_p$, where ω_p — frequency of long wavelength plasmon, n — number of plasmons generated, $\hbar\omega_g$ — band gap energy. Hence emission of photon and several plasmons is simultaneous process. In our previous works [1–6] we have introduced a coupling constant N_p (mean number of low-frequency optical plasmons, emitted along with one photon). Multiplasmon optical transitions are actual at $N_p > 1$ [1–6]. Analogous constant can be introduced for high frequency $\hbar\omega_p \cong 10$ eV plasmons $N_p = 1/a_B k_F = 0.52 r_s$ which are given in terms of Bohr radius $a_B = \hbar^2/(me^2)$ and Fermi momentum $k_F = (3\pi^2 N)^{1/3}$. In the most cases the value of parameter r_s is not small ($2 < r_s < 6$). Thus study of electron inelastic scattering associated with excitation of several high-frequency plasmons is actual as well.

The aim of this paper is to derive the multiplasmon theoretical approach, which can be applied to the corresponding problem in transmission electron energy loss spectroscopy. A large number of experimental and theoretical papers [12–16] are devoted to the problem of electron energy losses for various targets, in which multiple generation of high-frequency plasmons exhibiting the valence electron oscillations in metals and semiconductors are considered as consecutive process [12–16], characterized by electron mean free path concerning to plasmon generation. This process consists of separate consecutive collisions with generation of one plasmon in each collision ($\omega = \omega_p$). Thus for optical energy loss function $\mathrm{Im}\{1/\varepsilon^*\}$ with single

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plasmon pole is used Drude-Lindhard model [12–16]. In this work we investigate spectra of electron energy losses in solid state with taking into the consideration of simultaneous multiple generation of high-frequency plasmons. We give general integral multiquantum kinetic equation for a determination of a longitudinal dielectric function $\varepsilon(\kappa,\omega)$ derived on the basis of fluctuation-dissipation theorem with using of commutation operators.

We have to mention at first that in works [1–11] two-band model for semiconductors which includes the direct Coulomb interactions was considered. According to this model electrons and holes are in the dielectric media with background dielectric constant of material and well-known effective mass approximation is used. In this paper we examine the electron energy loss spectra without of using any model approximation.

2 General considerations

The starting point for our consideration is the Heisenberg equation of motion for microscopic polarization operators with taking into account of non-screened Coulomb interaction within the many-body electrons and nucleus system. The Hamiltonian from which we will derive the microscopic quantum kinetic equations is

$$\hat{H} = \sum_{\alpha,n,m} \hat{h}^{\alpha}_{nm}\hat{a}^{+}_{n\alpha}\hat{a}_{m\alpha} + \frac{1}{2}\sum_{\kappa,\alpha} V^{\alpha\alpha}_{\kappa} \sum_{m,n,l,p} e^{i\kappa r_\alpha}_{l,n} e^{-i\kappa r_\alpha}_{p,m} \hat{a}^{+}_{l\alpha}\hat{a}^{+}_{p\alpha}\hat{a}_{m\alpha}\hat{a}_{n\alpha},$$
$$V^{\alpha\beta}_{\kappa} = \frac{4\pi q_\alpha q_\beta}{V\kappa^2}. \quad (1)$$

Here $\hat{a}^{+}_{i\alpha}$, $\hat{a}_{f\alpha}$ are the creation and destruction operators, $V^{\alpha\beta}_{\kappa}$ is the Fourier transform of Coulomb potential. The i, f subscripts represent the quantum numbers sets, describing the many-body system eigen-functions. The $\alpha = e, n$ ($q_e = e$) index distinguishes electrons and nuclei. The last term in equation (1) describes the repulsive Coulomb interaction among electrons $\alpha = e$ and among nuclei $\alpha = n$. The interaction with electromagnetic field and Coulomb attraction between electrons and nuclei are included in operator

$$\hat{h}^{\alpha} = \frac{1}{2m_\alpha}\left(\hat{p}_\alpha - \frac{q_\alpha}{c}\hat{A}_\alpha(r_\alpha,t)\right)^2 + q_\alpha\varphi(r_\alpha,t) + \sum_{\beta}\sum_{\kappa} V^{\alpha,\beta}_{\kappa} e^{i\kappa r_\alpha}\hat{\rho}^{\beta}_{\kappa},\hat{\rho}^{\beta}_{\kappa} = \sum_{n,m} e^{-i\kappa r_\beta}_{nm}\hat{P}^{\beta}_{nm}, \hat{P}^{\alpha}_{nm} = \hat{a}^{+}_{n\alpha}\hat{a}_{m\alpha}, \beta \neq \alpha, \quad (2)$$

where $\hat{p}_\alpha$ is the momentum operator, r_α is the radius vector of particle with charge q_α. Other designations are standard. Here it is emphasized that field (φ, A), which appears in equation (2) is the microscopic local field.

The response under the external longitudinal and transversal electromagnetic fields perturbation of many-body system with Hamiltonian (1) is determined by the microscopic polarization operator time dependence [3,6]. In terms of creation and destruction operators according to the formula (2) the time dependence of the micropolarization operator $\hat{P}^{\alpha}_{if}$, has to be derived from the Heisenberg equation of motion. Working out commutators, Heisenberg equation of motion for operator $\hat{P}^{\alpha}_{if}(t)$ can be transformed to the next one

$$\frac{\partial\hat{P}^{\alpha}_{if}}{\partial t} = \frac{i}{\hbar}\left[\hat{H},\hat{P}^{\alpha}_{if}\right] = \frac{i}{\hbar}\sum_{n}\left(\hat{h}_{n\,i}P^{\alpha}_{n\,f} - P^{\alpha}_{i\,n}\hat{h}_{fn}\right) + \frac{i}{\hbar}\sum_{\kappa} V^{\alpha\alpha}_{\kappa}\sum_{m,n,l}\left(e^{i\kappa r_\alpha}_{l,n}e^{-i\kappa r_\alpha}_{f,m}\hat{a}^{+}_{l\alpha}\hat{a}^{+}_{i\alpha}\hat{a}_{n\alpha}\hat{a}_{m\alpha} - e^{i\kappa r_\alpha}_{l,i}e^{-i\kappa r_\alpha}_{n,m}\hat{a}^{+}_{n\alpha}\hat{a}^{+}_{l\alpha}\hat{a}_{m\alpha}\hat{a}_{f\alpha}\right). \quad (3)$$

The second part of equation (3) shows contributions due to correlated four-point terms. A simple approximation for the correlation contributions can be obtained within the density matrix theory or within a Green's function method [7–11]. The Heisenberg equation of motion (3) for operator $\hat{P}^{\alpha}_{if}$, with transforming the four operator terms, can be presented as (index α is temporarily omitted)

$$\frac{\partial\hat{P}_{if}}{\partial t} = \frac{i}{\hbar}(\hat{E}_{if}\hat{P}_{if} - \hat{P}_{if}\hat{E}^{+}_{fi}),$$
$$\hat{E}_{if} = \hat{\tilde{h}}^{f}_{ii} + \frac{1}{2}(1-\delta_{fi})\hat{\tilde{h}}_{fi}\hat{C}_{fi} + \sum_{n}^{i,f}\left(\hat{h}_{fn} + \frac{1}{2}\hat{\tilde{h}}^{C}_{fn}\right)\hat{C}_{fn}. \quad (4)$$

The symbols i, f at sum after n mean that the terms with $n = i, f$ have to be excluded. The first term in equation (4) determines the diagonal part of operator $\hat{E}_{if}$

$$\hat{\tilde{h}}^{f}_{ii} = \hat{h}_{ii} + \sum_{n}\sum_{m}^{f} M^{ii}_{nm}\hat{P}_{nm} = \hat{E}_i - \sum_{n} M^{ii}_{nf}\hat{P}_{nf},$$
$$M^{kl}_{nm} = \sum_{\kappa} V_\kappa\left(e^{i\kappa r}_{nm}e^{-i\kappa r}_{kl} - e^{i\kappa r}_{nl}e^{-i\kappa r}_{km}\right). \quad (5)$$

The matrix M^{kl}_{nm}, equation (5), is the sum of two terms describing the direct and exchange Coulomb interaction contributions respectively. Off-diagonal terms in equation of motion (3) are turned to diagonal form (4), using the commutation operators $\hat{C}_{nm}$. The commutation operator action upon the micropolarization operator $\hat{P}_{if}$, according to the definition, consist of

$$\hat{C}_{nm}\hat{P}_{if} = [\hat{P}_{nm},\hat{P}_{if}] = \hat{P}_{nf}\delta_{im} - \hat{P}_{im}\delta_{nf}. \quad (6)$$

Berezin has introduced in [17] the commutation operator of equation of motion (3) $\hat{C}\hat{P}_{if} = [\hat{H},\hat{P}_{if}]$. The explicit form of the expression for off-diagonal terms of operators $\hat{\tilde{h}}_{nm}$ in equation (4) is determined as

$$\hat{\tilde{h}}_{fn} = \hat{h}_{fn} + \hat{h}^{C}_{fn}, \quad \hat{h}^{C}_{fn} = \sum_{m}^{ifn}\sum_{l}^{if} M^{fn}_{lm}\hat{P}_{lm}. \quad (7)$$

The exchange energies in Hartree-Fock approximation can be easily obtained from the expectation value of an operator $\hat{\tilde{h}}^f_{ii}(t)$ (5). Correlation memory effects are described by terms in equation (4), which contain commutation operators. Exact solution of Heisenberg equation of motion (4) can be presented in the form

$$\hat{P}_{if}(t) = \exp\left\{\frac{i}{\hbar}\int_0^t \hat{E}_{if}(s)ds\right\}\hat{P}_{if}(0) \times \exp\left\{-\frac{i}{\hbar}\int_0^t \hat{E}^+_{fi}(s)ds\right\}. \quad (8)$$

The calculation of macroscopical response under external perturbations includes the operation of quantum statistic averaging with using of the equilibrium density matrix. Taking into account the diagonal part $\hat{\tilde{h}}^f_{ii}$ (5) of the operator $\hat{E}_{if}$, expanding T exponents (8) in series and carrying out the simple decoupling like

$$\left\langle \hat{\tilde{h}}^f_{ii}(t_1)\hat{\tilde{h}}^i_{ff}(t_2)...\hat{\tilde{h}}^i_{ff}(t_n)\right\rangle \cong \left\langle \hat{\tilde{h}}^f_{ii}(t_1)\right\rangle \left\langle \hat{\tilde{h}}^i_{ff}(t_2)\right\rangle ... \left\langle \hat{\tilde{h}}^i_{ff}(t_n)\right\rangle \quad (9)$$

we obtain

$$P_{if}(t) = \langle \hat{P}_{if}(t)\rangle = G_{if}(t-s)P_{if}(s),$$
$$G_{if}(t-s) = \exp\left\{i\int_s^t \omega_{if}(s)ds\right\}. \quad (10)$$

This result for micropolarization (10) corresponds to taking into consideration of first term in cumulant expansion method [18]. The first order cumulant is

$$\hbar\omega_{if} = \hbar\omega^{HF}_{if} = \left\langle \hat{\tilde{h}}^f_{ii} - \hat{\tilde{h}}^i_{ff}\right\rangle = \left\langle \hat{E}_i - \hat{E}_f\right\rangle - M^{ii}_{ff}(n_f - n_i), n_f = \left\langle \hat{P}_{ff}\right\rangle, \quad (11)$$

where angular brackets denote average over the density of states. Beside the Hartree-Fock difference of proper energies $\langle \hat{E}_i - \hat{E}_f\rangle$ of initial i and final f states (Cumans approximation), equation (11) takes into account the unscreened Coulomb interaction between the quasi-particle in f state and quasi-hole in i state. The second order cumulant describes the effect of exciton Coulomb interaction screening and multiquantum transitions.

To develop a quantum integral kinetic equation for solid states with account of correlation effects we use the exact solution of Heisenberg equation (3) for operator $\hat{P}_{if}(t)$in the form

$$\hat{P}_{if}(t) = G_{if}(t-s)\hat{P}_{if}(s) + \int_s^t G_{if}(t-t_1)\left\{\frac{\partial \hat{P}_{if}(t_1)}{\partial t_1} - i\omega_{if}(t_1)\hat{P}_{if}(t_1)\right\}dt_1. \quad (12)$$

To obtain $\omega_{if}(t)$ first of all one notes that Heisenberg equations (3, 4) contain diagonal and off-diagonal contributions. Upon inserting equation (4) into (12) and averaging with respect to the density of states we obtain the expression for average transition frequency $\omega_{if}(t)$ from the diagonal part of expectation values, defined as

$$\left\langle \frac{\partial P_{if}(t_1)}{\partial t_1}\right\rangle = \left\langle \left(\frac{\partial P_{if}(t_1)}{\partial t_1}\right)^d\right\rangle + \left\langle \left(\frac{\partial P_{if}(t_1)}{\partial t_1}\right)^{off}\right\rangle,$$
$$\left\langle \left(\frac{\partial P_{if}(t_1)}{\partial t_1}\right)^d\right\rangle = \omega_{if}\langle \hat{P}_{if}\rangle. \quad (13)$$

Within the Hartree-Fock like approximation, the average microscopic equation of motion.

$$\left\langle \frac{\partial P_{if}(t_1)}{\partial t_1}\right\rangle \cong \frac{i}{\hbar}\left\langle \left(\hat{\tilde{h}}^f_{ii} - \hat{\tilde{h}}^i_{ff}\right)\hat{P}_{if}\right\rangle \cong \frac{i}{\hbar}\left\langle \left(\hat{\tilde{h}}^f_{ii} - \hat{\tilde{h}}^i_{ff}\right)\right\rangle \left\langle \hat{P}_{if}\right\rangle = \omega_{if}\left\langle \hat{P}_{if}\right\rangle \quad (14)$$

leads to the results obtained in equations (9–11). It should be noted, that simple approximation (14) is exactly equivalent to that obtained by infinite series summarizing in equations (8–11). Upon inserting equation (4) into (12) and taking the off-diagonal term $h_{if}(t_1)$ into consideration we get

$$\hat{P}_{if}(t) = G_{if}(t-s)\hat{P}_{if}(s) + \int_s^t G_{if}(t-t_1)\hat{\tilde{h}}_{if}(t_1))(n_f - n_i)dt_1. \quad (15)$$

As it is known, the random phase approximation supposes the replacement of instantaneous field created by plasma with average one [15]. The micropolarization operator (15), calculated in the Hartree-Fock like approximation with using of average transition frequency, can be used next for evaluation of the response function, the light absorption coefficient and the spontaneous recombination rate. Equation (15) for many-body system is more realistic one comparison with equation (10). In arriving at equation (15) we have taken into account off-diagonal contribution in Hartree-Fock like approximation

$$\left\langle \left(\frac{\partial \hat{P}_{if}(t_1)}{\partial t_1}\right)^{off}_{HF}\right\rangle = \frac{i}{\hbar}\left\langle \left(\hat{P}_{ff} - \hat{P}_{ii}\right)\hat{\tilde{h}}_{fi}\right\rangle = \frac{i}{\hbar}\left\langle \hat{\tilde{h}}_{fi}\right\rangle (n_f - n_i). \quad (16)$$

This term leads to the important consequences for kinetic properties of solid states. Equations (11, 15, 16) at the level of dynamical HF approximation allow to include random phase like approximation into the consideration. To study the correlation contributions to $\omega_{if}(t)$ one has to go beyond the HF approximation. We start by considering

the deviations of the exact terms from their corresponding HF factorized parts

$$\left\langle \frac{\partial \hat{P}_{if}(t_1)}{\partial t_1} \right\rangle - \left\langle \left(\frac{\partial \hat{P}_{if}(t_1)}{\partial t_1} \right)^{d}_{HF} \right\rangle - \left\langle \left(\frac{\partial \hat{P}_{if}(t_1)}{\partial t_1} \right)^{off}_{HF} \right\rangle = \left\langle \left(\frac{\partial \hat{P}_{if}(t_1)}{\partial t_1} \right)_{C} \right\rangle. \quad (17)$$

One remarkable feature of equation (17) is that for the quasi-particle picture ($H \to H_q$) the correlation part is identically zero

$$\left\langle \left(\frac{\partial \hat{P}^q_{if}(t_1)}{\partial t_1} \right)_{C} \right\rangle = 0, \quad \hat{P}^q_{if}(t) = e^{\frac{i}{\hbar}\hat{H}_q t} \hat{P}_{if} e^{-\frac{i}{\hbar}\hat{H}_q t},$$
$$\hat{H}_q = \sum_{n\alpha} E_{n\alpha} \hat{P}^{\alpha}_{nn}. \quad (18)$$

The simple analysis shows that the exact solution for operator of evolution $\exp(-i\hat{H}t/\hbar)$ can be written in the form

$$\exp\left(-\frac{i}{\hbar}\hat{H}t\right) = \exp\left(-\frac{i}{\hbar}\hat{H}_q t\right) - \frac{i}{\hbar}\int_0^t \exp\left(-\frac{i}{\hbar}\hat{H}t\right) T \times \exp\left(\frac{i}{\hbar}\int_s^t \hat{V}(s_1)ds_1\right)\hat{V}(s)ds,$$
$$\hat{V} = \hat{H} - \hat{H}_q. \quad (19)$$

To develop the equation of motion for microscopic polarization with account of memory effect we use the approximation

$$\left\langle \left(\frac{\partial \hat{P}^{(}_{if}t_1)}{\partial t_1} \right)_{C} \right\rangle = \frac{i}{\hbar}\int_0^t \left\langle \left[\hat{V}(s), \left(\frac{\partial \hat{P}_{if}(t)}{\partial t} \right)_{C} \right] \right\rangle ds,$$
$$\hat{V}(t) = e^{\frac{i}{\hbar}\hat{H}t}\hat{V}e^{-\frac{i}{\hbar}\hat{H}t}. \quad (20)$$

It is instructive to note here that time-dependent operators are expressed in the Heisenberg picture. Consequently, a fluctuation-dissipation theorem [15] can be used to calculate the correlator density-density. Making use of equation (20) and averaging equation (12) we obtain

$$\omega_{if}(t) = \omega^{HF}_{if}(t) + \left\langle \hat{\omega}^f_i(t) - \hat{\omega}^{i+}_f(t) \right\rangle$$

$$\hat{\omega}^f_i(t) = \frac{i}{\hbar^2}\int_0^t \left\{ \hat{\tilde{h}}^f_{ii}(s) \left(\hat{\tilde{h}}^f_{ii}(t) - \hat{\tilde{h}}^i_{ff}(t) \right) + \sum_n^i G_{ni}(t-s) \Big\{ \hat{\tilde{h}}_{in}(s)\hat{\tilde{h}}_{ni}(t)\left(1 - n_n(s)\right) + \hat{\tilde{h}}_{ni}(t)\hat{\tilde{h}}_{in}(s) n_n(s) \Big\} ds. \quad (21)$$

At the level of the quasi-particle approximation $G_{ni}(t) = \exp(i\omega_{ni}t)$, were one ignores Coulomb interaction, equation (21) can be written as

$$\omega_{if}(t) = \omega^{HF}_{if}(t) + \omega^{(2)}_{if}(t),\ \omega^{(2)}_{if}(t) = \omega_i(t) - \omega^*_f(t),\ \omega_i(t) = \frac{1}{\hbar^2}\sum_n^i \frac{\left\langle \left|\hat{h}_{in}\right|^2 \right\rangle}{\omega_{in}} \left(1 - \exp(-i\omega_{in}t)\right). \quad (22)$$

Transition frequencies ω_{ni} have to be calculated self-consistently in quasi-particle picture. This approximation allows the evaluation of energy spectrum for close levels, when the perturbation theory is not valid. Coulomb interaction gives the contribution into equation (21).

$$\left\langle \hat{\tilde{h}}^C_{in}(t)\hat{\tilde{h}}^C_{ni}(s) \right\rangle = \sum_\kappa V^2_\kappa \left| e^{i\kappa r}_{in} \right|^2 \langle \rho_\kappa(t)\rho_\kappa(s)\rangle \quad (23)$$

which is proportional to V^2_κ and diverges as κ^{-4} at $\kappa \to 0$ if we restrict the consideration with second order correction of perturbation theory. To avoid this difficulty some authors replaces the unscreened Coulomb potential by its screened value. But screening should result from the proper treatment of correlations. The expectation value of charge density operator's multiplication $\langle \rho_\kappa(t)\rho_\kappa(s)\rangle$, taken in different time points in equation (23), specify memory effects. It can be precisely calculated within the fluctuation-dissipation theorem [15] and can be expressed in terms of structure factor or inverse dielectric function.

$$\langle \rho_\kappa(t)\rho_\kappa(s)\rangle = \frac{\hbar}{\pi V_\kappa}\int_{-\infty}^{\infty} \exp(-i\omega(t-s))[n(\omega) + 1]\mathrm{Im}\left\{ \frac{1}{\varepsilon^*(\kappa,\omega)} \right\} d\omega,$$
$$n(\omega) = \left[\exp\left(\frac{\hbar\omega}{k_0 T} \right) - 1 \right]^{-1}. \quad (24)$$

At this stage we already have a closed set of integral equations. The correlator density-density from equation (22) depends on the longitudinal dielectric function $\varepsilon(\kappa,\omega)$ equation (24) [15]. On the other hand dielectric function $\varepsilon(\kappa,\omega)$ depends on the micro-polarization in its turn. Hence equations (15, 21–24) constitute a self-consistent set of multi-quantum integral kinetic equations. The four-operator expectation values are multi-dimensional integrals over products of distribution functions and micro-polarization's with matrix elements of the dynamically screened Coulomb potential (24). The next section is used to present and discuss several results, which stem from this formulation.

3 Electron energy loss spectrum

Experiments in transmission electron energy loss spectroscopy determine the structure factor — correlator

density-density, which can be obtained from the imaginary part of inverse longitudinal dielectric function $\varepsilon(\kappa,\omega)$ by using of fluctuation dissipation theorem [15]. At the level of Born approximation, the collision rate $R(\kappa,\omega)$

$$R(\kappa,\omega) = 2V_\kappa(n(\omega)+1)\mathrm{Im}\left\{\frac{1}{\varepsilon^*(\kappa,\omega)}\right\},$$
$$V_\kappa = \frac{4\pi e^2}{V\kappa^2}, \tag{25}$$

depends on losses of electron energy $\hbar\omega$ and losses of momentum $\hbar\kappa$ which in theirs turn define energy and momentum of longitudinal plasmas eigenmodes. According to the energy and momentum conservation laws we have

$$\frac{\hbar^2\kappa^2}{2m} = 2E + \hbar\omega - 2\cos\theta\sqrt{E(E+\hbar\omega)}, \tag{26}$$

where E is the energy of electrons having passed through target, θ is the scattering angle. Valence electrons can be considered as practically free electrons at high energies $\approx\hbar\omega_p \cong 10$ eV and don't "feel" periodical potential of solid state. According to the definition [15] in linear approximation

$$\frac{1}{\varepsilon(\kappa,\omega)} = 1 + F(\kappa,\omega),$$
$$F(\kappa,\omega) = \frac{i}{\hbar}V_\kappa\int_0^\infty e^{i\omega t}\left\langle[\hat\rho_{-\kappa}(0),\hat\rho_\kappa(t)]\right\rangle dt,$$
$$\hat\rho_\kappa = \sum_k a_k^+ a_{k+\kappa}. \tag{27}$$

Using results obtained, inserting the equation (15) into (27), we can calculate the longitudinal dielectric function with account of multiquantum processes

$$\varepsilon(\kappa,\omega) = 1 - \frac{i}{\hbar}V_\kappa\sum_\kappa G_{k+\kappa,k}(\omega)(n_k - n_{k+\kappa})$$
$$\times\left\{1 - \frac{i}{\hbar}\sum_q V_q(n_{k+q} - n_{k+\kappa+q})G_{k+q,k+q+\kappa}(\omega)\right\},$$
$$n_k = \langle\hat P_{kk}\rangle. \tag{28}$$

We have included here the Coulomb-exchange contribution (Fock-field) to the polarization field. The results of the work [19] can be obtained from equations (27, 28) if one has calculated the transition frequency $\omega_{k,k+\kappa}$ equation (21) at the level of first order approximation of exchange-interacting electrons $\hbar\omega^{HF}_{k,k+\kappa} = e_k - e_{k+\kappa} - \sum_{kq} V_q(n_{k+q} - n_{k+q+\kappa})$. Equation (28) is integral equation for dielectric function $\varepsilon(\kappa,\omega)$, because there are integral dependence between the transition frequency

$$\hbar\omega_{k+\kappa\,k} = \tilde e_{k+\kappa} - \tilde e_k^*,$$
$$\tilde e_k = e_k - \sum_{kq} V_q n_{k+q} + i\int_0^t ds\int_{-\infty}^{\infty} d\nu$$
$$\times\sum_q K_q(\nu)\left\{n_{k+q}\exp(-i\nu(t-s))\right.$$
$$\left.+\,(1-n_{k+q})\exp(i\nu(t-s))\right\}G_{k+q,k}(t-s), \tag{29}$$

and dielectric function $\varepsilon(\kappa,\omega)$, which has to be calculated selfconsistently by iteration method, since according to the definition

$$K_q(v) = \frac{V_q}{\pi}(n_v+1)\mathrm{Im}\left\{\frac{1}{\varepsilon^*(q,v)}\right\} \tag{30}$$

the function $K_q(v)$ (30) is expressed in terms of dielectric function itself. The two first nonintegrative terms in equation (29) defines transition energy in Hartree-Fock like approximation, the second term describes the energy renormalization (memory correlation corrections to electrons energies) and multiquantum collision processes. The dynamical screening of correlation contribution in quasi-particle energy (time independent contribution in Eq. (29)) is taken into account and is in accordance with that obtained by Green function method [20] if one takes random phase approximation for dielectric function in equation (30). More than that, the Coulomb interaction leads to the multiquantum collision processes.

As it is known [15] in random phase approximation the dielectric function $\varepsilon(q,\nu)$ is equals to zero if $\nu = \omega_p = \left(4\pi Ne^2/m\right)^{1/2}$ (ω_p is the plasma frequency, N — the electron concentration). Considering plasmons as undamped excitations the contribution from high frequencies into integral over v (29) can be calculated using so-called plasmon-pole approximation

$$\mathrm{Im}\left\{\frac{1}{\varepsilon^*(q,\nu)}\right\} = \frac{\pi}{2}\omega_p[\delta(v-\omega_p) - \delta(v+\omega_p)]. \tag{31}$$

At small values of plasmon momentum q ($q < q_c$, $q_c = \omega_p/v_F$) plasma oscillations damp weakly and $\omega_{k,k+q} < \omega_p$. Hence we can get $G_{k+q,k}(t-s) \cong 1$ in equation (29). By taking into account of Landau damping γ_p and relaxation time of quasi-particles $1/\gamma$ we obtain

$$G_{k,k+\kappa}(t) = \exp\left\{i\omega^{HF}_{k,k+\kappa}t - \gamma t - N_p\right\}\sum_{n=-\infty}^{\infty} I_n(z)$$
$$\times\exp\{ny + in\omega_p t - n\gamma_p t\}, \tag{32}$$

where

$$z = N_p\left[1 - (n_k - n_{k+\kappa})^2\right]^{1/2},$$
$$\exp(y) = \frac{1 + n_k - n_{k+\kappa}}{1 + n_{k+\kappa} - n_{kk+\kappa}},$$
$$N_p = \frac{2}{\pi}(a_B k_F)^{-1} = 0,33\, r_s. \tag{33}$$

Damping coefficient γ is obtained at $t \to \infty$ limit, when $\lim_{t\to\infty}(1-e^{-i\omega t})/\omega^2 = \pi t\delta(\omega) + i\,\pi\, sign\, t\delta'(\omega)$ and is determined by imaginary part of dielectric function $\varepsilon(q,v)$ at frequencies $v = \omega_{k,q+\kappa}$ and $v = \omega_{k+q+\kappa,\kappa+q}$

$$\gamma = \frac{\pi}{\hbar}\sum_q [K_q(\omega_{k,k+q}) + K_q(\omega_{q+k+\kappa,k+q})]. \qquad (34)$$

The mean number of high-frequency plasmons N_p, generated in every collision process, is determined by the expression $N_p = 2e^2 q_c/\pi\hbar\omega_p$. For typical values of metal parameters the numerical value of N_p is more then one. Using sum rule for dielectric function in accordance with [3] we get $N_p = e^2\kappa_{FT}/\hbar\omega_p = \frac{\sqrt{3}}{a_B k_F} = 0.9 r_s$ ($\kappa_{FT}^2 = 6\pi N e^2/E_F$, κ_{FT}^{-1} is Thomas-Fermi radius). In the simplest kind of approximation for the polarization operator one obtains $\hat{P}_{k,k+\kappa}(t) = G_{k,k+\kappa}(t)\hat{P}_{k,k+\kappa}(0)$. Using equation (31) we get expression for collision rate equation (25) ($k_0 T \ll \hbar\omega_p$) in the form

$$R(\kappa,\omega) = \exp(-N_p)\sum_{n=-\infty}^{\infty} I_n(N_p) R^0(\kappa,\omega+n\omega_p),$$

$$R^0(\kappa,\omega+n\omega_p) = 2(n(\omega)+1)\mathrm{Im}Q^*(\kappa,\omega+n\omega_p). \qquad (35)$$

Here $R^0(\kappa,\omega)$ is the collision rate for non-interacting electron gas [15] without account of damping

$$Q(\kappa,\omega) = V_\kappa \sum_k \frac{n_k - n_{k+\kappa}}{e_k - e_{k+\kappa} + \hbar\omega + i\gamma}, \quad \gamma \to 0. \qquad (36)$$

Function $R(\kappa,\omega)$ can be transformed to $R^0(\kappa,\omega)$ at $N_p = 0$. In Figure 1 there are represented results of energy loss spectra calculations generated by equation (35) for different values of N_p and κ (plasmon momentum κ is in units of k_F, $k_F = (3\pi^2 N)^{1/3}$). As it is clear from equation (35) and Figure 1 electron energy loss spectra have multiplasmon structure. At $N_p > 1$ there is the Poison distribution for satellites intensities.

As the more realistic approximation for electron energy loss spectra function we use equations (28) and (32). For dielectric function we obtain the expression:

$$\varepsilon(\kappa,\omega) = 1 - \exp(-N_p) \times \sum_{n=-\infty}^{\infty} I_n(N_p) Q(\kappa,\omega+n\omega_p+in\gamma_p+i\gamma). \qquad (37)$$

Random phase approximation for dielectric function $\varepsilon(\kappa,\omega)$ is easy to obtain from equation (37) if we equate N_p to zero and neglect damping ($\gamma = \gamma_p = 0$). As we can see from equation (37) the frequency dependence for plasmonless ($n = 0$) and multiplasmon contributions in dielectric function is similar. Account of dispersion of plasmons and electrons kinetic transition energy $\hbar\omega_{k,k+q}$ in the function $G_{k,k+q}(t)$ leads to the additional enhancement of loss spectra line-width. Here we restrict the analysis in this paper to the simple situation, when the line-width of multiplasmon replicas is determined by the damping coefficients γ, γ_p and by the transition energy $e_k - e_{k+\kappa}$ in the function $G_{k+q,k}(t-s)$ in equation (31). The electron energy loss spectra resulting from our model are presented in Figure 2.

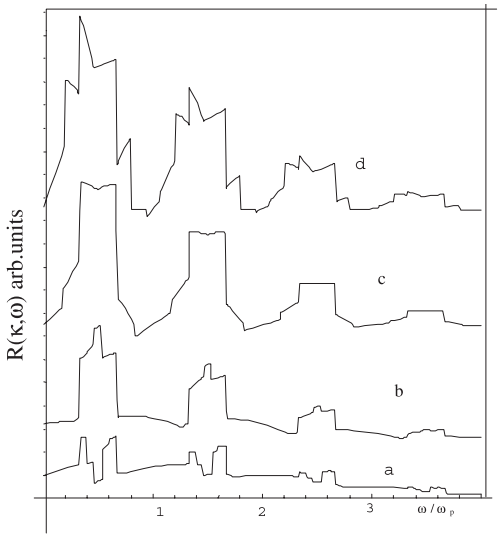

Fig. 1. Collision rate distribution of scattering electrons in metals calculated with (35), (36) at $\gamma = 0$, $N = 10^{23}$ cm^{-3}, $N_p = 2, 3$ and different values k: (a) $k = 0.9\ k_F$, (b) $k = 0.7\ k_F$, (c) $k = 0.5\ k_F$, (d) $k = 0.3\ k_F$.

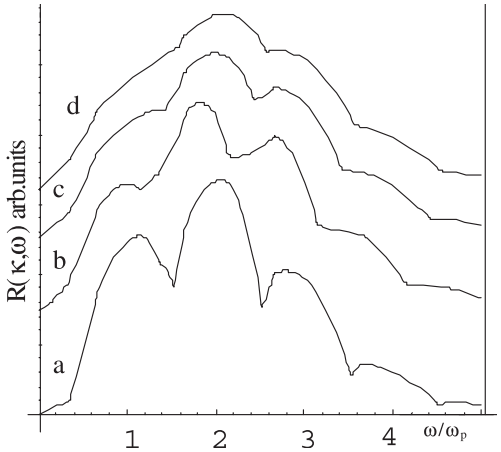

Fig. 2. Frequency dependence of collision rate R, calculated with using of equations (25) and (37), $G_{k+q,k}(t-s) \cong 1$ at different values of k, (a) $\kappa = 0.81\ k_F$, (b) $\kappa = 1.05\ k_F$, (c) $\kappa = 1.15\ k_F$, (d) $\kappa = 1.2\ k_F$, and $\gamma = 10^{-2}\omega_p$, $\gamma_p = 0$, $N = 10^{23}$ cm^{-3}, $N_p = 2$.

The results obtained for collision rate (Fig. 2) as function of ω are in accordance with experimental data [12–16]. Electron energy loss spectra have multiplasmon structure. If $N_p > 1$ the generation of a good few plasmons is simultaneous event not consecutive.

Many body interactions cause the electron energy loss spectra to change. This change is a result of simultaneous generation of several plasmons.

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