

ELECTRON COLLISION RATE AND MULTIQUANTUM TRANSITIONS IN SOLIDS

A. Klyukanov, A. Catanoi, A. Petrushca, and A. Awawdeh

*Department of Physics, Moldova State University, 60, A. Mateevich str., MD-2009,
Chisinau, Moldova*

E-mail: klukanov@cinf.usm.md

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Abstract

Multiquantum integral kinetic equation for the longitudinal complex dielectric function is derived. Heisenberg equation of motion and fluctuation-dissipation theorem has been used to calculate multiplasmon transmission electron energy loss spectra. Strong interaction between high-energy 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 of plasmons is simultaneous event. Calculated multiplasmon structure of electron energy loss spectra coincides with experimental one.

1. Introduction

The correlation effects give rise different collective and coherence effects which are intensively investigated at the present time [1-11]. 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]. A spectroscopic study of free-to-free, free-to-bound, and bound-to-bound e-h multiplasmon 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 ($\hbar\omega_p \cong 10 \text{ meV}$) as a typical electronic eigenmodes induced by band charge carrier correlations were investigated experimentally and theoretically in detail [1-6].

Emission of photon and that of several plasmons are simultaneous processes. 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 $\hbar\omega_p \cong 10 \text{ eV}$ plasmons $N_p = 1/a_B k_F = 0.52r_s$, which are given in terms of Bohr radius

$a_B = \frac{\hbar^2}{me^2}$ 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 relevant 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 is considered as consecutive process [12-16], characterized by electron mean free path concerning plasmon generation. This process consists of separate consecutive collisions with generation of one plasmon in each collision ($\omega = \omega_p$). In this work we investigate spectra of electron energy losses in solid state taking into the consideration simultaneous multiple generation of high-frequency plasmons. We give general integral multiquantum kinetic equation for a determination of a longitudinal dielectric function $\varepsilon(\mathbf{k}, \omega)$ derived on the basis of fluctuation-dissipation theorem. We have to mention at first that in works [1-6] 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 using any model approximation.

2. General considerations

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 $\varepsilon(\mathbf{k}, \omega)$ by using of fluctuation dissipation theorem [15]. In the frame of Born approximation, the collision rate $R(\mathbf{k}, \omega)$

$$R(\mathbf{k}, \omega) = 2V_{\kappa} (n(\omega) + 1) \text{Im} \left\{ \frac{1}{\varepsilon^*(\mathbf{k}, \omega)} \right\}, \quad V_{\kappa} = \frac{4\pi e^2}{V_{\kappa}^2} \quad (1)$$

depends on losses of electron energy $\hbar\omega$ and losses of momentum $\hbar\mathbf{k}$ which, in their turn, define energy and momentum of longitudinal plasma eigenmodes. According to the definition of [15] in linear approximation

$$\frac{1}{\varepsilon(\mathbf{k}, \omega)} = 1 + F(\mathbf{k}, \omega), \quad F(\mathbf{k}, \omega) = \frac{i}{\hbar} V_{\kappa} \int_0^{\infty} e^{i\omega t} \langle [\hat{\rho}_{-\mathbf{k}}(0), \hat{\rho}_{\mathbf{k}}(t)] \rangle dt, \quad \hat{\rho}_{\mathbf{k}} = \sum_{\mathbf{k}} a_{\mathbf{k}}^+ a_{\mathbf{k}+\mathbf{k}}. \quad (2)$$

In terms of creation and destruction operators according to formula (2) the time dependence of the micropolarization operator $\hat{P}_{k,k+\kappa}(t) = a_{\mathbf{k}}^+ a_{\mathbf{k}+\mathbf{k}}$ has to be derived from the Heisenberg equation of motion.

Using the notations and the results of paper [7] we obtain

$$P_{if}(t) = G_{if}(t) P_{if}(0) + \frac{i}{\hbar} \int_0^t G_{if}(t-t_1) \left\{ \sum_n (\hat{h}_{ni} P_{nf} - \hat{h}_{fn} P_{in})_{t_1} + \frac{i}{\hbar} \int_0^{t_1} \sum_{nm} \sum_{n_1 m_1} \langle \delta \hat{h}_{n_1 m_1}(t_2) \delta \hat{h}_{nm}(t_1) \rangle \right. \\ \left. \langle \delta \hat{P}_{n_1 m_1}(t_2) [\hat{P}_{nm}(t_1), \hat{P}_{if}(t_1)] \rangle - \langle \delta \hat{h}_{nm}(t_1) \delta \hat{h}_{n_1 m_1}(t_2) \rangle \langle [\hat{P}_{nm}(t_1), \hat{P}_{if}(t_1)] \delta \hat{P}_{n_1 m_1}(t_2) \rangle - \hbar \omega_{if} \hat{P}_{if}(t_1) \right\} dt_2, \quad (3)$$

The i, f subscripts represent the quantum number sets.

The longitudinal dielectric function $\varepsilon(\mathbf{k}, \omega)$ depends on the correlator density-density from Eq. (2). On the other hand, the micro-polarization $\hat{P}_{k,k+\kappa}(t) = a_{\mathbf{k}}^+ a_{\mathbf{k}+\mathbf{k}}$ depends on the dielectric function $\varepsilon(\mathbf{k}, \omega)$ in its turn. Hence, equations (1-3) constitute a self-consistent set of multi-quantum integral kinetic equations. Using the obtained results, we can calculate the longitudinal dielectric function with account of multiquantum processes

$$\varepsilon(\mathbf{k}, \omega) = 1 - \frac{i}{\hbar} V_{\kappa} \sum_{\mathbf{k}} G_{\mathbf{k}+\mathbf{k}, \mathbf{k}}(\omega) (n_{\mathbf{k}} - n_{\mathbf{k}+\mathbf{k}}) \left\{ 1 - \frac{i}{\hbar} \sum_{\mathbf{q}} V_{\mathbf{q}} (n_{\mathbf{k}+\mathbf{q}} - n_{\mathbf{k}+\mathbf{k}+\mathbf{q}}) G_{\mathbf{k}+\mathbf{q}, \mathbf{k}+\mathbf{q}+\mathbf{k}}(\omega) \right\}, \quad n_{\mathbf{k}} = \langle \hat{P}_{\mathbf{k}\mathbf{k}} \rangle. \quad (4)$$

Equation (4) is integral equation for dielectric function $\varepsilon(\mathbf{k}, \omega)$, because there is an integral dependence between the transition frequency

$$\hbar\omega_{\mathbf{k}+\mathbf{q},\mathbf{k}} = \tilde{e}_{\mathbf{k}+\mathbf{q}} - \tilde{e}_{\mathbf{k}}^*, \quad \tilde{e}_{\mathbf{k}} = e_{\mathbf{k}} - \sum_{\mathbf{q}} V_{\mathbf{q}} n_{\mathbf{k}+\mathbf{q}} +$$

$$+ i \int_0^t ds \int_{-\infty}^{\infty} d\nu \sum_{\mathbf{q}} K_{\mathbf{q}}(\nu) \{ n_{\mathbf{k}+\mathbf{q}} \exp(-i\nu(t-s)) + (1 - n_{\mathbf{k}+\mathbf{q}}) \exp(i\nu(t-s)) \} G_{\mathbf{k}+\mathbf{q},\mathbf{k}}(t-s) \quad (5)$$

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

$$K_{\mathbf{q}}(\nu) = \frac{V_{\mathbf{q}}}{\pi} (n_{\mathbf{q}} + 1) \text{Im} \left\{ \frac{1}{\varepsilon^*(\mathbf{q}, \nu)} \right\}, \quad G_{\mathbf{k},\mathbf{k}+\mathbf{q}}(t-s) = \exp \left\{ i \int_s^t \omega_{\mathbf{k},\mathbf{k}+\mathbf{q}}(t_1) dt_1 \right\} \quad (6)$$

the function $K_{\mathbf{q}}(\nu)$ (6) is expressed in terms of dielectric function itself. We have included here the Coulomb-exchange contribution (Fock-field) to the polarization field. The two first nonintegrative terms in Eq. (5) define transition energy in Hartree-Fock like approximation $\hbar\omega_{\mathbf{k},\mathbf{k}+\mathbf{q}}^{HF} = e_{\mathbf{k}} - e_{\mathbf{k}+\mathbf{q}} - \sum_{\mathbf{q}} V_{\mathbf{q}} (n_{\mathbf{k}+\mathbf{q}} - n_{\mathbf{k}+\mathbf{q}+\mathbf{q}})$, the next term describes the energy renormalization (memory correlation corrections to electron energies) and multiquantum collision processes. The dynamical screening of correlation contribution in quasi-particle energy (time independent contribution in Eq. (5)) is taken into account and, in accordance with that, is obtained by the Green function method. Moreover, the Coulomb interaction leads to the multiquantum collision processes.

3. Electron energy loss spectrum

As it is known [15], in random phase approximation the dielectric function $\varepsilon(\mathbf{q}, \nu)$ is equal to zero if $\nu = \omega_p = (4\pi N e^2 / m)^{1/2}$ (ω_p is the plasma frequency, N is the electron concentration). Considering plasmons as undamped excitations the contribution from high frequencies to integral over ν (6) can be calculated using so-called plasmon-pole approximation

$$\text{Im} \left\{ \frac{1}{\varepsilon^*(\mathbf{q}, \nu)} \right\} = \frac{\pi}{2} \omega_p [\delta(\nu - \omega_p) - \delta(\nu + \omega_p)]. \quad (7)$$

At small values of plasmon momentum q ($q < q_c$, $q_c = \omega_p / v_F$) plasma oscillations damp weakly and $\omega_{\mathbf{k},\mathbf{k}+\mathbf{q}} < \omega_p$. Hence, we can get $G_{\mathbf{k}+\mathbf{q},\mathbf{k}}(t-s) \cong 1$ in equation (5). By taking into account Landau damping γ_p and relaxation time of quasi-particles $\frac{1}{\gamma}$ we obtain

$$G_{\mathbf{k},\mathbf{k}+\mathbf{q}}(t) = \exp \{ i \omega_{\mathbf{k},\mathbf{k}+\mathbf{q}}^{HF} t - \gamma t - N_p \} \sum_{n=-\infty}^{\infty} I_n(z) \exp \{ n y + i n \omega_p t - n \gamma_p t \}, \quad (8)$$

where

$$z = N_p [1 - (n_{\mathbf{k}} - n_{\mathbf{k}+\mathbf{q}})^2]^{1/2}, \quad \exp(y) = \frac{1 + n_{\mathbf{k}} - n_{\mathbf{k}+\mathbf{q}}}{1 + n_{\mathbf{k}+\mathbf{q}} - n_{\mathbf{k}+\mathbf{q}+\mathbf{q}}}, \quad N_p = \frac{2}{\pi} (a_B k_F)^{-1} = 0,33 r_s. \quad (9)$$

Damping coefficient γ is obtained at $t \rightarrow \infty$ limit, when $\lim_{t \rightarrow \infty} \frac{1 - e^{-i\omega t}}{\omega^2} = \pi t \delta(\omega) + i \pi \text{sgnt} \delta'(\omega)$ and is determined by imaginary part of dielectric function $\varepsilon(q, \nu)$ at frequencies $\nu = \omega_{\mathbf{k},\mathbf{q}+\mathbf{q}}$ and $\nu = \omega_{\mathbf{k}+\mathbf{q},\mathbf{q}+\mathbf{q}}$.

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 than 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 $\hat{P}_{\mathbf{k}, \mathbf{k}+\mathbf{\kappa}}(t) = G_{\mathbf{k}, \mathbf{k}+\mathbf{\kappa}}(t) \hat{P}_{\mathbf{k}, \mathbf{k}+\mathbf{\kappa}}(0)$.

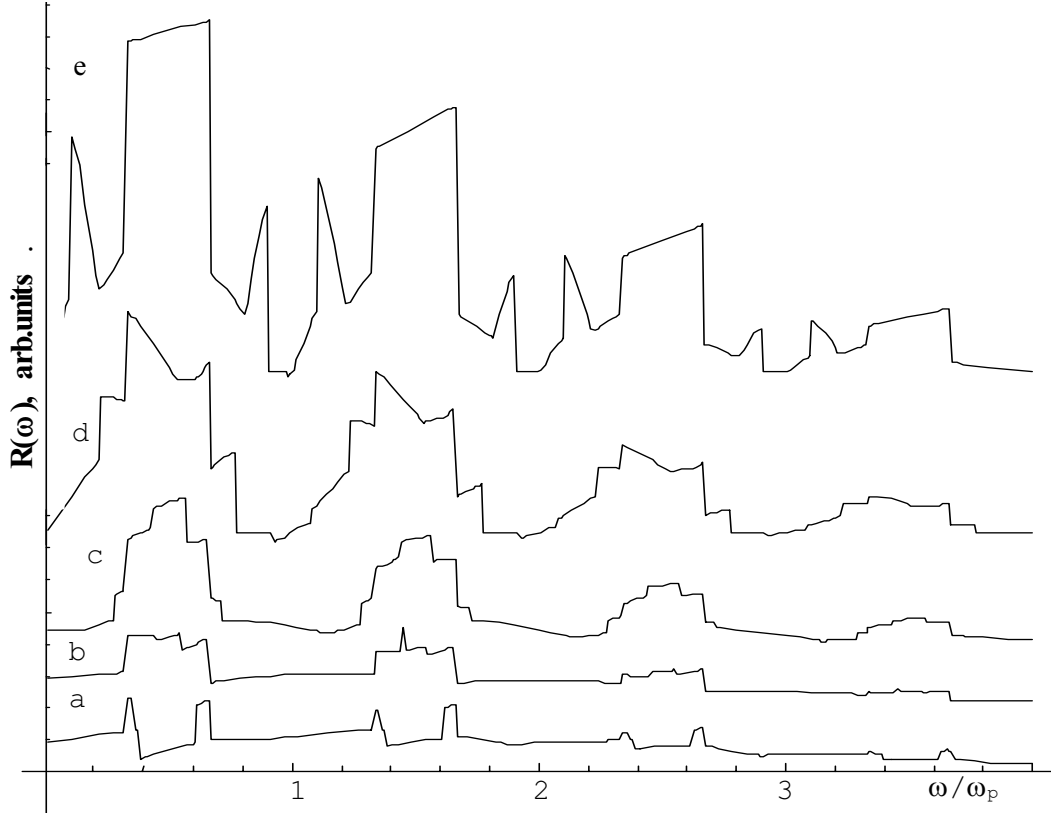


Fig. 1. Collision rate distribution of scattering electrons in metals calculated with (35), (36) at $\gamma = 0$, $N = 10^{23} \text{ cm}^{-3}$, $N_p = 2, 3$ and different values of κ : a) $\kappa = 0.96 k_F$, b) $\kappa = 0.82 k_F$, c) $\kappa = 0.65 k_F$, d) $\kappa = 0.34 k_F$, e) $\kappa = 0.15 k_F$.

Using Eq. (8) we obtain expression for collision rate ($k_0 T \ll \hbar \omega_p$) in the form

$$R(\mathbf{\kappa}, \omega) = \exp(-N_p) \sum_{n=-\infty}^{\infty} I_n(N_p) R^0(\mathbf{\kappa}, \omega + n\omega_p), \quad (10)$$

$$R^0(\mathbf{\kappa}, \omega + n\omega_p) = 2(n(\omega) + 1) \text{Im} Q^*(\mathbf{\kappa}, \omega + n\omega_p)$$

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

$$Q(\mathbf{\kappa}, \omega) = V_\kappa \sum_{\mathbf{k}} \frac{n_{\mathbf{k}} - n_{\mathbf{k}+\mathbf{\kappa}}}{e_{\mathbf{k}} - e_{\mathbf{k}+\mathbf{\kappa}} + \hbar \omega + i\gamma}, \quad \gamma \rightarrow 0. \quad (11)$$

Function $R(\mathbf{k}, \omega)$ can be transformed to $R^0(\mathbf{k}, \omega)$ at $N_p = 0$. In Fig. 1 there are represented results of energy loss spectra calculations generated by Eq. (10) 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 seen from Eq. (10) and Fig. 1, electron energy loss spectra have multiplasmon structure. At $N_p > 1$ there is the Poisson distribution for satellite intensities. As the more realistic approximation for electron energy loss spectra function we use Eqs. (8) and (4). For dielectric function we obtain the expression

$$\varepsilon(\mathbf{k}, \omega) = 1 - \exp(-N_p) \sum_{n=-\infty}^{\infty} I_n(N_p) Q(\mathbf{k}, \omega + n\omega_p + in\gamma_p + i\gamma). \quad (12)$$

Random phase approximation for dielectric function $\varepsilon(\mathbf{k}, \omega)$ is easy to obtain from Eq. (12) if we equate N_p to zero and neglect damping ($\gamma = \gamma_p = 0$). As we can see from Eq. (12) the frequency dependence for plasmonless ($n = 0$) and multiplasmon contributions in dielectric function is similar.

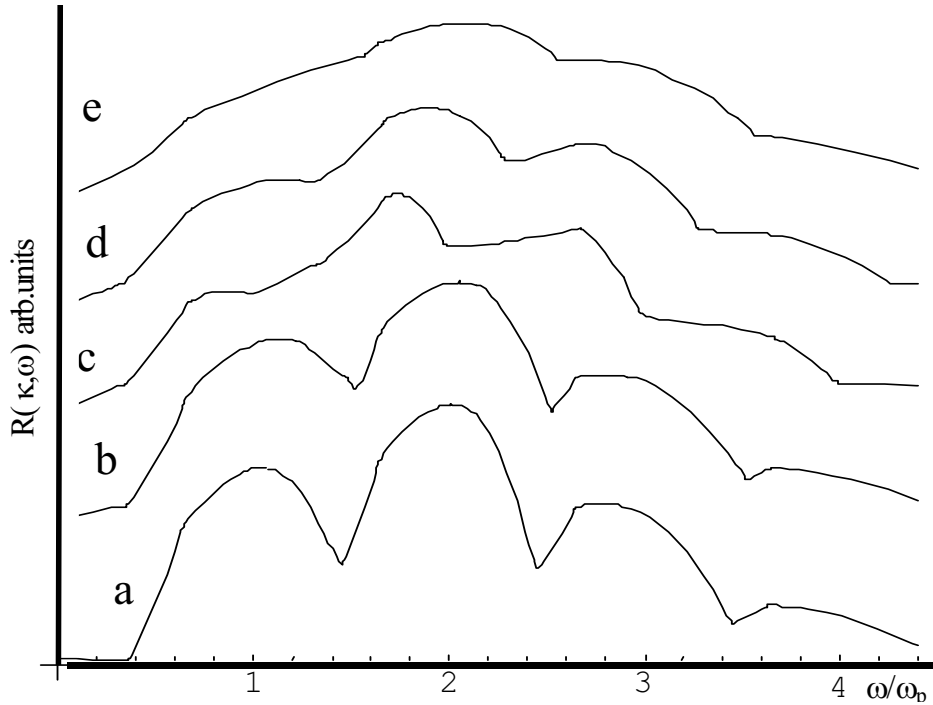


Fig. 2. Frequency dependence of collision rate R , calculated with using of Eq. (25) and Eq. (37) ($G_{\mathbf{k}+\mathbf{q},\mathbf{k}}(t-s) \cong 1$) at different values of κ : a) $\kappa = 0.78 k_F$, b) $\kappa = 0.81 k_F$, c) $\kappa = 0.99 k_F$, d) $\kappa = 1.1 k_F$, e) $\kappa = 1.2 k_F$ and $\gamma = 10^{-2} \omega_p$, $\gamma_p = 0$, $N = 10^{23} \text{ cm}^{-3}$, $N_p = 2$.

Account of dispersion of plasmons and electrons kinetic transition energy $\hbar\omega_{\mathbf{k},\mathbf{k}+\mathbf{q}}$ in the function $G_{\mathbf{k},\mathbf{k}+\mathbf{q}}(t)$ leads to the additional enhancement of loss spectra line-width. In this paper we restrict the analysis to the simple situation, when the line-width of multiphonon replicas is determined by the damping coefficients γ , γ_p and by the transition energy $e_{\mathbf{k}} - e_{\mathbf{k}+\mathbf{\kappa}}$ in the function $G_{\mathbf{k}+\mathbf{q},\mathbf{k}}(t-s)$ in Eq. (8). The electron energy loss spectra resulting from our model are

presented in Fig. 2. The results obtained for collision rate (Fig. 2) as a 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 of 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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