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V.V. Ignatyuk, V.G. Morozov

NON-MARKOVIAN QUANTUM KINETIC EQUATIONS FOR  
LASER-PULSE EXCITED SEMICONDUCTORS

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**Немарківські квантові кінетичні рівняння для напівпровідників у полі лазерного імпульсу**

В.В.Ігнатюк, В.Г. Морозов

**Анотація.** Отримано немарківські кінетичні рівняння в другому борнівському наближенні для двозонного напівпровідника в полі фемтосекундного лазерного імпульсу. Враховано як зіткненневу динаміку, так і внесок початкових кореляцій. Розглядається проблема балансу енергії при наявності квазічастинкового загасання, а також релаксація системи до рівноваги.

**Non-Markovian quantum kinetic equations for laser-pulse excited semiconductors**

V.V. Ignatyuk, V.G. Morozov

**Abstract.** We obtained non-markovian kinetic equations in the second Born approximation for two-zone semiconductor, excited by femtosecond laser pulse. Both collisional dynamics and initial correlations are taken into account. The problems of energy balance in the presence of quasiparticle damping and relaxation of the system to equilibrium is considered.



## 1. Introduction

The processes induced by short-lasting laser pulses in semiconductors permanently excite the attentions of technologists in microelectronics, experimentalists and theoretical physicists. Speaking about microelectronic technologies one has to mention that microelectronics technologists in the process of the creation of faster and faster switching devices reached “quantum limitation” on frequency when characteristics of the elements changed drastically. It could be said the scientists arrived at a restriction imposed by quantum uncertainty  $\Delta E \cdot \Delta \tau \sim \hbar$  itself, hence more experiments on ultrafast dynamics of semiconductors are needed to investigate new features in behaviour of such materials.

There are essentially two different classes of experiments for the study of carriers dynamics [1]: luminescence and pump-probe measurements. In both cases a pump pulse is used to generate electron-hole pairs and bring semiconductor into the state far from thermal equilibrium. In a luminescence experiment the radiation emitted in a direction different from that of incident pulse due to recombination processes is analyzed spectrally and/or temporally. Both carriers and photons dynamics are being described quantum mechanically that allows one to investigate spontaneous and/or stimulated emission and adsorption of the light in semiconductors.

In a pump-probe experiment the semiconductor is excited by a pump pulse travelling in a direction  $\mathbf{q}_1$  and the dynamics of the carriers induced by this excitation are studied by looking at some property related to a delayed pulse in a direction  $\mathbf{q}_2$ . In pump-probe experiments one obtains differential transmission/reflection spectra (DTS). Though the only values entering DTS are spectral characteristics of probe signal and change of interband polarization, to calculate complete dynamics of the excited semiconductor and, consequently, to verify experimental data, one has to take into account the distribution functions of carriers and phonons as well. Usually, light-matter interaction is considered to be classical one in pump-probe measurements while carrier-carrier interaction is of minor importance in comparison with carrier-phonon one, when a maximum pair density is about  $10^{17} \div 10^{18} \text{ cm}^{-3}$ . The investigation of ultrafast dynamics of laser excited semiconductors showed [1,2,10] that no purely empirical model with collision integrals in  $\tau$ -approximation manifested its efficiency.

Thus, the task for theoretists is to construct a reliable model that allows to describe kinetic processes in semiconductors taking into account all processes running in the systems: effects of “memory” - the

most important when one deals with phenomena on femtosecond scale, quantum coherence effects, influence of the dynamical correlations, formation of quasiparticle behaviour etc.

One can point out three main directions which are being followed during investigation of quantum kinetics in semiconductors: matrix density theory [2–6]; Green functions method [8–10]; the method of nonequilibrium statistical operator [11–13]. In our Introduction we touch upon main advantages and shortcomings of all these approaches.

In the theory of density matrix one deals with the chain of coupled equations for electron and phonon distribution functions [4,5]. In fact, this chain of equations appears to be infinite, incorporating more and more complex (with the increasing number of creation/annihilation operators) correlation functions. This system of equations is decoupled in the certain approximation in interaction, the obtained set of differential equations is being solved numerically or analyzed theoretically with appropriate initial conditions. Though such an approach formally admits taking into account higher correlations, there are some weak points here. The first one consists in the fact that it is impossible to trace the validity of conservation laws - the main point for any kinetic equation to be quite reliable. Another shortcoming is dealt with the lack of information about memory phenomena: the chain of equation does not contain time convolution, being in fact the system of differential equations with constant coefficients. Though taking into account higher correlations in the 4-th Born approximation manifested its efficiency in the framework of density matrix approach (for instance, one succeeded to obtain the solutions for one-particle distribution function which do not contain any artefact like negative parts and instabilities - the main problem of the 2-nd Born approximation), it was necessary to include time retardation. In Ref. [3] both time retardation for the electron distribution function and quasiparticle damping in the equation for polarization were taken into consideration. Temporal evolution of the electron distribution function was calculated both in Markovian and non-Markovian approximation. In the second case, in the electron distribution function phonon replica evolves as time goes by, but each of them starts rather broad before sharpening until reaching the zero-phonon shape. This could be considered as a fingerprint of quantum kinetics. Another manifestation of purely quantum phenomenon is quantum beating in interband polarization [2,10].

In the works by Kuznetsov [8,9] there were calculated relaxation times for electron distribution function in the approximation of phonon thermal bath and linear excitation as functions of excess energy and pulse duration. However, no quasiparticle damping was included into initial

kinetic equations, and the solution of coherent problem was taken as a zeroth approximation that does not allow to investigate an actual thermalization of the system. Green's function method was improved in Ref. [10], and kinetics of phonon was included in the chain of equations to obtain proper nonequilibrium one-particle distribution functions - for carriers and polarization. The equations for retarded/advanced Green functions were solved in the diagonal Wigner-Weisskopf approximation so that quasiparticle damping was not included into the field-dependent off-diagonal self-energy matrixes. Investigation showed well-pronounced quantum beats in the interband polarization - a typical feature of quantum coherence phenomena.

However, neither density matrix nor Green function method can incorporate nonequilibrium thermodynamics relations. On the other hand, the question how to include the initial correlations into the system, which are of great importance in such short-lasting processes, naturally appears. An attempt to do this has been performed in [12,13] where kinetic equations in the 2-nd Born approximation were obtained using the nonequilibrium statistical operator (NSO) formalism. Initial correlations were introduced by means of quasiequilibrium (relevant) distribution functions. The set of abbreviated description including one-particle densities and interaction energy was taken as a basic one. Collision integral consists of the two parts: due to collision dynamics and as a result of initial correlations. In the state of complete equilibrium both integrals compensate each other. It is worthy to note, that correlational contribution related to nonequilibrium temperature (quasitemperature) of the system vanishes in the Markovian approximation which indicates an existence of two typical relaxation times:  $\tau_{col}$  related to fast one-particle effects and  $\tau_{corr}$  connected with slower many-particle interactions. However, in the evaluation of kinetic equations, Wick's decomposition of the creation/annihilation operators was performed, hence correlations enter in the kinetic equations only indirectly - via mean value of interaction energy. Another point is that it is difficult to introduce a quasiparticle damping in the NSO method self-consistently. In Ref. [14] this problem was partially solved in the framework of the mixed (thermodynamic and time) Green's functions formalism. Despite of rather formidable structure of obtained expressions it is possible to solve them numerically for the simplest case of, say, electron-phonon relaxation.

However, the picture becomes much more complicated in the presence of the external field. Markovian picture is found to be ineffective in such a case [1,10], and one has to deal with the system of integro-differential equations for one-particle distribution functions of carriers,

phonons and polarization. The principal question is an introduction of slowly varying variable  $\langle H_{int} \rangle^t$  into the basic set of dynamic variable, since only taking into consideration of mean value of interaction energy allows one to obtain a proper transition from essentially nonequilibrium behaviour to Markovian one and, consequently, ensures equilibrium solution of the kinetic equations [12-14].

It is challenging to investigate whether introduction of interaction energy could stabilize the solutions of kinetic equations in the second Born approximation, or introduction of quasiparticle damping is necessary. We plan to touch upon all these problems in our paper on ultrafast kinetics in laser-excited semiconductors. The article is structured as follows: in the next Section we write down Hamiltonian, describing the basic interactions in laser-induced semiconductor and present coherent solutions to kinetic equations. In the third Section we construct collision integrals which are found to consist of two terms: due to collisional dynamics and due to consideration of initial correlations in the system. Additionally, the equation for quasitemperature will be obtained, and generalized heat capacity will be introduced. In the 4-th Section we consider some aspects related to energy balance requirement. In the Section 5 we examine a simplified version of the chain of kinetic equations in approximation of zero polarization. In fact, we deal with effective one-zone semiconductor investigated in Ref. [12]. Special attention will be paid to Markovian limit and transition of the system to equilibrium. The last Section contains some numerical estimations for relaxational dynamics of the excited two-zone semiconductor and discussion of the results.

## 2. Basic model of two-zone laser excited semiconductor

To describe the kinetics of laser-excited semiconductor let us introduce a Hamiltonian of the system:

$$H(t) = H_0 + H_f(t) + H_{int}, \quad (2.1)$$

where

$$H_0 = \sum_{\mathbf{k}, \alpha} e_{\alpha\mathbf{k}} a_{\alpha\mathbf{k}}^\dagger a_{\alpha\mathbf{k}} + \sum_{\mathbf{q}} \hbar\omega_{\mathbf{q}} \left( b_{\mathbf{q}}^\dagger b_{\mathbf{q}} + \frac{1}{2} \right) \quad (2.2)$$

denotes the kinetic part of the Hamiltonian in representation of secondary quantization,  $a_{\alpha\mathbf{k}}^\dagger$  and  $a_{\alpha\mathbf{k}}$ ,  $\alpha = \{c, v\}$  mean creation and annihilation operators of conduction ( $\alpha = c$ ) and valence electrons

( $\alpha = v$ ), while  $b_{\mathbf{q}}^\dagger$ ,  $b_{\mathbf{q}}$  correspond to creation and annihilation operators of phonons. We shall deal with parabolic two-band semiconductor; the energies of the electrons in the vicinity of the  $\Gamma$  point are to be described by the effective mass  $m_c$  of the electrons in the conduction band and  $m_v$  of the holes in the valence band:

$$e_{c\mathbf{k}} = \frac{E_g}{2} + \frac{\hbar^2 \mathbf{k}^2}{2m_c} = \frac{E_g}{2} + \varepsilon_{c\mathbf{k}}, \quad e_{v\mathbf{k}} = -\frac{E_g}{2} - \frac{\hbar^2 \mathbf{k}^2}{2m_v} = -\frac{E_g}{2} - \varepsilon_{v\mathbf{k}}. \quad (2.3)$$

Field-dependent part of Hamiltonian  $H_f(t)$  is presented in the form

$$H_f(t) = - \sum_{\alpha \neq \beta, \mathbf{k}} \mu_{\alpha\beta}(\mathbf{k}) a_{\alpha\mathbf{k}}^\dagger a_{\beta\mathbf{k}} E(t), \quad (2.4)$$

that describes the interband polarization of carriers by external field  $E(t)$  via optical matrix elements  $\mu_{\alpha\beta}(\mathbf{k})$ :

$$\mu_{cv}(\mathbf{k}) = e \int d\mathbf{r} u_{c\mathbf{k}} \frac{\mathbf{E}_0 \mathbf{r}}{E_0} u_{v\mathbf{k}} + \text{h.c.} \quad (2.5)$$

where denotation h.c. means Hermitian conjugation, and  $u_{\alpha\mathbf{k}}$  is the lattice periodic Bloch function of the band  $\alpha$  and the crystal momentum  $\mathbf{k}$ . Usually,  $\mu_{\alpha\beta}(\mathbf{k})$  are supposed to be constants,  $\mu_{\alpha\beta}(\mathbf{k}) \approx d_0$ ; they define Rabi energy  $\hbar\omega_R = d_0 E_0$  of the laser pulse [9,10], where  $E_0$  is the field amplitude.

Time profile of the external electric field could be approximated as a leading edge of the excitation pulse with duration  $\tau_p$  and central frequency  $\omega$  in the form

$$\mathbf{E}(t) = \mathbf{E}_0(t) \cos(\omega t), \quad \mathbf{E}_0(t) = \mathbf{E}_0 \exp[-t^2/\tau_p^2]. \quad (2.6)$$

Note that we deal with homogeneous system under uniform excitation, the length of the wave being determined by energy  $1 \div 3$  eV (characteristic gap-width in two-zone semiconductor) is about  $10^{-7}$  m that is much more than lattice period in several nanometers.

The last term  $H_{int}$  in the Hamiltonian, which is supposed to be small, describes the carrier-phonon interaction and could be presented as

$$H_{int} = \sum_{\alpha\mathbf{k}\mathbf{q}} D(\mathbf{q}) \left( b_{\mathbf{q}} a_{\alpha\mathbf{k}}^\dagger a_{\alpha\mathbf{k}-\mathbf{q}} + b_{\mathbf{q}}^\dagger a_{\alpha\mathbf{k}-\mathbf{q}}^\dagger a_{\alpha\mathbf{k}} \right), \quad (2.7)$$

where the constant of interaction  $D(\mathbf{q})$  could be chosen in the Fröhlich-type form [8,10]:

$$D(\mathbf{q})^2 = \frac{2\pi e_c^2 \omega_{LO}}{\epsilon^* \mathbf{q}^2},$$

$\hbar\omega_{LO}$  means the energy of a longitudinal optical phonon (in all subsequent calculations we shall deal with dispersionless longitudinal optical phonons, so we put  $\omega_{\mathbf{q}} = \omega_{LO}$  everywhere), and  $\epsilon^*$  denotes an effective dielectric constant. Note, that first two terms of the Hamiltonian could be presented in the matrix form as

$$H_0(t) \equiv H_0 + H_f(t) = \sum_{\alpha\beta, \mathbf{k}} M_{\alpha\beta}(\mathbf{k}, t) a_{\alpha\mathbf{k}}^\dagger a_{\beta\mathbf{k}},$$

$$M_{\alpha\beta}(\mathbf{k}, t) = \begin{pmatrix} e_{c\mathbf{k}} & -d_0 E(t) \\ -d_0 E(t) & e_{v\mathbf{k}} \end{pmatrix}. \quad (2.8)$$

The subject of our interest are the kinetic equations for nonequilibrium distribution functions of carriers  $f_{\alpha\beta}(\mathbf{k}, t)$  and phonons  $n(\mathbf{q}, t)$ , which can be determined as follows:

$$f_{\alpha\beta}(\mathbf{k}, t) = \begin{pmatrix} \langle a_{c\mathbf{k}}^\dagger a_{c\mathbf{k}} \rangle^t & \langle a_{v\mathbf{k}}^\dagger a_{c\mathbf{k}} \rangle^t \\ \langle a_{c\mathbf{k}}^\dagger a_{v\mathbf{k}} \rangle^t & \langle a_{v\mathbf{k}}^\dagger a_{v\mathbf{k}} \rangle^t \end{pmatrix} \equiv \begin{pmatrix} f_c(\mathbf{k}, t) & p(\mathbf{k}, t) \\ p^*(\mathbf{k}, t) & f_v(\mathbf{k}, t) \end{pmatrix},$$

$$n(\mathbf{q}, t) = \langle b_{\mathbf{q}}^\dagger b_{\mathbf{q}} \rangle^t, \quad (2.9)$$

where averaging is being performed over nonequilibrium statistical operator  $\rho(t)$  and the notation for polarization  $p(\mathbf{k}, t)$  from Ref. [9] is used. Taking into account the evolution expression for NSO  $\rho(t)$  (von Neumann equation)

$$\frac{\partial \rho(t)}{\partial t} + \frac{1}{i\hbar} [\rho(t), H(t)] = 0 \quad (2.10)$$

and definition for distribution functions, one can write down the kinetic equations in the following form [11]:

$$\frac{\partial f_{\alpha\beta}(\mathbf{k}, t)}{\partial t} - \frac{i}{\hbar} [M(\mathbf{k}, t), f(\mathbf{k}, t)]_{\alpha\beta} = I_{\alpha\beta}(\mathbf{k}, t), \quad (2.11)$$

$$\frac{\partial n(\mathbf{q}, t)}{\partial t} = I^{ph}(\mathbf{q}, t),$$

where  $[M(\mathbf{k}, t), f(\mathbf{k}, t)]_{\alpha\beta}$  denotes corresponding matrix elements of the commutator of two matrixes, while for collision integrals one can write down the following general expressions:

$$I_{\alpha\beta}(\mathbf{k}, t) = \frac{1}{i\hbar} \text{Tr} \left\{ [\hat{f}_{\alpha\beta}(k), H_{int}] \rho(t) \right\},$$

$$I^{ph}(\mathbf{q}, t) = \frac{1}{i\hbar} \text{Tr} \left\{ [\hat{n}(\mathbf{q}), H_{int}] \rho(t) \right\}. \quad (2.12)$$

The former kinetic equation (2.11) without collision integral  $I_{\alpha\beta}(\mathbf{k}, t)$  is known as a coherent approximation<sup>1</sup>: in the Green's functions formalism [8,10] it corresponds to singular terms (coherent part) of self-energy matrix only. Coherent kinetic equations could be written as follows:

$$\begin{aligned} \frac{\partial f_c(\mathbf{k}, t)}{\partial t} &= -2\text{Im}(p^*(\mathbf{k}, t)d_0E(t)), \\ \frac{\partial f_v(\mathbf{k}, t)}{\partial t} &= -\left(\frac{\partial f_c(\mathbf{k}, t)}{\partial t}\right), \end{aligned} \quad (2.13)$$

$$\frac{\partial p(\mathbf{k}, t)}{\partial t} = -\frac{i}{\hbar}(e_{c\mathbf{k}} - e_{v\mathbf{k}})p(\mathbf{k}, t) + \frac{i}{\hbar}d_0E(t)(f_c(\mathbf{k}, t) - f_v(\mathbf{k}, t)).$$

Relations (2.13) are nothing but the Bloch equations which are widely used to describe the optical response of a two-level system. A general solution of these equations for an arbitrary form of  $E(t)$  is not known; however, Eqs. (2.13) yield a useful integral of motion [9]:

$$4|p(\mathbf{k}, t)|^2 + (f_c(\mathbf{k}, t) - f_v(\mathbf{k}, t))^2 = 1 \quad (2.14)$$

for radius of the Bloch sphere, which makes clear that as long as there is no relaxation,  $p(\mathbf{k}, t)$  and  $f_c(\mathbf{k}, t)$  are generally of the same order. This is particularly obvious in the low excitation limit ( $f_c \ll 1$ ), where (2.13) reduces to

$$|p(\mathbf{k}, t)|^2 \approx f_c(\mathbf{k}, t) \approx 1 - f_v(\mathbf{k}, t).$$

In the process of relaxation due to electron-phonon interaction, polarization finally decreases because of the finite spectral width of the pulse. In the last Section we perform numerical calculation in the assumption that non-diagonal elements of distribution function, related with interband polarization, damp faster than diagonal ones. This situation is well known in quantum kinetics [11], though during pulse transmission it is necessary to consider polarization on equal terms with diagonal elements of distribution functions.

### 3. Collision integrals and equation for quasitemperature

To obtain an explicit form of collision integrals (2.12) one has to construct a general solution of von Neumann equation. To do this let us supply the evolution equation (2.10) with initial conditions  $\rho(t_0) = \rho_{rel}(t_0)$ .

<sup>1</sup>It is often called as "collisionless" approximation

Following the standard procedure [12,13], we define relevant statistical operator  $\rho_{rel}(t)$  in generalized Gibbs form as

$$\rho_{rel}(t) = \frac{1}{Z_{rel}(t)} \exp\{-\beta(t)H_{int} - \sum_{\alpha\beta, \mathbf{k}\mathbf{q}} \Lambda_{\alpha\beta}(\mathbf{k}, t)\hat{f}_{\alpha\beta}(\mathbf{k}) - \Lambda^{ph}(\mathbf{q}, t)\hat{n}(\mathbf{q})\}, \quad (3.1)$$

$$Z_{rel}(t) = \text{Tr} \exp\{-\beta(t)H_{int} - \sum_{\alpha\beta, \mathbf{k}\mathbf{q}} \Lambda_{\alpha\beta}(\mathbf{k}, t)\hat{f}_{\alpha\beta}(\mathbf{k}) - \Lambda^{ph}(\mathbf{q}, t)\hat{n}(\mathbf{q})\}, \quad (3.2)$$

where Lagrange multipliers  $\beta(t)$ ,  $\Lambda_{\alpha\beta}(\mathbf{k}, t)$ ,  $\Lambda^{ph}(\mathbf{q}, t)$  should be calculated from self-consistency conditions

$$\langle a_{\beta\mathbf{k}}^\dagger a_{\alpha\mathbf{k}} \rangle_{rel}^t = f_{\alpha\beta}(\mathbf{k}, t), \quad \langle b_{\mathbf{q}}^\dagger b_{\mathbf{q}} \rangle_{rel}^t = n(\mathbf{q}, t), \quad \langle H_{int} \rangle_{rel}^t = \langle H_{int} \rangle^t \quad (3.3)$$

and are attributed to nonequilibrium thermodynamic forces. For instance,  $\beta(t)$  corresponds to nonequilibrium inverse temperature. Later we shall demonstrate that generalized temperature  $T(t)$  on large time scales determines an equilibrium background to which an electron temperature tends from above and phonon temperature from below, as long as one operates with subsystem temperatures. Hence, being initially determined as Lagrange multiplier,  $T(t)$  subsequently is associated with a generalized thermodynamic characteristic of the system; besides,  $T(t)$  tends to its equilibrium value  $T_{eq}$  when  $t \rightarrow \infty$ . Note, that we included  $H_{int}$  into the basic set of abbreviated description variables. In Green's functions formalism it corresponds to introduction of temperature and mixed Green's functions [14] when Keldysh contour goes along both time and thermodynamic axes. The advantage of NSO method consists in the direct introduction of generalized temperature  $T(t)$  while in the mixed Green's function formalism one can talk only about initial value of correlation function. It is a common problem how to introduce generalized temperature in the framework of Green's functions method.

Now we are in a position to obtain an iterative solution to von Neumann equation (2.10). In the second Born approximation one can present NSO  $\rho(t)$  in the following form, neglecting higher orders in interaction [12]:

$$\rho(t) = \rho_{rel}(t) - \int_{t_0}^t dt' U_0(t, t') \frac{1}{i\hbar} [\rho_{rel}(t'), H_0(t') + H_{int}] U_0^+(t, t'). \quad (3.4)$$

According to the definition (2.12), collision integrals in the second Born

approximations could be written as follows [12,13]:

$$I_{\alpha\beta}(\mathbf{k}, t) = -\frac{1}{\hbar^2} \int_{t_0}^t dt' \text{Tr} \left\{ \left[ U_0^+(t, t') \left[ a_{\alpha\mathbf{k}}^\dagger a_{\beta\mathbf{k}}, H_{int} \right] U_0(t, t'), H_{int} \right] \rho_{rel}(t') \right\} \quad (3.5)$$

$$+ \frac{1}{\hbar^2} \int_{t_0}^t dt' \text{Tr} \left\{ U_0^+(t, t') \left[ a_{\alpha\mathbf{k}}^\dagger a_{\beta\mathbf{k}}, H_{int} \right] U_0(t, t') [\rho_{rel}(t'), H_0(t')] \right\},$$

$$I^{ph}(\mathbf{q}, t) = -\frac{1}{\hbar^2} \int_{t_0}^t dt' \text{Tr} \left\{ \left[ U_0^+(t, t') \left[ b_{\mathbf{q}}^\dagger b_{\mathbf{q}}, H_{int} \right] U_0(t, t'), H_{int} \right] \rho_{rel}(t') \right\} \quad (3.6)$$

$$+ \frac{1}{\hbar^2} \int_{t_0}^t dt' \text{Tr} \left\{ U_0^+(t, t') \left[ b_{\mathbf{q}}^\dagger b_{\mathbf{q}}, H_{int} \right] U_0(t, t') [\rho_{rel}(t'), H_0(t')] \right\}.$$

In Eqns.(3.5)-(3.6)  $U_0(t, t') = \exp_+ \left\{ -\frac{i}{\hbar} \int_{t'}^t H_0(\tau) d\tau \right\}$  denotes an evolution operator with time-ordered exponent. The action of  $U_0^+(t, t')$ ,  $U_0(t, t')$  on corresponding operators could be calculated explicitly only for bosons:

$$U_0^+(t, t') b_{\mathbf{q}} U_0(t, t') = \exp[-i\omega_{\mathbf{q}}(t - t')] b_{\mathbf{q}}, \quad (3.7)$$

$$U_0^+(t, t') b_{\mathbf{q}}^\dagger U_0(t, t') = \exp[i\omega_{\mathbf{q}}(t - t')] b_{\mathbf{q}}^\dagger,$$

while for fermions the problem does not have an analytical solution. We present corresponding transformations in a symbolic matrix form via time-ordered exponent:

$$U_0^+(t, t') a_{\alpha\mathbf{k}} U_0(t, t') = \exp_+ \left[ -\frac{i}{\hbar} \int_{t'}^t M(\mathbf{k}, t'') dt'' \right]_{\alpha\beta} a_{\beta\mathbf{k}}, \quad (3.8)$$

$$U_0^+(t, t') a_{\alpha\mathbf{k}}^\dagger U_0(t, t') = \exp_- \left[ \frac{i}{\hbar} \int_{t'}^t M(\mathbf{k}, t'') dt'' \right]_{\alpha\beta} a_{\beta\mathbf{k}}^\dagger, \quad (3.9)$$

Up to a factor  $i\theta(t - t')$ , the time-ordered exponents in (3.8)-(3.9), determined by single particle energy matrix  $M(\mathbf{k}, t)$  from Eqn.(2.8) are

nothing but retarded  $G^r(\mathbf{k}; t, t')$  and advanced  $G^a(\mathbf{k}; t, t')$  Green's functions, respectively [10,11]. We will keep the generalized matrix notations for further convenience.

Thus, prior to solving of kinetic equations, one has to solve the Green's function problem<sup>2</sup>. Formally, kinetic kernels (3.8) could be found from relation

$$\frac{\partial A_{\alpha\beta}(\mathbf{k}, t)}{\partial t} = -\frac{i}{\hbar} \sum_{\gamma} M_{\alpha\gamma}(\mathbf{k}, t) A_{\gamma\beta}(\mathbf{k}, t) \quad (3.10)$$

and written down in the following way:

$$\exp_+ \left[ -\frac{i}{\hbar} \int_{t'}^t M(\mathbf{k}, t'') dt'' \right]_{\alpha\beta} = \sum_{\gamma} A_{\alpha\gamma}(\mathbf{k}, t) [A(\mathbf{k}, t')]_{\gamma\beta}^{-1}, \quad (3.11)$$

where  $M(\mathbf{k}, t)$  denotes single particle elements (2.8).

In general case of time-dependent perturbation, the system of equations (3.10) could be solved only numerically [10]; only linear in field amplitude approximation admits analytical solution, and this case is considered in Appendix A. However, since we are about to verify the results of [10], we restrict ourselves to unperturbed Wigner-Weisskopf approximation for matrix exponents  $\exp_+ \left[ -\frac{i}{\hbar} \int_{t'}^t M(\mathbf{k}, t'') dt'' \right]_{\alpha\beta}$ ,  $\exp_- \left[ \frac{i}{\hbar} \int_{t'}^t M(\mathbf{k}, t'') dt'' \right]_{\alpha\beta}$ , taking into consideration in our numerical calculations only the diagonal elements. Off-diagonal elements, vanishing in the limit of zero field, give the main contribution in the energy domain  $\Delta = \hbar\omega - e_{c\mathbf{k}} - e_{v\mathbf{k}}$ , which is close to an excess energy region and could be neglected in the problem of the relaxation to equilibrium. Note that our approach does not impose any limitation on the strength of  $E(t)$ ; the only requirement for the electric field is not to cause damages in crystal lattice.

Now we are in a position to calculate the first terms in (3.5)-(3.6). We call them ‘‘collisional’’ contributions in total collision integrals  $I_{\alpha\beta}(\mathbf{k}, t)$ ,  $I^{ph}(\mathbf{q}, t)$ , in order to distinguish them from ‘‘correlational’’ ones, appearing because of taking into consideration initial correlations in the system. Using Wick's decomposition of the operators, we have:

$$I_{\alpha\beta}^{(coll)}(\mathbf{k}, t) = -\frac{1}{\hbar^2} \int_{t_0}^t dt' \sum_{\mathbf{q}} D(\mathbf{q})^2 \left\{ \exp[-i\omega_{LO}(t - t')] \times \right.$$

<sup>2</sup>We shall use a term ‘‘Green's function’’ to denote time-ordered exponents (3.8) when appropriate.

$$\begin{aligned} & \times \exp_+ \left[ -\frac{i}{\hbar} \int_{t'}^t M(\mathbf{k} - \mathbf{q}, t'') dt'' \right] \cdot \\ & \{N^+(\mathbf{q})(\mathcal{I} - f(\mathbf{k} - \mathbf{q})) \cdot f(\mathbf{k}) - N^-(\mathbf{q})f(\mathbf{k} - \mathbf{q}) \cdot (\mathcal{I} - f(\mathbf{k}))\}_{t'} \cdot \\ & \left. \exp_- \left[ \frac{i}{\hbar} \int_{t'}^t M(\mathbf{k}, t'') dt'' \right] + \text{h.c.} \right\}_{\alpha\beta} - (\mathbf{k} \longleftrightarrow \mathbf{k} - \mathbf{q}), \quad (3.12) \end{aligned}$$

$$\begin{aligned} I_{ph}^{(coll)}(\mathbf{q}, t) &= -\frac{1}{\hbar^2} \int_{t_0}^t dt' D(\mathbf{q}^2) \sum_{\mathbf{k}} \text{Tr} \left\{ \exp[-i\omega_{LO}(t - t')] \right. \\ & \times \exp_+ \left[ -\frac{i}{\hbar} \int_{t'}^t M(\mathbf{k} - \mathbf{q}, t'') dt'' \right] \cdot \\ & \{N^+(\mathbf{q})(\mathcal{I} - f(\mathbf{k} - \mathbf{q})) \cdot f(\mathbf{k}) - N^-(\mathbf{q})f(\mathbf{k} - \mathbf{q}) \cdot (\mathcal{I} - f(\mathbf{k}))\}_{t'} \cdot \\ & \left. \exp_- \left[ \frac{i}{\hbar} \int_{t'}^t M(\mathbf{k}, t'') dt'' \right] + \text{h.c.} \right\}. \quad (3.13) \end{aligned}$$

Here we denote  $N^-(\mathbf{q}, t) = n(\mathbf{q}, t)$ ,  $N^+(\mathbf{q}, t) = 1 + n(\mathbf{q}, t)$ , use the symbol  $\cdot$  for matrix product and  $\text{Tr}$  for trace over zone indexes, while  $\mathcal{I}$  is a unit matrix. Though the form of  $I_{\alpha\beta}^{(coll)}(\mathbf{k}, t)$  looks rather complicated, it is seen that (3.12)-(3.13) have a well defined “gain–loss” structure. Taking only diagonal elements of the time-ordered exponents (that corresponds to Wigner-Weisskopf approximation for Green’s functions) we can write down the following expression:

$$\begin{aligned} & \exp_- \left[ \frac{i}{\hbar} \int_{t'}^t M(\mathbf{k}, t'') dt'' \right]_{\alpha\mu} \exp_+ \left[ \frac{-i}{\hbar} \int_{t'}^t M(\mathbf{k} - \mathbf{q}, t'') dt'' \right]_{\beta\nu} \rightarrow \\ & \exp \left[ \frac{i}{\hbar} (e_{\alpha\mathbf{k}} - e_{\beta\mathbf{k}-\mathbf{q}})(t - t') \right] \delta_{\alpha\mu} \delta_{\beta\nu}. \end{aligned}$$

However, the mentioned here approximation for kinetic kernels has one essential shortcoming: its field-free form does not obey energy balance equations. We shall come back to this important question in the next Section.

Kinetic equations with collision integrals (3.12)-(3.13) turn out to be in a closed form, since we neglect a contribution of correlation after decoupling of Bose and Fermi operators. Taking into account of higher correlations leads to quasiparticle damping (related to the imaginary part of self-energy operator in Green’s function method [10]). In the density matrix approach it corresponds to consideration of equations for higher correlation functions. Again, the question how to cut off the hierarchy of equations with not allowing the energy disbalance arises [3,4]. Recently the procedure of obtaining of quasiparticle damping function after the solution of quantum B-B-G-K-I hierarchy has been proposed in Ref. [18]. However, this procedure seems to be quite complicated to be strictly implemented in numerical solution of kinetic equations (in contrast to Green’s functions method when the equation for quasiparticle damping function  $\gamma(\mathbf{k}, t, t')$  supplements the system of kinetic equations [14].) In the method of nonequilibrium statistical operator one encounters similar difficulties describing formation of long-time quasiparticle behaviour of the system. Usually quasiparticle damping factor  $\exp[-\gamma(t-t')]$  serves as a fitting parameter, the system stabilizing behaviour at long times and preventing numerical solutions of kinetic equations from non-physical effects (electron and hole distribution functions should be in the interval [0,1]). Summing up all mentioned above, we shall treat quasiparticle damping as a result of all interaction processes in the system which did not enter the kinetic equations explicitly, keeping in mind that, in the first turn, quasiparticle behaviour arises from exceeding the bounds of second Born approximation <sup>3</sup>

Remind that we take into consideration the equation for interaction energy  $\langle H_{int} \rangle_t$ . Hence, we take into account *running correlations* in the system, though we neglect correlations during Wick’s decoupling. Taking into consideration the equation for  $\langle H_{int} \rangle_t$  is known to be of essential importance when describing relaxation toward equilibrium [13,16]; our numerical estimations completely confirm the theory predictions.

The contribution of dynamic correlations is being determined by the second terms in (3.5)-(3.6). We shall call them “correlational” to underline their dependence on initial correlations introduced by the relevant distribution function  $\rho_{rel}(t)$ , Eqn.(3.1). After some algebra (see Appendix B) we come to the explicit form of correlational constituents of the collision integrals:

<sup>3</sup>Often one can encounter the term “collision broadening” which is found to be equivalent to quasiparticle damping. It originates from Lorentzian shape of spectral function while one has energy-conserving delta-function in case of zero  $\gamma$ .



$$\begin{aligned}
I_{\alpha\beta}^{(corr)}(\mathbf{k}, t) = & -\frac{1}{\hbar^2} \int_{t_0}^t dt' \sum_{\mathbf{q}} D(\mathbf{q})^2 \beta(t') \left\{ \exp[-i\omega_{LO}(t-t')] \right. \\
& \times \exp_+ \left[ -\frac{i}{\hbar} \int_{t'}^t M(\mathbf{k}-\mathbf{q}, t'') dt'' \right] \cdot \\
& \{ N^+(\mathbf{q}, t') (\mathcal{I} - f(\mathbf{k}-\mathbf{q}, t')) \cdot B(\mathbf{k}, \mathbf{k}-\mathbf{q}, t') \cdot f(\mathbf{k}, t') \\
& - N^-(\mathbf{q}, t') f(\mathbf{k}-\mathbf{q}, t') \cdot B(\mathbf{k}, \mathbf{k}-\mathbf{q}, t') \cdot (\mathcal{I} - f(\mathbf{k}, t')) \} \\
& \cdot \exp_- \left[ \frac{i}{\hbar} \int_{t'}^t M(\mathbf{k}, t'') dt'' \right]_{\alpha\beta} + \text{h.c.} \left. \right\} - (\mathbf{k} \longleftrightarrow \mathbf{k}-\mathbf{q}), \quad (3.14)
\end{aligned}$$

where matrix  $B(\mathbf{k}, \mathbf{k}-\mathbf{q}, t)$  is being defined as follows:

$$B(\mathbf{k}, \mathbf{k}-\mathbf{q}, t) = \lim_{x \rightarrow 0} \int dx \exp[x\Lambda^{ph}(\mathbf{q}, t)] \exp[-x\Lambda(\mathbf{k}, t)]. \quad (3.15)$$

$$(M(\mathbf{k}, t) - M(\mathbf{k}-\mathbf{q}, t) - \hbar\omega_{LO}\mathcal{I}) \cdot \exp[x\Lambda(\mathbf{k}-\mathbf{q}, t)],$$

$\beta(t) = 1/T(t)$  means generalized inverse temperature, the explicit expressions for Lagrange multipliers could be found in Appendix B, and the equation for phonon correlational counterpart is of a similar structure, see Eqn. (3.13). To obtain a closed set of equations we have to compose the equation for quasitemperature  $\beta(t)$ . We proceed in the similar manner as in the Ref. [12]. Again, we start from the self-consistency conditions (3.3) which could be rewritten in a somewhat different form, taking into account the work of the external field:

$$\begin{aligned}
\text{Tr} \left\{ \hat{f}_{\alpha\beta}(\mathbf{k}) \frac{\partial \rho_{rel}(t)}{\partial t} \right\} = I_{\alpha\beta}(\mathbf{k}, t), \quad \text{Tr} \left\{ \hat{n}(\mathbf{q}) \frac{\partial \rho_{rel}(t)}{\partial t} \right\} = I^{ph}(\mathbf{q}, t), \\
\text{Tr} \left\{ \hat{H}_{int} \frac{\partial \rho_{rel}(t)}{\partial t} \right\} = - \sum_{\mathbf{q}} \hbar\omega_{\mathbf{q}} I^{ph}(\mathbf{q}, t) - \sum_{\alpha\mathbf{k}} e_{\alpha\mathbf{k}} I_{\alpha\alpha}(\mathbf{k}, t) \\
+ \sum_{\alpha \neq \beta \mathbf{k}} I_{\alpha\beta}(\mathbf{k}, t) d_0 E(t). \quad (3.16)
\end{aligned}$$

Note that the coherent term - the second summand  $[M(\mathbf{k}, t), f(\mathbf{k}, t)]_{\alpha\beta}$  from Eqn. (2.11) with external field does not contribute to balance equation: only the collisional dynamics determines the equation for generalized temperature.

The last equation for mean interaction energy could be rewritten as an equation for  $\beta(t)$ , taking into account the time dependence of the relevant statistical operator via time dependence of Lagrange multipliers. One can write down:

$$\begin{aligned}
\frac{d\beta(t)}{dt} = \frac{1}{C(t)} \left\{ \sum_{\alpha\mathbf{k}} \varepsilon_{\alpha}(\mathbf{k}) I_{\alpha\alpha}(\mathbf{k}, t) + \sum_{\mathbf{q}} \hbar\omega_{\mathbf{q}} I^{ph}(\mathbf{q}, t) \right. \\
\left. - \sum_{\alpha \neq \beta \mathbf{k}} I_{\alpha\beta}(\mathbf{k}, t) d_0 E(t) \right\}, \quad (3.17)
\end{aligned}$$

where we use denotation  $C(t) = \left( \hat{H}_{int}, \hat{H}_{int} \right)_t$  for nonequilibrium correlation function “energy–energy” defined in a usual manner:

$$\left( \hat{A}, \hat{B} \right)_t = \int_0^1 dx \langle \Delta \hat{A} \tilde{\rho}_{rel}^x \Delta \hat{B} \tilde{\rho}_{rel}^{-x} \rangle_{rel}, \quad \Delta \hat{A} = \hat{A} - \langle \hat{A} \rangle_{rel}. \quad (3.18)$$

Again, using Wick’s decomposition we can present the following expression for the generalized heat capacity  $C(t)$  via one-particle nonequilibrium distribution functions:

$$\begin{aligned}
C(t) = \sum_{\mathbf{k}\mathbf{q}} D(\mathbf{q})^2 \text{Tr} \left\{ N^-(\mathbf{q}, t) f(\mathbf{k}-\mathbf{q}, t) \cdot B(\mathbf{k}, \mathbf{k}-\mathbf{q}, t) \cdot (\mathcal{I} - f(\mathbf{k}, t)) - \right. \\
\left. N^+(\mathbf{q}, t) (\mathcal{I} - f(\mathbf{k}-\mathbf{q}, t)) \cdot B(\mathbf{k}, \mathbf{k}-\mathbf{q}, t) \cdot f(\mathbf{k}, t) + \text{h.c.} \right\}. \quad (3.19)
\end{aligned}$$

As it is, we obtained the closed form of kinetic equations for the description of the ultrafast dynamics in a laser excited semiconductor. The collision integrals as well as equation for generalized temperature are obtained in the closed form, being expressed through one-particle nonequilibrium distribution functions. Kinetic equations are known to posses two basic properties [11,12,14]:

- They have to allow equilibrium distribution function to be a solution for kinetic equations. In other words, the collision integrals are vanishing when all distribution functions simultaneously are taken to be zero.
- Collision integrals have to obey the energy balance requirement, as long as no additional relaxation (like quasiparticle damping or any phenomenological relaxation terms) is introduced in the kinetic equations.

In the next section we dwell our attention on balance equation, pointing at the fact that in the absence of quasiparticle damping and without any simplifications in kinetic kernels the collision integrals satisfy the second basic property. In the fifth Section we will show that equilibrium distribution functions are the solution to kinetic equations that agrees completely with the first basic requirement.

#### 4. Energy balance equation

As has been already said, the kinetic equations without quasiparticle damping should obey energy balance. To verify this requirement, let us multiply the first equation from (2.11) by single-particle energy matrix  $M(\mathbf{k}, t)$  and the second equation by  $\hbar\omega_{LO}$ . Using definition of time-ordered exponents (3.8)-(3.9), their symmetry properties (see Appendix A) and taking the trace over zone indexes, one can write down the following expression:

$$\frac{d}{dt}(\varepsilon_{kin}(t) + \varepsilon_{ph}(t) + \varepsilon_{int}(t)) = j_{pol}(t)E(t), \quad (4.1)$$

where polarization current is defined via the time derivative of the real part of polarization:

$$j_{pol}(t) = d_0 \frac{d}{dt} \sum_{\mathbf{k}} (f_{cv}(\mathbf{k}, t) + f_{vc}(\mathbf{k}, t)), \quad (4.2)$$

averaged kinetic energy of the carriers  $\varepsilon_{kin}(t)$  and phonons  $\varepsilon_{ph}(t)$  is being defined via the corresponding one-particle nonequilibrium distribution functions

$$\varepsilon_{kin}(t) = \sum_{\alpha\mathbf{k}} e_{\alpha\mathbf{k}} f_{\alpha\alpha}(\mathbf{k}, t), \quad \varepsilon_{ph}(t) = \sum_{\mathbf{q}} \hbar\omega_{LO} n(\mathbf{q}, t), \quad (4.3)$$

while for the mean value of interaction energy  $\varepsilon_{int}(t)$  one can present after some algebra the following explicit relation:

$$\varepsilon_{int}(t) = \frac{1}{i\hbar} \int_{t_0}^t dt' \sum_{\mathbf{q}} D(\mathbf{q})^2 \text{Tr} \left\{ \exp[-i\omega_{LO}(t-t')] \right. \\ \left. \times \exp_+ \left[ -\frac{i}{\hbar} \int_{t'}^t M(\mathbf{k}-\mathbf{q}, t'') dt'' \right] \right\}.$$

$$\{N^+(\mathbf{q}, t') (\mathcal{I} - f(\mathbf{k}-\mathbf{q}, t')) \cdot A(\mathbf{k}, \mathbf{k}-\mathbf{q}, t') \cdot f(\mathbf{k}, t') \\ - N^-(\mathbf{q}, t') f(\mathbf{k}-\mathbf{q}, t') \cdot A(\mathbf{k}, \mathbf{k}-\mathbf{q}, t') \cdot (\mathcal{I} - f(\mathbf{k}, t'))\}. \quad (4.4)$$

$$\exp_- \left[ \frac{i}{\hbar} \int_{t'}^t M(\mathbf{k}, t'') dt'' \right] - \text{h.c.} \Big\},$$

$$A(\mathbf{k}, \mathbf{k}-\mathbf{q}, t) = \mathcal{I} - \beta(t)B(\mathbf{k}, \mathbf{k}-\mathbf{q}, t).$$

We will show in the next section that generalized temperature  $T(t)$ , conjugated to mean interaction energy  $\varepsilon_{int}(t)$ , takes its constant value much earlier than distribution functions become quasiequilibrium. In Markovian limit  $\varepsilon_{int}(t) \rightarrow 0$ ,  $T(t) \rightarrow \text{const}$  while collision integrals remain finite. Basically, it agrees completely with two-time hierarchy spoken about in Introduction. On the other hand, one can introduce a characteristic time of correlation damping  $\tau_{mark}$ , which is smaller than time  $\tau_{asympt}$  at which one-particle distribution functions approach their equilibrium values. We will call  $\tau_{mark}$  “a time of Markovization” (see next Section for detailed explanations) keeping in mind that this is a time scale at which correlational components  $I_{\alpha\beta}^{corr}(\mathbf{k}, t)$ ,  $I_{ph}^{corr}(\mathbf{q}, t)$  of collision integrals vanish [12,13].

A laser pulse diminishes with time, and the right hand side of the Eqn.(4.1) tends to zero; thus we pass from energy balance equation to energy conservation [12,13]. At long times, when carrier distribution functions tend to quasi Fermi distribution and phonon distribution function tends to quasi Bose distribution, it is possible to introduce two-temperature formalism [19]. In cited paper the authors showed the two-temperature model to become valid at times of thermalization, when the kinetic energy starts to evolve in a quasi Fermian manner.<sup>4</sup> Contrary, there are some doubts about validity of two-temperature model at short times, though it does not prevent us from introduction of generalized electron temperature  $T_{el}(t)$  and lattice temperature  $T_{ph}(t)$  in a way similar to (3.17). Namely, at the times when laser pulse has ceased one can rewrite a balance equation (4.1) as follows:

$$c_{el}(t) \frac{dT_{el}(t)}{dt} + c_{ph}(t) \frac{dT_{ph}(t)}{dt} = -c_{corr}(t) \frac{dT(t)}{dt}, \quad (4.5)$$

<sup>4</sup>In order to the reader does not confuse  $\tau_{therm}$  with time of the establishing of quasiequilibrium the authors explain:  $\tau_{therm}$  gives a time when an electron gas exchanges energy with a cold lattice at the same rate as a Fermi-distributed electron gas, which has the same internal energy. Hence,  $\tau_{therm}$  “is not a time, after which Fermi distribution is established but the time at which the laser excited electron gas behaves as a Fermi distributed electron gas.” In any case, since the authors of Ref. [19] have already chosen a Markovian form of the kinetic equations, there is not any reason to relate thermalization time to  $\tau_{mark}$ : obviously,  $\tau_{mark} \ll \tau_{therm} < \tau_{asympt}$ .

where we have denoted  $c_{corr}(t) = C(t)/T(t)^2$  for correlational contribution to the heat capacity while one can write down the following relations for the electron and lattice generalized heat capacities:

$$c_{el}(t) = \frac{(\hat{\varepsilon}_{kin}, \hat{\varepsilon}_{kin})_t}{T_{el}(t)^2}, \quad (\hat{\varepsilon}_{kin}, \hat{\varepsilon}_{kin})_t = \sum_{\alpha \mathbf{k}} e_{\alpha \mathbf{k}}^2 f_{\alpha}(\mathbf{k}, t)(1 - f_{\alpha}(\mathbf{k}, t)), \quad (4.6)$$

$$c_{ph}(t) = \frac{(\hat{\varepsilon}_{ph}, \hat{\varepsilon}_{ph})_t}{T_{ph}(t)^2}, \quad (\hat{\varepsilon}_{ph}, \hat{\varepsilon}_{ph})_t = \sum_{\mathbf{q}} \hbar^2 \omega_{LO}^2 n(\mathbf{q}, t)(1 + n(\mathbf{q}, t)). \quad (4.7)$$

Right hand side of (4.5), which is related to energy loss due to correlation, tends to zero in Markovian limit, and we have the following picture: an electron gas is cooling to  $T(t \rightarrow \infty) = T_{eq}$  from above, a lattice is heating to the same equilibrium temperature from below. As to generalized nonequilibrium temperature  $T(t)$  associated with  $\varepsilon_{corr}$ , it forms equilibrium background at long times. All these statements to be confirmed at the Section 6, when we estimate the evolution of generalized temperatures of an effective one-zone semiconductor.

If one makes any approximation for kinetic kernels (for instance, expansion in series in external field amplitude), balance equation (4.1) will be broken automatically. In such a case, one can only talk about balance equation up to a certain order in  $E_0$ . Since we do not have an exact expression for kinetic kernels anyway, in subsequent numerical evaluations we will operate with diagonal matrix only, which corresponds to Wigner–Weisskopf approximation to Green’s functions.

The last remark which has to be made in this section is related to quasiparticle damping and its influence on energy balance equation. As has been already said, quasiparticle damping arises from going beyond the framework of the 2-nd Born approximation. In a general case, writing down the kinetic equations one has to compose the dynamic equation for time–energy dependent quasiparticle damping function  $\gamma(\mathbf{k}, t, t')$  in addition to modified kinetic kernels

$$\exp_+ \left[ -\frac{i}{\hbar} \int_{t'}^t M(\mathbf{k}, t'') dt'' \right]_{\alpha\beta} \longrightarrow \exp_+ \left[ -\frac{i}{\hbar} \int_{t'}^t M(\mathbf{k}, t'') dt'' \right]_{\alpha\beta} \exp[-\gamma(\mathbf{k}, t, t')].$$

However, once we have introduced nonzero  $\gamma(\mathbf{k}, t, t')$ , we cannot avoid violation of energy balance. We shall not talk about this problem any

more, referring reader to the literature [10,14,18]. The only reason to introduce nonzero quasiparticle damping is dealt with necessity to stabilize numerical solutions to kinetic equations at long time scales. However, as will be shown in the last section, the “fee” for such a procedure is overheating of the system: the more pronounced, the larger value of  $\gamma$  taken into consideration.

## 5. An effective one-zone semiconductor. Transition to Markovian limit.

In previous sections we obtained kinetic equations in the 2-nd Born approximation taking into account running dynamic correlation with the aid of  $\langle H_{int} \rangle_t$ . Correlational component of the collision integral involves the generalized temperature, for which we can obtain the equation (3.17). All kinetic equations are of the closed form but rather complicated matrix structure. Moreover, kinetic kernels in the general case could be evaluated only numerically. Therefore we consider the problem of relaxation towards equilibrium. To get rid of the complicated matrix structure in the expressions for collision integrals we made two approximations:

- (i) we neglect off-diagonal elements in kinetic kernels;
- (ii) polarization is supposed to vanish more rapidly than electron, hole and phonon distribution functions.

Formally, it means that we neglect off-diagonal elements of Lagrange multipliers (B.8) which, in their turn, tend to zero in zero-field limit. In our viewpoint, such an assumption is, at least, more grounded than approximation of phonon bath made in Ref. [10], when phonon collision integral is considered to be zero, while carriers distribution functions as well as polarization are essentially nonequilibrium. In our approximation, the two-level problem decouples into effective one-zone problems. We have the particles in conduction zone, the holes in valence zone after laser pulse irradiations, electrons and holes interact with nonequilibrium phonon subsystem but c-v transitions are vanishing.

With all mentioned assumptions we are in a position to rewrite the kinetic equations and the equation for quasitemperature in the following form:

$$I_{\alpha\alpha}(\mathbf{k}, t) = -\frac{2}{\hbar^2} \sum_{\mathbf{q}} D(\mathbf{q})^2 \int_{t_0}^t dt' \cos[\Omega_{\mathbf{k}, \mathbf{k}-\mathbf{q}, \mathbf{q}}^{\alpha\alpha}(t-t')] \times$$

$$\begin{aligned}
& \times \left( 1 - \beta(t') \frac{\hbar \Omega_{\mathbf{k}, \mathbf{k}-\mathbf{q}, \mathbf{q}}^{\alpha\alpha}}{\ln \mathcal{K}_{\mathbf{k}, \mathbf{k}-\mathbf{q}, \mathbf{q}}^{\alpha\alpha}(\{f(t'), n(t')\})} \right) \\
& \times \left( f_{\alpha\alpha}(\mathbf{k})(1 - f_{\alpha\alpha}(\mathbf{k} - \mathbf{q}))(1 + n(\mathbf{q})) - (1 - f_{\alpha\alpha}(\mathbf{k}))f_{\alpha\alpha}(\mathbf{k} - \mathbf{q})n(\mathbf{q}) \right)_{t'} \\
& - \frac{2}{\hbar^2} \sum_{\mathbf{q}} D(\mathbf{q})^2 \int_{t_0}^t dt' \cos [\Omega_{\mathbf{k}-\mathbf{q}, \mathbf{k}, \mathbf{q}}^{\alpha\alpha}(t - t')] \times \\
& \left( 1 - \beta(t') \frac{\hbar \Omega_{\mathbf{k}-\mathbf{q}, \mathbf{k}, \mathbf{q}}^{\alpha\alpha}}{\ln \mathcal{K}_{\mathbf{k}-\mathbf{q}, \mathbf{k}, \mathbf{q}}^{\alpha\alpha}(\{f(t'), n(t')\})} \right) \times \\
& \left( f_{\alpha\alpha}(\mathbf{k})(1 - f_{\alpha\alpha}(\mathbf{k} - \mathbf{q}))n(\mathbf{q}) - (1 - f_{\alpha\alpha}(\mathbf{k}))f_{\alpha\alpha}(\mathbf{k} - \mathbf{q})(1 + n(\mathbf{q})) \right)_{t'}, \quad (5.1)
\end{aligned}$$

$$\begin{aligned}
I_{ph}(\mathbf{q}, t) &= \frac{2D(\mathbf{q})^2}{\hbar^2} \sum_{\alpha\mathbf{k}} \int_{t_0}^t dt' \cos [\Omega_{\mathbf{k}, \mathbf{k}-\mathbf{q}, \mathbf{q}}^{\alpha\alpha}(t - t')] \\
& \times \left( 1 - \beta(t') \frac{\hbar \Omega_{\mathbf{k}, \mathbf{k}-\mathbf{q}, \mathbf{q}}^{\alpha\alpha}}{\ln \mathcal{K}_{\mathbf{k}, \mathbf{k}-\mathbf{q}, \mathbf{q}}^{\alpha\alpha}(\{f(t'), n(t')\})} \right) \times \\
& \left( f_{\alpha\alpha}(\mathbf{k})(1 - f_{\alpha\alpha}(\mathbf{k} - \mathbf{q}))(1 + n(\mathbf{q})) - (1 - f_{\alpha\alpha}(\mathbf{k}))f_{\alpha\alpha}(\mathbf{k} - \mathbf{q})n(\mathbf{q}) \right)_{t'}, \quad (5.2)
\end{aligned}$$

where we have introduced the designations:

$$\Omega_{\mathbf{k}_1, \mathbf{k}_2, \mathbf{q}}^{\alpha\alpha} = \frac{1}{\hbar} (e_{\alpha\mathbf{k}_1} - e_{\alpha\mathbf{k}_2} - \hbar\omega_{\mathbf{q}}), \quad (5.3)$$

$$\mathcal{K}_{\mathbf{k}_1, \mathbf{k}_2, \mathbf{q}}^{\alpha\alpha}(\{f, n\}) = \frac{(1 - f_{\alpha\alpha}(\mathbf{k}_1, t))f_{\alpha\alpha}(\mathbf{k}_2, t)n(\mathbf{q}, t)}{f_{\alpha\alpha}(\mathbf{k}_1, t)(1 - f_{\alpha\alpha}(\mathbf{k}_2, t))(1 + n(\mathbf{q}, t))}. \quad (5.4)$$

The system of kinetic equations has to be complemented by the equation for quasitemperature, which has the following structure after all simplifications mentioned above:

$$\frac{d\beta(t)}{dt} = -\frac{2}{\hbar C(t)} \sum_{\alpha\mathbf{k}\mathbf{q}} D(\mathbf{q})^2 \Omega_{\mathbf{k}, \mathbf{k}-\mathbf{q}, \mathbf{q}}^{\alpha\alpha} \int_{t_0}^t dt' \cos [\Omega_{\mathbf{k}, \mathbf{k}-\mathbf{q}, \mathbf{q}}^{\alpha\alpha}(t - t')] \times$$

$$\begin{aligned}
& \times \left( 1 - \beta(t') \frac{\hbar \Omega_{\mathbf{k}, \mathbf{k}-\mathbf{q}, \mathbf{q}}^{\alpha\alpha}}{\ln \mathcal{K}_{\mathbf{k}, \mathbf{k}-\mathbf{q}, \mathbf{q}}^{\alpha\alpha}(\{f(t'), n(t')\})} \right) \times \\
& \left( f_{\alpha\alpha}(\mathbf{k})(1 - f_{\alpha\alpha}(\mathbf{k} - \mathbf{q}))(1 + n(\mathbf{q})) - (1 - f_{\alpha\alpha}(\mathbf{k}))f_{\alpha\alpha}(\mathbf{k} - \mathbf{q})n(\mathbf{q}) \right)_{t'}, \quad (5.5)
\end{aligned}$$

with the expression for generalized heat capacity

$$\begin{aligned}
C(t) &= (\hat{H}_{int}, \hat{H}_{int})_t = 2 \sum_{\alpha\mathbf{k}\mathbf{q}} D(\mathbf{q})^2 \\
& \times \frac{n(\mathbf{q})(1 - f_{\alpha\alpha}(\mathbf{k}))f_{\alpha\alpha}(\mathbf{k} - \mathbf{q}) - (1 + n(\mathbf{q}))f_{\alpha\alpha}(\mathbf{k})(1 - f_{\alpha\alpha}(\mathbf{k} - \mathbf{q}))}{\ln \left[ \frac{n(\mathbf{q})(1 - f_{\alpha\alpha}(\mathbf{k}))f_{\alpha\alpha}(\mathbf{k} - \mathbf{q})}{(1 + n(\mathbf{q}, t))f_{\alpha\alpha}(\mathbf{k})(1 - f_{\alpha\alpha}(\mathbf{k} - \mathbf{q}))} \right]}. \quad (5.6)
\end{aligned}$$

Since  $\{(x - 1)/\ln x\} > 0$ , we see from the last equation that  $C(t) > 0$ . From the expressions for kinetic equations and the equation for generalized temperature one can easily verify that equilibrium (quasiequilibrium) solutions satisfy Eqns.(5.1),(5.2), (5.5). Indeed, let us write down quasiequilibrium distribution functions for electron and phonon subsystems:

$$\begin{aligned}
f_{\alpha\alpha}^0(\mathbf{k}, t) &= \frac{1}{\exp\{\beta(t)(e_{\alpha\mathbf{k}} - \mu_{\alpha}(t))\} + 1}, \\
n^0(t) &= \frac{1}{\exp\{\beta(t)\hbar\omega_{LO}\} - 1}, \quad (5.7)
\end{aligned}$$

where  $\mu_{\alpha}(t)$  means nonequilibrium chemical potential of the electrons/holes. It can easily be checked that

$$\beta(t)\hbar\Omega_{\mathbf{k}_1, \mathbf{k}_2, \mathbf{q}}^{\alpha\alpha} = \mathcal{K}_{\mathbf{k}_1, \mathbf{k}_2, \mathbf{q}}^{\alpha\alpha}(\{f^0(t), n^0(t)\}), \quad (5.8)$$

where the  $\mathcal{K}$ -function in the right-hand side is obtained from the function (5.4) by replacing nonequilibrium distribution functions with their quasiequilibrium correspondences. Using equation (5.7), we see that the collision integrals (5.1) and (5.2) are equal to zero if  $f_{\alpha}(\mathbf{k}, t) = f_{\alpha}^0(\mathbf{k}, t)$  and  $n(\mathbf{q}, t) = n^0(t)$ . Note that in this case the right-hand side of equation (5.5) is also zero. Contrary, the collision integrals without their correlational components do not have an equilibrium solution!

Let us consider in more details approaching of the system to the Markovian limit. The Markovian approximation implies that the collision integrals (5.1), (5.2), and the right-hand side of equation (5.5) depend on the distribution functions and the quasi-temperature taken at time  $t$ .

The Markovian approximation is inadequate for the initial stage of evolution because of initial correlations and, strictly speaking, describes the limiting long-time regime where  $\partial f_{\alpha\alpha}(\mathbf{k}, t)/\partial t \rightarrow 0$ ,  $\partial n(\mathbf{q}, t)/\partial t \rightarrow 0$ , and  $\partial\beta(t)/\partial t \rightarrow 0$ . Thus, on passage to the Markovian limit in the collision integrals (5.1), (5.2), and in equation (5.5) for the quasi-temperature, we put  $f_{\alpha\alpha}(\mathbf{k}, t') \approx f_{\alpha\alpha}(\mathbf{k}, t)$ ,  $n(\mathbf{q}, t') \approx n(\mathbf{q}, t)$ , and then pass to the limit  $t - t_0 \rightarrow \infty$ . The remainder integrals over  $t'$  are calculated in the standard way:

$$\lim_{t-t_0 \rightarrow \infty} \int_{t_0}^t dt' \cos[\Omega(t-t')] = \lim_{\varepsilon \rightarrow +0} \int_{-\infty}^0 d\tau e^{\varepsilon\tau} \cos(\Omega\tau) = \pi\delta(\Omega). \quad (5.9)$$

From the last equation one can see immediately that in Markovian limit correlational components of the collision integrals (terms including  $\beta(t)$ ) as well as right hand side of (5.5) become zeros. Indeed, in all these expressions we face with a construction like  $\Omega\delta(\Omega) \equiv 0$ . Nevertheless, collisional components of  $I_{el}$ ,  $I_{ph}$  remain finite and have the form of Uehling–Uhlenbeck collision integrals [7]. Equilibrium distribution functions are known to obey kinetic equations with collisions Uehling–Uhlenbeck type integrals.

Let us to sum up all results from this section. We have shown that only taking into consideration a correlational component of collision integrals one can guarantee the kinetic equations to have an equilibrium solution. In Markovian limit  $I^{(corr)}$  tends to zero, but it is a correlational component, which allows system to approach this limit. When behaviour of the system became Markovian, generalized temperature stop to change, but in no way it means that all collisions ceased. From this time one is able to introduce a two-temperature model of the system and can observe how distribution functions approach their (quasi)equilibrium values.<sup>5</sup>

## 6. Discussion

In numerical calculations we investigated both dynamics of two-zone semiconductor during laser pulse irradiation and its relaxation to-

<sup>5</sup>During numerical studies we can talk only about approaching to quasiequilibrium value as long as we are not able to obtain the exact relation (5.9) numerically, integrating the cosine function. As will be shown in the next section, introduction of quasiparticle damping in the simplest form  $\exp[-\gamma(t-t')]$  can stabilize the long time solutions and guarantee long time asymptotics to exist. However, the problem of violation of the energy balance appears.

ward equilibrium with additional assumption that polarization relaxes much faster than carriers and phonon distribution functions. Both thermal phonons approximation (phonon bath) and complete dynamics of phonons have been taken into consideration. As for a complete dynamics during laser pulse, we verified the results of the previous papers [3,10,16]. We considered  $\gamma$  as a fitting parameter, stabilizing the solutions of quantum kinetic equations at long time scales.

We chose the following typical energies and time scales: the energy of longitudinal optical phonon equals  $\hbar\omega_{LO}=36\text{mEv}$ , Rabi energy  $\hbar\omega_R=26.3\text{mEv}$ , laser pulse duration  $\tau_p=50\text{fs}$ , the width of band gap was equal  $E_g=1.8\text{eV}$ , excess energy  $\Delta = \hbar\omega - E_g=60\text{mEv}$ . Dimensionless constant of electron-phonon interaction equals 0.069; mass ratio of holes to electrons equals 6.86 and the initial temperature of the phonon bath was 164K. We use values  $\hbar\omega_{LO}$  and  $\tau_p$  as characteristic energy and time scales for obtaining of dimensionless units.

To solve the system of kinetic equations we made use of factorization of the kinetic kernels and borrowed from the authors of Ref. [10] the idea how to pass from the initial integro-differential equations to the extended set of ordinary differential equations. Details of the transition from integro-differential equations to the extended system of ordinary differential equations are presented in Appendix C. This extended set was solved by applying the fifth order Runge–Kutta integration with automatic step-size control. We chose dense enough energy grid in  $N = 60$  points that corresponds to  $\text{dim} = 5N + 12N^3 + 1 \sim 2.5 \times 10^6$  ordinary differential equations.

In figure (6.1) we presented behaviour of the electron distribution function at fixed times depending on dimensionless energy. The phonon subsystem was chosen to be equilibrium. Complete dynamics of the system (polarization was governed by its own kinetic equation) was allowed, but we neglected the correlational part of collision integral and put  $\gamma = 0$ . One can see as initial excitation on the excess energy  $\Delta/\hbar\omega_{LO} = 5/3$  starts to relax at long times, but does not reach its equilibrium value. Quite contrary, the lower panel shows unphysical behaviour at large times  $t > 20\tau_p$ . We have to mention that similar behaviour, though less pronounced, was observed even with nonequilibrium phonons.

As soon as we introduced nonzero quasiparticle damping, the behaviour of the system becomes regular. In figure (6.2) one can see that even small enough  $\gamma$  clears away all unphysical effects at long times ensuring relaxation toward quasiequilibrium. Running a few steps forward we note that this long-time asymptotics is not a true equilibrium because nonzero  $\gamma$  does not guarantee energy balance.

Starting from figure (6.3) we will talk about the relaxation problem. Namely, we start from an initially perturbed system with assumption that polarization is negligible at time  $t \geq 4\tau_p$ , when the external field tends to zero. Figure 3 demonstrates that consideration of nonequilibrium phonon distribution function  $n_{ph}(\varepsilon_q, t)$  (and even quasiequilibrium one) prevents from unphysical behaviour, though does not ensure relaxing to equilibrium. It becomes quite apparent in figure (6.4a), where time dependence of total phonon number  $N(t) = \sum_{\mathbf{q}} n_{ph}(\mathbf{q}, t)$  is presented. We see that  $N(t)$  does not saturate unless additional relaxation mechanism is taken into consideration. This relaxation consists in adding to  $I_{ph}(\mathbf{q}, t)$  a simple damping term  $(n_{ph}^{eq} - n_{ph}(\mathbf{q}, t))/\mathcal{T}_{ph}$  in so-called  $\tau$  approximation with finite phonon relaxation time  $\mathcal{T}_{ph}=350$ fs. An additional relaxation term ensures relaxation of total phonon number to its asymptotic value depending on  $\mathcal{T}_{ph}$ . As one can see from figure (6.4b), taking into consideration the correlational part of collision integral results in smooth relaxation (solid line), while disregard of running correlations leads to non-motivated flexure of dashed line at long times.

In figure (6.5) we plotted the energy dependence of nonequilibrium phonon distribution function  $n_{ph}(\varepsilon, t)$  at fixed times with finite phonon relaxation  $\mathcal{T}_{ph}=350$ fs and zero quasiparticle damping. One can observe relaxation from initial distribution with maximum at zero energy to dispersionless asymptotic value at long times. A remarkable feature should be noted: this asymptotic value differs from quasiequilibrium phonon distribution  $n_{ph}(\varepsilon, t)|_{t \gg \tau_p} = n_{ph}(t) = 1/[\exp(\hbar\omega_{LO}/T(t)) - 1]$  (marked by horizontal dashed line) by less than 8%! In our point of view, this small difference is induced by relatively small value of additional relaxation term in phonon collision integral, which, in its turn, is found to be indispensable for relaxation to equilibrium. In fact, here we face with dilemma: either to neglect any relaxation mechanism put “by hand” or not to reach equilibrium asymptotics on long-time scales. Situation could be partially improved if one composes dynamic equation for quasiparticle damping function  $\gamma(\varepsilon, t, t')$ , that is, considers damping self-consistently [14] unlike in our case of fixed  $\gamma$ . However, even then one cannot avoid energy disbalance. This is not surprising, because the extra terms in the energy balance equation are formally beyond the 2-nd Born approximation used in the derivation of the kinetic equation. In Green’s functions method, to obtain the collision integrals which involve the quasiparticle damping and are consistent with the energy conservation, one has to calculate the  $T$ -matrices in the full collision integrals, keeping terms of higher-order in the interactions. In NSO method, presented here, the problem of consideration of higher order interactions seems to be even

more difficult [12].

In any case, figure (6.5) is the best possible illustration of the fact that Lagrange multiplier  $\beta(t)$  in relevant statistical operator (3.1) really means inverse generalized temperature, defining quasiequilibrium phonon distribution.<sup>6</sup>

The importance of running correlations could be traced from figures (6.6), (6.7) and (6.11). In figures (6.6)-(6.7) we presented the energy dependence of  $f_c(\varepsilon, t)$  at fixed times and  $\gamma = 0$ . There is no complete relaxation toward equilibrium in both cases, but neglecting dynamic correlations leads to pronounced unphysical behaviour of electron distribution functions at small energies (Fig.6), while taking of  $I^{(corr)}$  into account improves situation greatly (Fig.7). A similar remark concerns the energy balance in the case of one-zone “semiconductor” disturbed initially from equilibrium (figure (6.11))<sup>7</sup>. One can see that neglecting of initial correlations leads immediately to a pronounced energy disbalance, while in the case of nonzero  $I^{(corr)}$  we keep the constant value  $\varepsilon_{kin}(t) + \varepsilon_{ph}(t) + \varepsilon_{int}(t)$  within 3-4% accuracy. (The greater accuracy is not reachable because of non-zero quasiparticle damping which leads to a deviation from energy balance.)

Figure 8, which describes energy disbalance, is no less interesting. One can see that taking into account quasiparticle damping (still relatively small!) leads to “overheating” of the system: generalized temperature  $T(t)$  does not reach its asymptotic value whereas zero damping and smaller phonon relaxation term assure transition to quasiequilibrium regime. Another interesting point is that the period of temperature oscillation is twice shorter than inverse phonon frequency  $2\pi/\omega_{LO}$ . The other dynamic characteristics (generalized heat capacity, total phonon number, phonon distribution function) oscillate with phonon period  $\mathcal{T}_{ph} = 2\pi/\omega_{LO}$ . At present we do not have an answer why energy exchange between two subsystems gives doubled frequency of temperature oscillation.

Figure 9 is a logical consequence of previous plots. One can see that long-time asymptotic of phonon distribution function in case of nonzero  $\gamma$  (circles) differs much greater from its quasiequilibrium value than with no quasiparticle damping (squares). When  $\gamma \neq 0$  the system tends

<sup>6</sup>More precisely, we have to talk about a generalized phonon temperature  $T_{ph}(t)$ , introduced in the section 4. However, the analysis of the behaviour of the electron and phonon generalized temperatures (see figure (6.10)) shows them to be equal to  $T(t) = \beta(t)^{-1}$  at long time-scales.

<sup>7</sup>In fact, we reduce the basic formulae of the previous section to the case of one-zone model. That is why we took a word “semiconductor” in inverted commas, talking actually about a one zone metal.

to its asymptotic characteristic but this state is not equilibrium one: collision integral does not conserve the energy and semiconductor remains overheated.

The results for two-temperature model are presented on the figure (6.10). One can see that initially heated electron subsystem is cooling down, lattice is heating up, while generalized temperature  $T(t)$ , conjugated to mean interaction energy, forms an equilibrium background. A maximum of  $T(t)$  is located at a point, which corresponds to the largest value of  $\langle \hat{H}_{int} \rangle_t$ .

It is interesting also to investigate relaxation of the system after non-resonant excitation when  $\Delta = \hbar\omega - E_g < 0$ . Numerical calculations from Ref. [10] manifested phonon replicas in conducting zone on long-time scales due to indirect excitation of the electrons. And, to be able to describe a complete dynamics of the system during laser pulse as well as at long times, one has to consider a non-simplified version of total collision integral with off-diagonal  $\Lambda$  in (3.14)-(3.15) and non-simplified matrix exponents  $\exp_+ \left[ -\frac{i}{\hbar} \int_{t'}^t M(\mathbf{k}, t'') dt'' \right]_{\alpha\beta}$ . This problem, though difficult enough, is very promising and the results could give an answer, how running correlations form quite complicated dynamics of two-zone semiconductor under short-lasting laser excitation.

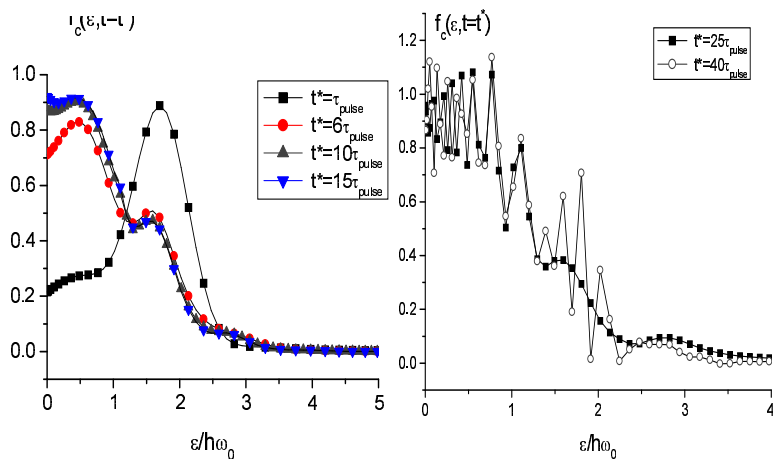


Figure 6.1. Electron distribution function at fixed times. Thermal phonons, no quasiparticle damping.

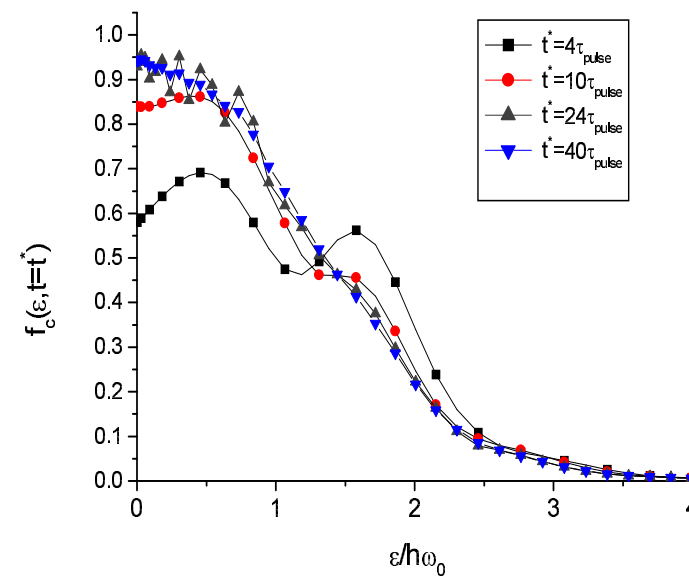


Figure 6.2. Electron distribution function at fixed times. Thermal phonons,  $\gamma = 1/36$ .

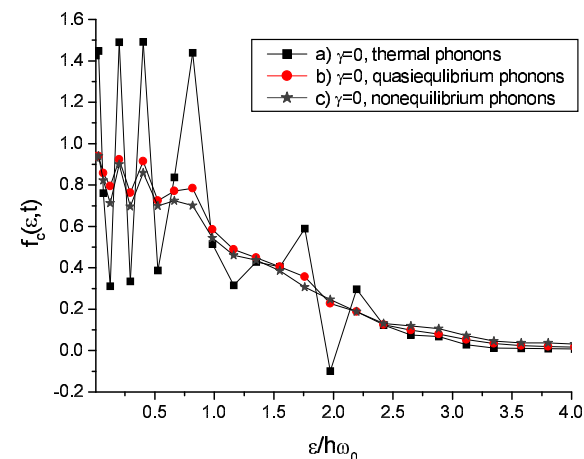


Figure 6.3. Electron distribution function  $f_c(\epsilon, t)$  at  $t = 20\tau_{pulse}$ ,  $\gamma = 0$ . Dynamical correlations are taken into account in b) and c).

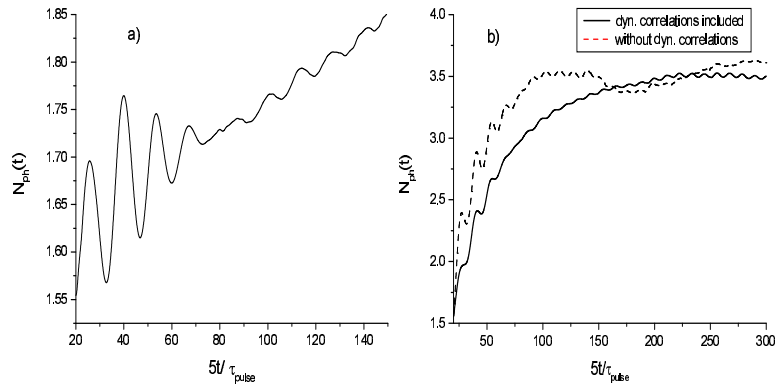


Figure 6.4. Time dependence of total phonon number  $N_{ph}(t)$ : a)  $\gamma = 0$ ,  $T_{ph} \rightarrow \infty$ ; b)  $\gamma = 0$ ,  $T_{ph} = 7\tau_{pulse}$ .

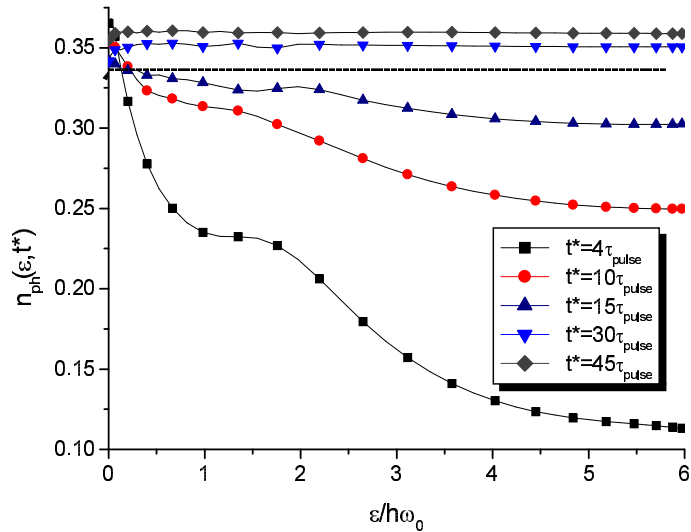


Figure 6.5. Phonon distribution functions  $n_{ph}(\epsilon, t)$  at fixed times. Horizontal dashed line marks equiasiequilibrium value.

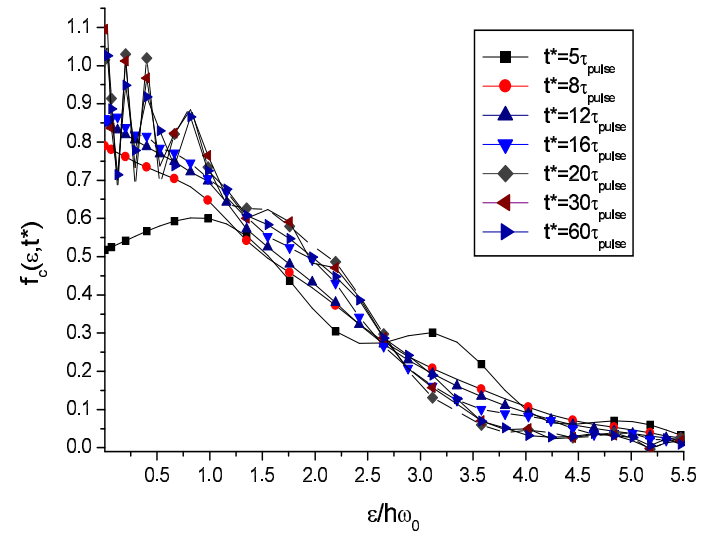


Figure 6.6. Electron distribution function  $f_c(\epsilon, t)$  at fixed times. No initial correlations.

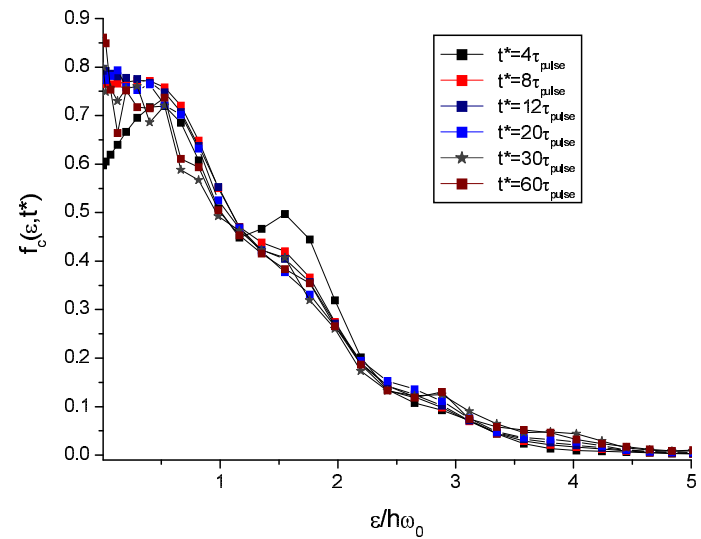


Figure 6.7. Electron distribution function  $f_c(\epsilon, t)$  at fixed times. Initial correlations are taken into account.



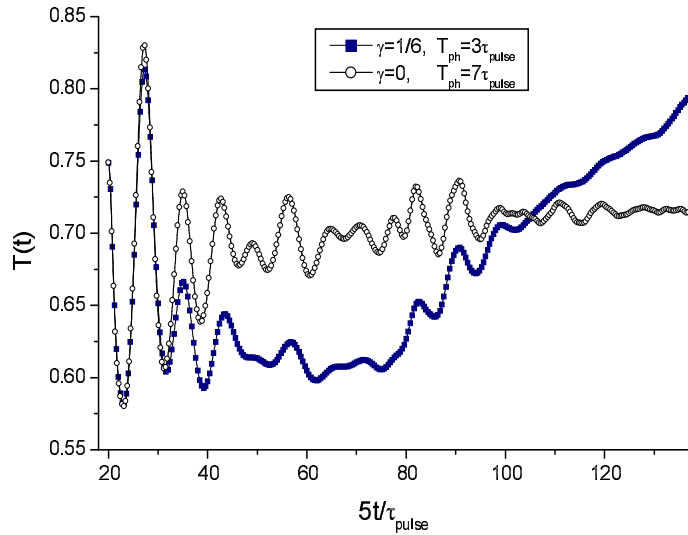


Figure 6.8. Generalized temperature  $T(t)$  in dimensionless units.

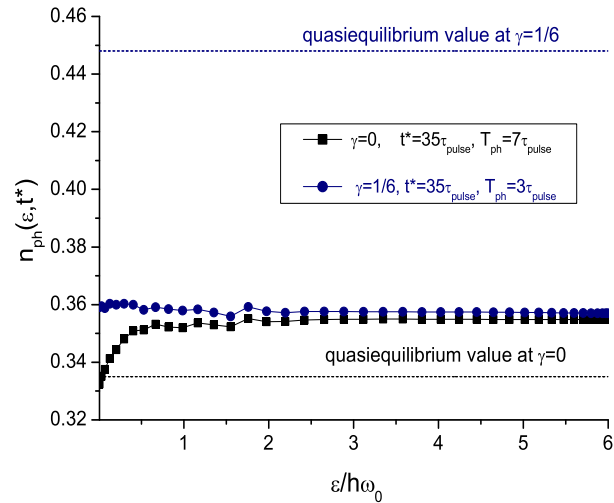


Figure 6.9. Phonon distribution functions  $n_{ph}(\varepsilon, t)$  at fixed times. Horizontal dashed lines mark quasiequilibrium values.

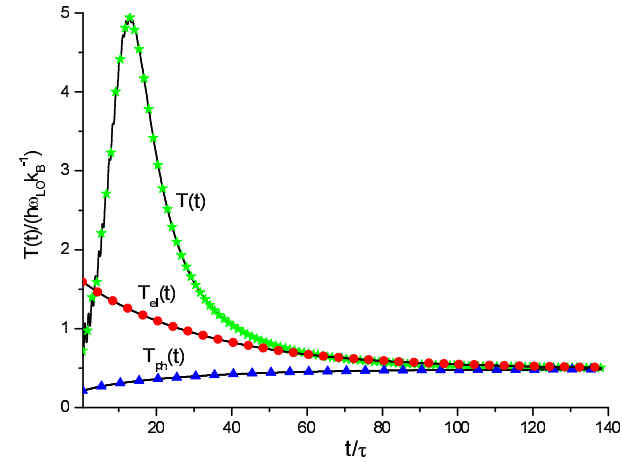


Figure 6.10. Generalized temperatures as functions of dimensionless time:  $T(t)$  - green stars,  $T_{el}(t)$  - red circles,  $T_{ph}(t)$  - blue triangles. Initial correlations are taken into account,  $\gamma = 0.1$

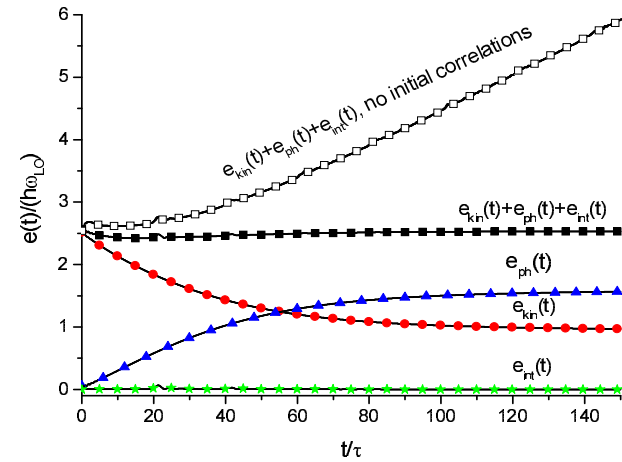


Figure 6.11. Mean energies as functions of dimensionless time:  $\varepsilon_{int}(t)$  - green stars,  $\varepsilon_{kin}(t)$  - red circles,  $\varepsilon_{ph}(t)$  - blue triangles. Total energy  $\varepsilon_{kin}(t) + \varepsilon_{ph}(t) + \varepsilon_{int}(t)$  is marked by solid squares (when initial correlations are taken into consideration) and by opened squares (with no initial correlations).  $\gamma = 0.1$ .

## Appendix A

To obtain the solution for time-ordered matrix exponents (3.8)-(3.9) one has to consider the system of differential equations (3.10). In general case of time-dependent field the system can not be solved analytically. To find the explicit expressions for kinetic kernels one has to neglect the field-dependent terms in the equations of diagonal  $A(\mathbf{k}, t)$ . Then one can obtain immediately unperturbed Wigner-Weisskopf form for diagonal matrix elements:

$$\exp_+ \left[ -\frac{i}{\hbar} \int_{t'}^t M(\mathbf{k}, t'') dt'' \right]_{cc} \approx \exp \left( -\frac{i}{\hbar} e_{c\mathbf{k}}(t-t') \right) + o(E^2), \quad (\text{A.1})$$

$$\exp_+ \left[ -\frac{i}{\hbar} \int_{t'}^t M(\mathbf{k}, t'') dt'' \right]_{vv} \approx \exp \left( -\frac{i}{\hbar} e_{v\mathbf{k}}(t-t') \right) + o(E^2) \quad (\text{A.2})$$

with similar expressions for anti-chronologically ordered exponents:

$$\exp_- \left[ \frac{i}{\hbar} \int_{t'}^t M(\mathbf{k}, t'') dt'' \right]_{cc} \approx \exp \left( \frac{i}{\hbar} e_{c\mathbf{k}}(t-t') \right) + o(E^2), \quad (\text{A.3})$$

$$\exp_- \left[ \frac{i}{\hbar} \int_{t'}^t M(\mathbf{k}, t'') dt'' \right]_{vv} \approx \exp \left( \frac{i}{\hbar} e_{v\mathbf{k}}(t-t') \right) + o(E^2). \quad (\text{A.4})$$

After substitution of the diagonal elements into the equations (3.10) for off-diagonal ones and supposing the field amplitude to change much slower than cosine factor in (2.6), we come to the following form of field-dependent kinetic kernels:

$$\exp_+ \left[ -\frac{i}{\hbar} \int_{t'}^t M(\mathbf{k}, t'') dt'' \right]_{cv} = \exp_- \left[ \frac{i}{\hbar} \int_t^{t'} M(\mathbf{k}, t'') dt'' \right]_{vc} \approx (\text{A.5})$$

$$\approx \frac{1}{2} d_0 E_0 \left[ \exp \left( \frac{i}{\hbar} \delta_{\mathbf{k}}^- t \right) - \exp \left( \frac{i}{\hbar} \delta_{\mathbf{k}}^- t' \right) \right] \exp \left( \frac{i}{\hbar} e_{v\mathbf{k}} t' - \frac{i}{\hbar} e_{c\mathbf{k}} t \right) + o(E^3),$$

where  $\delta_{\mathbf{k}}^- = e_{c\mathbf{k}} - e_{v\mathbf{k}} - \hbar\omega$  denotes detuning of the system <sup>8</sup>

<sup>8</sup>In rotating wave-approximation for external field [1,10] we neglect non resonant strongly oscillating term with  $\delta_{\mathbf{k}}^+ = e_{c\mathbf{k}} - e_{v\mathbf{k}} + \hbar\omega$  in favour of resonant one  $\delta_{\mathbf{k}}^-$ .

Let us make some conclusions from the expressions (A.1)-(A.5). On the first hand, the kinetic kernels possess the symmetric properties similar to those of retarded and advanced Green's functions. On the second hand, one can verify all expressions for matrix kernel to factorize in  $t, t'$  (see Eqn.(3.11) for comparison). The last conclusion follows from the expression for off-diagonal element (A.5): this kernel turns out to be relevant in a narrow zone around excitation energy and could be omitted when one describes low and high energy domains of distribution functions.

## Appendix B

Let us rewrite the last commutator in (3.5) via Kubo identity

$$[\rho_{rel}(t'), H_0(t')] = \beta(t') \int_0^1 \tilde{\rho}_{rel}^x(t') [H_0(t'), H_{int}] \tilde{\rho}_{rel}^{-x}(t'), \quad (\text{B.1})$$

where the symbol “tilde” denotes truncated statistical operator

$$\tilde{\rho}_{rel}(t) = \frac{1}{\tilde{Z}_{rel}(t)} \exp \left\{ - \sum_{\alpha\beta, \mathbf{k}\mathbf{q}} \Lambda_{\alpha\beta}(\mathbf{k}, t) \hat{f}_{\alpha\beta}(\mathbf{k}) - \Lambda^{ph}(\mathbf{q}, t) \hat{n}(\mathbf{q}) \right\} \quad (\text{B.2})$$

without interaction part  $H_{int}$  [12] (remember, we have to work in the framework of the 2-nd Born approximation). The action of operators  $\tilde{\rho}_{rel}^x(t'), \tilde{\rho}_{rel}^{-x}(t')$  can be presented in the similar matrix form like (3.8), namely:

$$\begin{aligned} \tilde{\rho}_{rel}^x(t') a_{\mathbf{k}} \tilde{\rho}_{rel}^{-x}(t') &= \exp [x\Lambda(\mathbf{k}, t')] a_{\mathbf{k}}, \\ \tilde{\rho}_{rel}^x(t') a_{\mathbf{k}}^\dagger \tilde{\rho}_{rel}^{-x}(t') &= \exp [-x\Lambda(\mathbf{k}, t')] a_{\mathbf{k}}^\dagger, \\ \tilde{\rho}_{rel}^x(t') b_{\mathbf{q}} \tilde{\rho}_{rel}^{-x}(t') &= \exp [x\Lambda^{ph}(\mathbf{q}, t')] b_{\mathbf{q}}, \\ \tilde{\rho}_{rel}^x(t') b_{\mathbf{q}}^\dagger \tilde{\rho}_{rel}^{-x}(t') &= \exp [-x\Lambda^{ph}(\mathbf{q}, t')] b_{\mathbf{q}}^\dagger, \end{aligned} \quad (\text{B.3})$$

with the only exception that  $\exp [x\Lambda(\mathbf{k}, t')]_{\alpha\beta}$  can be evaluated explicitly from the corresponding system of ordinary differential equations ( $\mathbf{k}, t$ -dependence in the right-hand side is not written for brevity):

$$\begin{aligned} \exp [x\Lambda(\mathbf{k}, t)]_{cc} &= \\ \frac{\exp [xz_c] \left( \Lambda_{cc} - \Lambda_{vv} + \sqrt{D} \right) + \exp [xz_v] \left( \Lambda_{vv} - \Lambda_{cc} + \sqrt{D} \right)}{2\sqrt{D}}, \end{aligned}$$

$$\begin{aligned}
\exp [x\Lambda(\mathbf{k}, t)]_{cv} &= \frac{\exp[xz_c] - \exp[xz_v]}{\sqrt{D}} \Lambda_{cv}, \\
\exp [x\Lambda(\mathbf{k}, t)]_{vc} &= \frac{\exp[xz_c] - \exp[xz_v]}{\sqrt{D}} \Lambda_{vc}, \\
\exp [x\Lambda(\mathbf{k}, t)]_{vv} &= \\
\frac{\exp[xz_v] (\Lambda_{cc} - \Lambda_{vv} + \sqrt{D}) + \exp[xz_c] (\Lambda_{vv} - \Lambda_{cc} + \sqrt{D})}{2\sqrt{D}} & \quad (\text{B.4})
\end{aligned}$$

In Eqn. (B.4)  $z_c(\mathbf{k}, t)$ ,  $z_v(\mathbf{k}, t)$  denote eigenvalues of the quadratic form in the exponent of relevant operator  $\tilde{\rho}_{rel}(t)$ :

$$\begin{aligned}
z_c(\mathbf{k}, t) &= \frac{\Lambda_{cc}(\mathbf{k}, t) + \Lambda_{vv}(\mathbf{k}, t) + \sqrt{D(\mathbf{k}, t)}}{2}, \\
z_v(\mathbf{k}, t) &= \frac{\Lambda_{cc}(\mathbf{k}, t) + \Lambda_{vv}(\mathbf{k}, t) - \sqrt{D(\mathbf{k}, t)}}{2}, \\
D(\mathbf{k}, t) &= (\Lambda_{cc}(\mathbf{k}, t) - \Lambda_{vv}(\mathbf{k}, t))^2 + 4|\Lambda_{cv}(\mathbf{k}, t)|^2. \quad (\text{B.5})
\end{aligned}$$

Lagrange multipliers  $\Lambda_{\alpha\beta}(\mathbf{k}, t)$  could be eliminated due to self-consistency conditions (3.3), which can be rewritten in the following form:

$$[\exp\{\Lambda(\mathbf{k}, t)\} + \mathcal{I}]_{\alpha\beta}^{-1} = f_{\alpha\beta}(\mathbf{k}, t), [\exp\{\Lambda^{ph}(\mathbf{q}, t)\} - 1]^{-1} = n(\mathbf{q}, t). \quad (\text{B.6})$$

The explicit form of  $\Lambda$  will be needed for further numerical calculations. For phonon counterpart it is easy to write down the following relation:

$$\Lambda^{ph}(\mathbf{q}, t) = \ln \left( \frac{1 + n(\mathbf{q}, t)}{n(\mathbf{q}, t)} \right). \quad (\text{B.7})$$

To obtain more than formal matrix relation  $\Lambda_{\alpha\beta}(\mathbf{k}, t) = [\ln f(\mathbf{k}, t)^{-1} - \mathcal{I}]_{\alpha\beta}$ , we have calculated all Lagrange multipliers explicitly taking use of (B.4)-(B.5) (again we omitted time-energy dependence of corresponding functions):

$$\begin{aligned}
\Lambda_{cc} &= \frac{1}{2} \ln \left( \frac{X^2 - \Delta}{4} \right) + \frac{[f]_{cc}^{-1} - [f]_{vv}^{-1}}{\sqrt{\Delta}} \text{arcth} A, \\
\Lambda_{cc} &= \frac{1}{2} \ln \left( \frac{X^2 - \Delta}{4} \right) - \frac{[f]_{cc}^{-1} - [f]_{vv}^{-1}}{\sqrt{\Delta}} \text{arcth} A, \quad (\text{B.8})
\end{aligned}$$

$$|\Lambda_{cv}|^2 = \frac{4\text{arcth}^2 A}{\Delta} [f]_{cv}^{-1} [f]_{vc}^{-1},$$

where

$$A = \frac{\sqrt{\Delta}}{X}, \quad \Delta = ([f]_{cc}^{-1} - [f]_{vv}^{-1})^2 + 4[f]_{cv}^{-1} [f]_{vc}^{-1}, \quad X = [f]_{cc}^{-1} + [f]_{vv}^{-1} - 2. \quad (\text{B.9})$$

As one can see, all Lagrange multipliers as well as matrix elements are in the closed form being expressed via one-particle distribution functions in a very complicated way. Noting that off-diagonal elements of distribution functions tend to zero in field-free case (after the action of laser pulse), one can easily reproduce a result for effective one-zone semiconductor [12].

## Appendix C

After transition from sum over wave-vectors to corresponding integrals with subsequent substitution  $\hbar^2 k^2 / (2\mu) \rightarrow e_{\mathbf{k}}$ , where  $\mu = \frac{m_c m_v}{m_c + m_v}$  and taking use of factorization properties (3.11) of the kinetic kernels, one can write down the following schematic system of equations:

$$\begin{aligned}
\frac{f(e_{\mathbf{k}}, t)}{dt} &= \\
&= \int_{t_0}^t dt' \sum_i \int de'_k de''_k \kappa_i(e_{\mathbf{k}}, t) \bar{\kappa}_i(e'_k, t') F_i(\{f(e_{\mathbf{k}}, t'), f(e'_k, t'), f(e''_k, t')\}) \\
&= \sum_i \int de'_k de''_k \kappa_i(e_{\mathbf{k}}, t) G_i(e_{\mathbf{k}}, e'_k, e''_k, t) \quad (\text{C.1})
\end{aligned}$$

with supplementary variables

$$G_i(e_{\mathbf{k}}, e'_k, e''_k, t) = \int_{t_0}^t dt' \bar{\kappa}_i(e'_k, t') F_i(\{f(e_{\mathbf{k}}, t'), f(e'_k, t'), f(e''_k, t')\}) \quad (\text{C.2})$$

for which one gets a local differential equations

$$\frac{dG_i(e_{\mathbf{k}}, e'_k, e''_k, t)}{dt} = \bar{\kappa}_i(e'_k, t) F_i(\{f(e_{\mathbf{k}}, t), f(e'_k, t), f(e''_k, t)\}) \quad (\text{C.3})$$

with initial conditions  $G_i(e_{\mathbf{k}}, e'_k, e''_k, t = t_0) = 0$ . Summing up, we come to the system of ordinary differential equations where energy appears

as a parameter. Then, passing from integrals over  $e'_k, e''_k$  to the integral sums with corresponding Gaussian weights, we obtain the system of

$$\dim = 5N^2 + 12N^3 + 1$$

equations. Here  $5N$  equations are for one particle distribution functions (for carriers, phonons and polarization),  $12N^3$  equations describe dynamics of the auxiliary kernels (C.2), and the last equation determines the generalized temperature  $T(t)$ . Taking the energy grid dense enough with  $N = 60$  points we come to the system of about  $2.5 \times 10^6$  equations which are being solved numerically with given initial conditions.

## References

1. J. Shah. Ultrafast Spectroscopy of Semiconductors and Semiconductor Microstructures. Springer Series in Solid-State Sci., vol. 115, Berlin/Heidelberg, Springer, 1997.
2. F.Rossi and T.Kuhn.// Rev. Mod. Phys., 2002, vol.74, No 3, p.895.
3. C. Fürst, A. Leitenstorfer, A. Laubereau and R. Zimmermann.// Phys. Rev Lett., 1997, vol. 78, p. 3733.
4. R. Zimmermann, J. Wauer, and A. Leitenstorfer: Non-Markovian dynamics in optically detected electron-phonon relaxation in: Radiative Processes and Dephasing in Semiconductors, ed. D. S. Citrin, Optical Society of America Trends in Optics and Photonic Series (OSA TOPS), 1998, vol. 18, p. 2.
5. R. Zimmermann, J. Wauer, and A. Leitenstorfer: Non-Markovian dynamics in electron-phonon relaxation, Proc. 24th. Int. Conf. on the Physics of Semiconductors, Jerusalem 1998, Ed. D. Gershoni (World Scientific, Singapore 1999), paper II-E-4.
6. R. Zimmermann, J. Wauer, A. Leitenstorfer, and C. Fürst.// Journ. Luminescence, 1998, vol. 76-77, p. 34.
7. V.G. Morozov, G. Röpke.// Physica A, 1995, vol. 221, p. 511.
8. A.V. Kuznetsov.// Phys. Rev. B, 1991, vol. 44, p. 8721.
9. A.V. Kuznetsov.// Phys. Rev. B, 1991, vol. 44, p. 13 381
10. H. Haug, A.-P. Jauho. Quantum Kinetics in Transport and Optics of Semiconductors, Springer Series in Solid State Sci., vol. 123, Berlin/Heidelberg, Springer, 1996.
11. D.N. Zubarev, V.G. Morozov, G. Röpke. Statistical Mechanics of Nonequilibrium Processes, vol.1, Akademie Verlag, Berlin, 1997.
12. V. G. Morozov.// Cond. Matt. Phys., 2000, vol. 3, No. 3(23), p. 577.
13. V.G. Morozov, G. Röpke.// Journ. Stat. Phys., 2001, vol.102, No 1, p.285.
14. V.G.Morozov and G.Röpke.// Annals of Physics, 1999, vol.278, p.127.
15. M. Bonitz, D. Kremp, D.C. Scott, R. Binder, W.D. Kraeft and H.S. Köhler.// J. Phys.: Condens. Matter, 1996, vol. 8, p. 6057.
16. K. Morawetz.// Phys. Lett. A, 1995, vol. 199, p.241.
17. K. Morawetz, H.S. Köhler.// The Euro. Phys. Journ., 1999, vol. 4, p.291.
18. D. Kremp, M. Bonitz, W.D. Kraeft and M. Schlanges.// Ann. of Phys., 1997, vol.258, p.320.
19. B. Rethfeld, A. Kaiser, M. Vičánek and G. Simon.// Phys. Rev. B, 2002, vol. 65, p.214303.



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Василь Васильович Ігнатюк  
Володимир Георгійович Морозов

НЕМАРКІВСЬКІ КВАНТОВІ КІНЕТИЧНІ РІВНЯННЯ ДЛЯ  
НАПІВПРОВІДНИКІВ У ПОЛІ ЛАЗЕРНОГО ІМПУЛЬСУ

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