Time-convolutionless reduced-density-operator theory of an arbitrary driven system coupled to a stochastic reservoir: Quantum kinetic equations for semiconductors

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(Received 18 May 1994)

In this paper two things are done. (1) A projection-operator formalism is used to derive the time-convolutionless stochastic equation of motion for the reduced density operator from the quantum Liouville equation for an arbitrary driven system coupled to a stochastic reservoir. As an initial condition, decoupling of the system and reservoir for the total-density operator is assumed in the formulation. Perturbation expansions of the generalized collision operator are carried out in powers of the driving field within the Born approximation for the interaction of the system with the reservoir. The time-convolutionless form of the equation for the reduced density operator allows one to include the memory effects systematically. (2) Time-convolutionless quantum kinetic equations for interacting electron-hole pairs near the band edge in semiconductors under an arbitrary optical field are obtained from the equation of motion for the reduced density operator. These equations generalize the semiconductor Bloch equations to incorporate the non-Markovian relaxation and the interference effects between the external driving field and the stochastic reservoir of the system and are valid to any time scale. It is shown that the interference term modulates the interband polarization and includes the renormalized memory effects.

I. INTRODUCTION

Recent advances in the theory of femtosecond optical pulse spectroscopy[1–4] and quantum transport theory[5–10] draw much attention to the ultrafast relaxation kinetics of the electrons and holes near the band edge in semiconductors. These relaxation kinetics in nonequilibrium cases are often characterized by the presence of memory effects and are also important in related areas such as nonlinear optical gain in semiconductors in which the competition between the stimulated emission and the intraband relaxation contribute to the spectral hole burning. In these nonequilibrium kinetics, the system has memory effects on a very short time scale and the equations of motion for the system have time-convolution forms of integral kernels which are responsible for the memory effects.[1–4] These quantum kinetic equations can be obtained with reduced-density matrices and with nonequilibrium Green's-function theory. In order to obtain numerically stable kinetic equations a consistent treatment of the memory kernels of the equations and the Green's functions or the density matrices for the scattering processes is needed.[4] In general, it is very difficult to solve for the memory kernels of the time-convolution forms of the equation self-consistently and almost always, one must be content with the narrowing limit or the fast modulation limit to obtain the non-Markovian relaxation.

Some time ago, Tokuyama and Mori[11] suggested the time-convolutionless equations of motion in the Heisenberg picture for problems in nonequilibrium statistical mechanics. These formulations were then developed in the Schrödinger picture by Shibata and co-workers[12–14] by using the projection operator technique. They obtained equations of motion for a reduced density operator of a system interacting with the surroundings. Saeki generalized these equations by considering the response of the system to an external driving field.[15–18] He derived generalized master equations for an arbitrary driven system interacting with the heat bath[16] and for a weakly driven system interacting with the stochastic reservoir.[17,18] It was shown that the time-convolutionless equations of motion incorporate both non-Markovian relaxation and renormalization of the memory effects.

Recently, Tomita and Suzuki used the time-convolutionless equations in the lowest Born approximation to obtain the density-matrix theory of nonlinear gain for noninteracting electron-hole pairs in semiconductors and showed that the non-Markovian relaxation enhances both linear and nonlinear optical gains.[19] Many-body effects such as band-gap renormalization and Coulomb enhancement are not considered in their work. On the other hand, recent calculations by the author showed that the band-gap renormalization effects are pronounced in microstructures such as strained-layer quantum wells and are important in analyzing the optical gain spectrum for long-wavelength semiconductor lasers.[20,21]

In this paper, we first extended the work[17] of Saeki on a stochastic Liouville equation for a weakly driven system to derive a time-convolutionless equation for a reduced density operator of an arbitrary driven system coupled to the stochastic reservoir. It is found that the density operator method is convenient for us to transform the memory kernel into a time-convolutionless form which is suitable for the perturbation expansions in the system-reservoir interaction and the driving field. Secondly we apply the formulation to obtain time-convolutionless quantum kinetic equations for the system of interacting electron-hole pairs under arbitrary external optical field. These equations are the generalization of the semiconductor Bloch equations[22–28] by incorporating the non-Markovian relaxation and the renormalization of the memory effects through the interference between the external driving field and the stochastic reservoir.

II. TIME-CONVOLUTIONLESS EQUATION FOR A REDUCED DENSITY OPERATOR OF AN ARBITRARY DRIVEN SYSTEM

In this section, we extend the work[17] of Saeki on a stochastic Liouville equation for a weakly driven system to derive an equation for a reduced density operator of an arbitrary driven system coupled to a stochastic reservoir. We consider an arbitrary driven system interacting with a stochastic reservoir and assume that the interaction of the system with its surroundings can be represented by a stochastic Hamiltonian. The Hamiltonian of the total system is assumed to be

$$\begin{aligned} H_T(t) &= H_0(t)+H_i(t)+H_{\rm ext}(t) \\ &= H(t)+H_{\rm ext}(t) \\ &= H_s(t)+H_i(t) , \end{aligned} \tag{1}$$

where $H_0(t)$ is the Hamiltonian of the system, $H_{\rm ext}(t)$ the interaction of the system with the external driving field, and $H_i(t)$ the Hamiltonian for the interaction of the system with its stochastic reservoir. The equation of motion for the density operator $\rho_T(t)$ of the total system is given by a stochastic Liouville equation

$$\begin{aligned} \frac{d\rho_T(t)}{dt} &= -i[H_T(t),\rho_T(t)] \\ &= -iL_T(t)\rho_T(t) , \end{aligned} \tag{2}$$

where

$$\begin{aligned} L_T(t) &= L_0(t)+L_i(t)+L_{\rm ext}(t) \\ &= L(t)+L_{\rm ext}(t)=L_s(t)+L_i(t) \end{aligned}$$

is the Liouville superoperator in one-to-one correspondence with the Hamiltonian. In this paper, we use a unit where $\hbar=1$. It is convenient to introduce the projection operators[29–31] which decompose the total system by eliminating the dynamical variables of the stochastic reservoir. We define time-independent projection operators $\underline{P}$ and $\underline{Q}$ as[17]

$$\underline{P}X=p_0(R)\langle X\rangle_i, \quad \underline{Q}=1-\underline{P} , \tag{3}$$

for any dynamical variable X. Here $p_0(R)$ is the initial distribution function of the random variable R and $\langle \cdots \rangle_i$ is the average over the stochastic process $H_i(t)$.

Projection operators $\underline{P}$ and $\underline{Q}$ satisfy the operator identity $\underline{P}^2=\underline{P}$, $\underline{Q}^2=\underline{Q}$, and $\underline{P}\,\underline{Q}=\underline{Q}\,\underline{P}=0$. The information of the system is then contained in the reduced density operator $\rho(t)$ which is defined by

$$\rho(t)=\langle \underline{P}\rho_T(t)\rangle_i . \tag{4}$$

In order to derive a time-convolutionless equation, we first multiply Eq. (1) by $\underline{P}$ and $\underline{Q}$ to obtain coupled equations for $\underline{P}\rho_T(t)$ and $\underline{Q}\rho_T(t)$:

$$\frac{d}{dt}\underline{P}\rho_T(t)=-i\underline{P}L_T(t)\underline{P}\rho_T(t)-i\underline{P}L_T(t)\underline{Q}\rho_T(t) \tag{5}$$

and

$$\frac{d}{dt}\underline{Q}\rho_T(t)=-i\underline{Q}L_T(t)\underline{Q}\rho_T(t)-i\underline{Q}L_T(t)\underline{P}\rho_T(t) , \tag{6}$$

where we use the identity $\underline{P}+\underline{Q}=1$.

We assume that the external driving field is turned on at $t=0$ and the total system was in an arbitrary initial condition $\rho_T(0)$.

It can be shown that the formal solution of (6) is given by

$$\begin{aligned} \underline{Q}\rho_T(t) = &-i\int_0^t d\tau\, \underline{H}(t,\tau)\underline{Q}L_T(\tau)\underline{P}\rho_T(\tau) \\ &+\underline{H}(t,0)\underline{Q}\rho_T(0) . \end{aligned} \tag{7}$$

where the projected propagator $\underline{H}(t,\tau)$ of the total system is defined as

$$\underline{H}(t,\tau)=\underline{T}\exp\left\{-i\int_\tau^t ds\, \underline{Q}L_T(s)\underline{Q}\right\} . \tag{8}$$

Here $\underline{T}$ denotes the time-ordering operator. Next, we transform the memory kernel in (7) into the time-convolutionless form by substituting the formal solution of (2)

$$\rho_T(\tau)=\underline{G}(t,\tau)\rho_T(t) \tag{9}$$

into Eq. (7). Here the evolution operator $\underline{G}(t,\tau)$ of the total system is given by

$$\underline{G}(t,\tau)=\underline{T}^c\exp\left\{i\int_\tau^t ds L_T(s)\right\} , \tag{10}$$

where $\underline{T}^c$ is the anti-time-ordering operator. Evolution operators $\underline{G}(t,\tau)$ and $\underline{H}(t,\tau)$ satisfy the following relations:

$$\underline{G}(t,\tau)\underline{G}(s,t)=\underline{G}(s,\tau) \tag{11}$$

and

$$\underline{H}(t,\tau)\underline{H}(\tau,s)=\underline{H}(t,s) . \tag{12}$$

From Eqs. (7) and (9), we obtain

$$\underline{Q}\rho_T(t)=\{\theta(t)-1\}\underline{P}\rho_T(t)+\theta(t)\underline{H}(t,0)\underline{Q}\rho_T(0) , \tag{13}$$

where

$$\begin{aligned} \theta(t)^{-1} &= g(t) \\ &= 1+i\int_0^t d\tau\, \underline{H}(t,\tau)\underline{Q}L_T(\tau)\underline{P}\,\underline{G}(t,\tau) . \end{aligned} \tag{14}$$

The time-convolutionless equation of motion for $\underline{P}\rho_T(t)$ can be obtained from (5) and (13) as

$$\begin{aligned} \frac{d}{dt}\underline{P}\rho_T(t) = &-i\underline{P}L_T(t)\underline{P}\rho_T(t)-i\underline{P}L_T(t)\{\theta(t)-1\}\underline{P}\rho_T(t) \\ &-i\underline{P}L_T(t)\theta(t)\underline{H}(t,0)\underline{Q}\rho_T(0) . \end{aligned} \tag{15}$$

It is now straightforward to obtain the time-convolutionless equation of motion for a reduced density operator $\rho(t)$. By taking the average of (15) over the stochastic process $H_i(t)$, we get

$$\frac{d}{dt}\rho(t)=-i[L_S(t)+\langle L_i(t)\rangle_i]\rho(t)+C(t)\rho(t) , \quad (16)$$

where the generalized collision operator $C(t)$ is defined by

$$C(t)=-i\langle L_i(t)\{\theta(t)-1\}\rangle_i$$
$$=-i\langle L_i(t)\Sigma(t)\{1-\Sigma(t)\}^{-1}\rangle_i , \quad (17)$$

in which

$$\Sigma(t)=1-\theta(t)^{-1}$$
$$=-i\int_0^t d\tau \underline{H}(t,\tau)\underline{Q}L_T(\tau)\underline{P}\ \underline{G}(t,\tau)$$
$$=-i\int_0^t d\tau\, \underline{U}(t)\underline{S}(t,\tau)\underline{U}^{-1}(\tau)\underline{Q}L_T(\tau)$$
$$\times \underline{P}\ \underline{U}(\tau)\underline{R}(t,\tau)\underline{U}^{-1}(t) . \quad (18)$$

Here we define

$$\underline{U}(t)=\underline{T}\exp\left\{-i\int_\tau^t ds\, L_s(s)\right\} , \quad (19)$$

$$\underline{S}(t,\tau)=\underline{T}\exp\left\{-i\int_\tau^t ds\, \underline{Q}\underline{U}^{-1}(s)L_i(s)\underline{U}(s)\underline{Q}\right\} , \quad (20)$$

and

$$\underline{R}(t,\tau)=\underline{T}^c\exp\left\{-i\int_\tau^t ds\, \underline{U}^{-1}(s)L_i(s)\underline{U}(s)\right\} , \quad (21)$$

where $\underline{U}(t)$ is the evolution operator of the system with driving field, and $\underline{R}(t,\tau)$ and $\underline{S}(t,\tau)$ are the evolution operators and the projected propagators of the total system in the interaction picture, respectively. In (16), we assumed that the initial condition $\rho_T(0)$ is given by

$$\rho_T(0)=\rho(0)p_0(R) , \quad (22)$$

which means that the system and the reservoir were uncoupled before the external driving field is turned on and that the system was in an arbitrary state $\rho(0)$ at $t=0$. Then it is obvious that $\underline{Q}\rho_T(0)=0$.

We now consider the case when the system is interacting weakly with the stochastic reservoir and expand (16) up to the second order in powers of the stochastic Hamiltonian $H_i(t)$. We assume that the random force vanishes on the average over the stochastic process,[17] i.e.,

$$\underline{P}L_i(t)\underline{P}=0 . \quad (23)$$

We further assume that the stochastic process is stationary.

The equation of motion for $\rho(t)$ up to the second-order expansion in $H_1(t)$ becomes

$$\frac{d}{dt}\rho(t)=-iL_s(t)\rho(t)+C^{(2)}(t)\rho(t) , \quad (24)$$

where

$$C^{(2)}(t)=-\langle L_i(t)\Sigma^{(1)}(t)\rangle_i$$
$$=-\int_0^t d\tau\langle L_i(t)\underline{U}(t,\tau)L_i(\tau)\underline{U}^{-1}(t,\tau)\rangle_i . \quad (25)$$

with

$$\Sigma^{(1)}(t)=-i\int_0^t d\tau\, \underline{U}(t,\tau)L_i(\tau)\underline{U}^{-1}(t,\tau) \quad (26)$$

and $\underline{U}(t,\tau)=\underline{U}(t)U^{-1}(\tau)$. Some important mathematical properties of the evolution operators are summarized in the Appendix. Using Eq. (A6) of the Appendix, we transform $C^{(2)}(t)$ into an expression more suitable for the perturbation expansions with respect to $H_{\text{ext}}(t)$:

$$C^{(2)}(t)=-\int_0^t d\tau\langle L_i(t)\underline{U}_0(t)\underline{U}_{\text{ext}}(t,\tau)\underline{U}_0^{-1}(\tau)$$
$$\times L_i(\tau)\underline{U}_0(\tau)\underline{U}_{\text{ext}}^{-1}(t,\tau)\underline{U}_0^{-1}(t)\rangle_i . \quad (27)$$

We can expand $C^{(2)}(t)$ in powers of the driving field as

$$C^{(2)}(t)=\sum_{m=0}^{\infty} C_m^{(2)}(t) , \quad (28)$$

where $C_m^{(2)}(t)$ is the mth-order term given by

$$C_m^{(2)}(t)=-\sum_{k=0}^{m}(-i)^k(i)^{m-k}\int_0^t d\tau\int_\tau^t d\tau_1\int_\tau^{\tau_1} d\tau_2\cdots\int_\tau^{\tau_{k-1}} d\tau_k\int_\tau^t d\tau_{k+1}\cdots\int_\tau^{\tau_{m-1}} d\tau_m\langle L_i(t)\Phi_k(t,\tau_1,\ldots,\tau_k,\tau)L_i(\tau)$$
$$\times\Psi_{m-k}(t,\tau_{k+1},\ldots,\tau_m,\tau)\rangle_i , \quad (29)$$

with

$$\Phi_0(t,\tau)=\underline{U}_0(t-\tau) , \quad (30)$$

$$\Phi_k(t,\tau_1,..,\tau_k,\tau)=\underline{U}_0(t-\tau_1)L_{ext}(\tau_1)\underline{U}_0(\tau_1-\tau_2)$$
$$\times L_{\text{ext}}(\tau_2)\underline{U}_0(\tau_2-\tau_3)\cdots\underline{U}_0(\tau_{k-1}-\tau_k)$$
$$\times L_{\text{ext}}(\tau_k)\underline{U}_0(\tau_k-\tau) , \quad (31)$$

$$\Psi_0(t,\tau)=\underline{U}_0(\tau-t) , \quad (32)$$

and

$$\Psi_k(t,\tau_1,\ldots,\tau_k,\tau)=\underline{U}_0(\tau-\tau_k)L_{\text{ext}}(\tau_k)$$
$$\times\underline{U}_0(\tau_k-\tau_{k-1})L_{\text{ext}}(\tau_{k-1})$$
$$\times\underline{U}_0(\tau_{k-2}-\tau_{k-3})\cdots\underline{U}_0(\tau_2-\tau_1)$$
$$\times L_{\text{ext}}(\tau_1)\underline{U}_0(\tau_1-t) . \quad (33)$$

The time-convolutionless equation of motion for a reduced density operator given in (24) with (28)–(33) can be used in any time scale such as the femtosecond regime and is valid up to the second order in powers in the interaction between the system and the stochastic reservoir. In the next section, a time-convolutionless equation for a reduced density operator is used to obtain quantum kinetic equations for the system of interacting electronhole pairs near the band edge in semiconductors under an arbitrary optical field.

III. TIME-CONVOLUTIONLESS QUANTUM KINETIC EQUATIONS FOR INTERACTING ELECTRON-HOLE PAIRS IN SEMICONDUCTORS

In this section, we apply a time-convolutionless Eq. (24) to the system of interacting electron-hole pairs in semiconductors with an external driving field. We assume that the system is weakly interacting with its stochastic reservoir. Many-body effects such as band-gap renormalization and phase-space filling are included by taking into account the Coulomb interaction in the Hartree-Fock approximation. The stochastic Hamiltonian $H_i(t)$ may include electron-electron interaction and electron-LO phonon interaction for both conduction and valence electrons. We will not specify the explicit forms of H_i in this work and leave the detailed calculations of correlation functions involving H_i for a future work. Instead, we obtain the intraband relaxation and the dephasing as correlation functions of $H_i(t)$ in the present work.

We employ the two-band model for the semiconductor and introduce two short-handed notations $|ck\rangle$ and $|vk\rangle$ such that

$$|ck\rangle=|c,\mathbf{k}\rangle \quad \text{and} \quad |vk\rangle=|v,\mathbf{k}\rangle \ , \tag{34}$$

where c and v denote conduction and valence bands, respectively, and $\mathbf{k}$ is the electron wave vector. In the following we suppress the vector notation for simplicity.

In the time-dependent Hartree-Fock approximation, the unperturbed Hamiltonian $H_0(t)$ is given by[23,24,32]

$$\langle \alpha k|H_0(t)|\beta k\rangle=E_\alpha^0(k)\delta_{\alpha\beta} -\sum_{k'} V(k-k')\langle \alpha k'|\rho(t)|\beta k'\rangle \ , \tag{35}$$

where $\alpha,\beta=c$ or v and $V(k-k')$ is the Coulomb interaction.

The interaction of the system with an external driving field gives the interaction Hamiltonian H_{ext} which is given by

$$H_{\text{ext}}(t)=-ME_p(t) \ , \tag{36}$$

where M is the dipole operator and E_p is the electric field strength of the optical radiation.

The equation of motion for the reduced density operator becomes

$$\frac{d}{d\tau}\rho(t)=-i[L_0(t)+I_{\text{ext}}(t)]\rho(t)+C_0^{(2)}(t)\rho(t)+D_1^{(2)} \ , \tag{37}$$

where

$$C_0^{(2)}(t)\rho(t)=-\int_0^t d\tau\langle L_i(t)\underline{U}_0(\tau) \times L_i(t-\tau)\underline{U}_0^{-1}(\tau)\rangle_i\rho(t) \ , \tag{38}$$

and

$$D_1^{(2)}=C_1^{(2)}(t)\underline{U}_0(t)\rho(0) \ . \tag{39}$$

$C_1^{(2)}(t)$ is responsible for the intracollisional field effects and can be derived from (29),

$$\begin{aligned}C_1^{(2)}(t)=i\int_0^t d\tau\int_\tau^t ds\{&\langle L_i(t)\underline{U}_0(t-s)L_{\text{ext}}(s) \\ &\times\underline{U}_0(s-\tau)L_i(\tau)\underline{U}_0(\tau-t)\rangle_i \\ &-\langle L_i(t)\underline{U}_0(t-\tau)L_i(\tau)\underline{U}_0(\tau-s) \\ &\times L_{\text{ext}}(s)\underline{U}_0(s-t)\rangle_i\} \ .\end{aligned} \tag{40}$$

It can be shown that $D_1^{(2)}$ contains information of the effects of the interference of the external driving field with a stochastic reservoir of the system and is the renormalization of the memory effects.

Nonequilibrium distributions $n_{ck}(t)$, $n_{vk}(t)$ for electrons in the conduction band and in the valence band, respectively, and the nondiagonal interband matrix element $p_k^*(t)$ which describes the interband polarization induced by the optical field, are the matrix elements of the reduced density operator and are given by

$$n_{ck}(t)=\rho_{cck}(t) =\langle ck|\rho(t)|ck\rangle \ , \tag{41}$$

$$n_{vk}(t)=\rho_{vvk}(t) =\langle vk|\rho(t)|vk\rangle \ , \tag{42}$$

and

$$p_k^*(t)=\rho_{vck}(t) =\langle vk|\rho(t)|ck\rangle \ . \tag{43}$$

Next, we calculate the matrix elements of the collision term $C_0^{(2)}(t)\rho(t)$, $\langle ck|C_0^{(2)}(t)\rho(t)|ck\rangle$, $\langle vk|C_0^{(2)}(t)\rho(t)|vk\rangle$, and $\langle vk|C_0^{(2)}(t)\rho(t)|ck\rangle$ to obtain the non-Markovian intraband relaxation and dephasing. After some mathematical manipulations, we obtain

$$\langle ck|C_0^{(2)}(t)\rho(t)|ck\rangle = -\int_0^t d\tau \langle\langle ck|L_i(t)\underline{U}_0(\tau)L_i(t-\tau)\underline{U}_0^{-1}(\tau)\rho(t)|ck\rangle\rangle_i$$
$$= -\int_0^t d\tau \langle\langle ck|[H_i(t),[(\underline{U}_0(\tau)H_i(t-\tau)),\rho(t)]]|ck\rangle\rangle_i$$
$$\approx -2\int_0^t d\tau \,\mathrm{Re}\{\langle\langle ck|[H_i(t)(\underline{U}_0(\tau)H_i(t-\tau))]|ck\rangle\rangle_i\}\times\{n_{ck}(t)-(\rho_0^{(0)})_{cck}(t)\} , \quad (44)$$

$$\langle vk|C_0^{(2)}(t)\rho(t)|vk\rangle = -\int_0^t d\tau \langle\langle vk|L_i(t)\underline{U}_0(\tau)L_i(t-\tau)\underline{U}_0^{-1}(\tau)\rho(t)|vk\rangle\rangle_i$$
$$= -\int_0^t d\tau \langle\langle vk|[H_i(t),[(\underline{U}_0(\tau)H_i(t-\tau)),\rho(t)]]|vk\rangle\rangle_i$$
$$\approx -2\int_0^t d\tau \,\mathrm{Re}\{\langle\langle vk|[H_i(t)(\underline{U}_0(\tau)H_i(t-\tau))]|vk\rangle\rangle_i\}\{n_{vk}(t)-(\rho_0^{(0)})_{vvk}(t)\} . \quad (45)$$

and

$$\langle vk|C_0^{(2)}(t)\rho(t)|ck\rangle = -\int_0^t d\tau \langle\langle vk|L_i(t)\underline{U}_0(\tau)L_i(t-\tau)\underline{U}_0^{-1}(\tau)\rho(t)|ck\rangle\rangle_i$$
$$= -\int_0^t d\tau \langle\langle vk|[H_1(t),[(\underline{U}_0(\tau)H_i(t-\tau)),\rho(t)]]|ck\rangle\rangle_i$$
$$= -\int_0^t d\tau\{\langle\langle vk|[H_i(t)(\underline{U}_0(\tau)H_i(t-\tau))]|vk\rangle\rangle_i$$
$$+\langle\langle ck|[(\underline{U}_0(\tau)H_i(t-\tau))H_i(t)]|ck\rangle\rangle_i\}p_k^*(t) , \quad (46)$$

where $\rho_0^{(0)}(t)=\underline{U}_0(t)\rho(0)$. Equation (46) is the non-Markovian optical dephasing which is the temporal decay of the interband polarization due to scattering processes.

Similarly,

$$\langle ck|-i[L_0(t)+L_{\mathrm{ext}}(t)]\rho(t)|ck\rangle = -2\,\mathrm{Im}\left\{\left[\mu(k)E_p(t)+\sum_{k'}V(k\text{-}k')p_{k'}(t)\right]p_k^*(t)\right\} , \quad (47)$$

$$\langle vk|-i[L_0(t)+L_{\mathrm{ext}}(t)]\rho(t)|vk\rangle = 2\,\mathrm{Im}\left\{\left[\mu(k)E_p(t)+\sum_{k'}V(k\text{-}k')p_{k'}(t)\right]p_k^*(t)\right\} , \quad (48)$$

and

$$\langle vk|-i[L_0(t)+L_{\mathrm{ext}}(t)]\rho(t)|ck\rangle = i[E_c(k)-E_v(k)]p_k^*(t)$$
$$+i\left[\mu^*(k)E_p(t)+\sum_{k'}V(k\text{-}k')p_{k'}^*(t)\right][n_{ck}(t)-n_{vk}(t)] , \quad (49)$$

where $\mu(k)=\langle ck|M|vk\rangle$, and $E_c(k)$, $E_v(k)$ are renormalized single-particle energies given by

$$E_c(k)=E_c^0(k)-\sum_{k'}V(k\text{-}k')n_{ck'}^0 , \quad (50)$$

and

$$E_v(k)=E_v^0(k)-\sum_{k'}V(k\text{-}k')n_{vk'}^0 . \quad (51)$$

In (44)–(48), Re and Im denote the real and the imaginary part of the complex variable, respectively.

The last term $D_1^{(2)}$ of (37) is the interference of the driving field with the surroundings and is given by

$$D_1^{(2)}(t)=C_1^{(2)}(t)\rho_0^{(0)}(t)$$
$$=i\int_0^t d\tau\int_0^\tau ds\{\langle L_i(t)\underline{U}_0(t-\tau)L_{\mathrm{ext}}(\tau)\underline{U}_0(\tau-s)L_i(s)\underline{U}_0(s)\rho(0)\rangle_i$$
$$-\langle L_i(t)\underline{U}_0(t-s)L_i(s)\underline{U}_0(s-\tau)L_{\mathrm{ext}}(\tau)\underline{U}_0(\tau)\rho(0)\rangle_i\}$$
$$=i\int_0^t d\tau\int_0^\tau ds\langle[H_i(t),[(\underline{U}_0(s)H_{\mathrm{ext}}(t-s)),(\underline{U}_0(\tau)H_i(t-\tau))],\rho_0^{(0)}(t)]]\rangle_i , \quad (52)$$

where we have made use of the transformation of integral variables and the commutation relation between the operators A, B, and C

$$[A,[B,C]]-[B,[A,C]]=[[A,B],C] .$$

It can be shown that the interference effects on the intraband collisions vanish to the first order if $\langle vk|\rho_0^{(0)}(t)|ck\rangle$ vanishes. In other words, we assume that

$$\langle ck|D_1^{(2)}(t)|ck\rangle = \langle vk|D_1^{(2)}(t)|vk\rangle = 0 .$$

In the following, we include the effects of the interference between the stochastic reservoir and the external optical field only in the interband kinetics. It is involved but straightforward to evaluate the matrix element of $D_1^{(2)}$ and we suppress the details of the algebra.

The result is

$$\begin{aligned}\langle vk|D_1^{(2)}(t)|ck\rangle &= i\int_0^t d\tau\int_0^\tau ds\,\exp\{-i[E_v(k)-E_c(k)](t-\tau)\}\{\langle\langle vk|[H_1(t)(\underline{U}_0(t-s)H_1(s))]|vk\rangle\rangle_i \\ &\quad + \langle\langle ck|[(\underline{U}_0(t-s)H_i(s))H_i(t)]|ck\rangle\rangle_i\} \\ &\quad \times\mu^*(k)E_p(t)\{(\rho_0^{(0)})_{cck}(t)-(\rho_0^{(0)})_{vvk}(t)\} \\ &\approx i\int_0^t d\tau\int_0^\tau ds\,\exp\{-i[E_v(k)-E_c(k)]s\}\{\langle\langle vk|[H_i(t)(\underline{U}_0(\tau)H_i(t-\tau))]|vk\rangle\rangle_i \\ &\quad + \langle\langle ck|[(\underline{U}_0(\tau)H_i(t-\tau))H_i(t)]|ck\rangle\rangle_i\} \\ &\quad \times\mu^*(k)E_p(t-s)\{(\rho_0^{(0)})_{cck}(t)-(\rho_0^{(0)})_{vvk}(t)\}\,, \end{aligned} \tag{53}$$

where we have made use of the transformation of integral variables and dropped the Coulomb exchange term between electrons in the conduction and the valence bands.

Using (41)–(53), we finally obtain time-convolutionless quantum kinetic equations for $n_{ck}(t)$, $n_{vk}(t)$, and $p_k^*(t)$,

$$\begin{aligned}\frac{\partial}{\partial t}n_{ck}(t) &= -2\,\mathrm{Im}\left\{\left[\mu(k)E_p(t)+\sum_{k'}V(k-k')p_{k'}(t)\right]p_k^*(t)\right\} \\ &\quad -2\int_0^t d\tau\,\mathrm{Re}\{\langle\langle ck|[H_i(t)(\underline{U}_0(\tau)H_i(t-\tau))]|ck\rangle\rangle_i\}\{n_{ck}(t)-(\rho_0^{(0)})_{cck}(t)\}\,,\end{aligned} \tag{54}$$

$$\begin{aligned}\frac{\partial}{\partial t}n_{vk}(t) &= 2\,\mathrm{Im}\{[\mu(k)E_p(t)+\sum_{k'}V(k-k')p_{k'}(t)]p_k^*(t)\} \\ &\quad -2\int_0^t d\tau\,\mathrm{Re}\{\langle\langle vk|[H_i(t)(\underline{U}_0(\tau)H_i(t-\tau))]|vk\rangle\rangle_i\}\{n_{vk}(t)-(\rho_0^{(0)})_{vvk}(t)\}\,.\end{aligned} \tag{55}$$

and

$$\begin{aligned}\frac{\partial}{\partial t}p_k^*(t) &= i[E_c(k)-E_v(k)]p_k^*(t)+i\left[\mu^*(k)E_p(t)+\sum_{k'}V(k-k')p_{k'}^*(t)\right][n_{ck}(t)-n_{vk}(t)] \\ &\quad -\int_0^t d\tau\{\langle\langle vk|[H_i(t)(\underline{U}_0(\tau)H_i(t-\tau))]|vk\rangle\rangle_i+\langle\langle ck|[(\underline{U}_0(\tau)H_i(t-\tau))H_i(t)]|ck\rangle\rangle_i\}p_k^*(t) \\ &\quad +i\int_0^t d\tau\int_0^\tau ds\,\exp\{-i[E_v(k)-E_c(k)]s\}\{\langle\langle vk|[H_i(t)(\underline{U}_0(\tau)H_i(t-\tau))|vk\rangle\rangle_i \\ &\quad + \langle\langle ck|[(\underline{U}_0(\tau)H_i(t-\tau))H_i(t)]|ck\rangle\rangle_i\} \\ &\quad \langle\mu^*(k)E_p(t-s)\{(\rho_0^{(0)})_{cck}(t)-(\rho_0^{(0)})_{vvk}(t)\}\,.\end{aligned} \tag{56}$$

The last term of (56), $\langle vk|D_1^{(2)}(t)|ck\rangle$, modulates the interband polarization due to the interference of the driving optical field and a stochastic reservoir of the system and gives the renormalized memory effects. Comparing our time-convolutionless quantum kinetic Eqs. (54)–(56) with previous quantum kinetic equations with memory kernels,[1–4] we can see that the time-convolutionless equations give the non-Markovian relaxation (both intraband relaxation and dephasing) and the renormalized memory effects through the modulation of the interband polarization, self-consistently. Moreover, time-convolutionless equations are in a form convenient for the perturbation expansions in powers of the stochastic Hamiltonian H_1 and the interaction with the driving field H_{ext} and are valid to very short time scale.

It can be shown that our quantum kinetic equations are reduced to the conventional density-matrix equations in the Markovian limit. In order to analyze the collision term $C_0^{(2)}(t)\rho(t)$ and the interference term $D_1^{(2)}(t)$ in the Markovian limit, we put

$$\langle\langle \alpha k|[H_1(t)(\underline{U}_0(\tau)H_1(t-\tau))]|\alpha k\rangle\rangle_1 = \frac{1}{2\tau_\alpha(k)}\delta(|\tau|)\,. \tag{57}$$

Then, the intraband relaxation and the dephasing terms become

$$\langle ck|C_0^{(2)}(t)\rho(t)|ck\rangle = -\frac{1}{\tau_c(k)}\{n_{ck}(t)-(\rho_0^{(0)})_{cck}(t)\}\,, \tag{58}$$

$$\langle vk|C_0^{(2)}(t)\rho(t)|vk\rangle = -\frac{1}{\tau_v(k)}\{n_{vk}(t)-(\rho_0^{(0)})_{vvk}(t)\}\,, \tag{59}$$

and

$$\langle vk|C_0^{(2)}(t)\rho(t)|ck\rangle = -\frac{1}{\tau_{vc}(k)}p_k^*(t)\,, \tag{60}$$

with

$$\frac{1}{\tau_{vc}(k)}=\frac{1}{2}\left\{\frac{1}{\tau_v(k)}+\frac{1}{\tau_c(k)}\right\} . \quad (61)$$

In the Markovian approximation, the interference term $\langle vk|D_1^{(2)}(t)|ck\rangle$ vanishes because in the integration over ds, the upper limit τ is zero and the resulting integral in (53) becomes zero because of (57). Intraband relaxation (58), (59) and dephasing (60) characterized by the dephasing time (61) agree well with the conventional density-matrix theory[33,34] with Markovian relaxation. As a result the memory effects vanish in the Markovian limit because there is no correlation between the stochastic process $H_i(t-\tau)$ and $H_i(t)$. Physically,

$$\langle\langle \alpha k|[H_i(t)(\underline{U}_0(\tau)H_i(t-\tau))]\alpha k\rangle\rangle_i$$

represents the averaged probability amplitude of finding a particle in the state $|\alpha k\rangle$ after being scattered at t by $H_i(t)$ when it was initially in the state $|\alpha k\rangle$, and then getting scattered at $t-\tau$ by $H_i(t-\tau)$ and go as a free particle for the time interval τ. It is obvious that the Markovian approximation assumes each interaction with the reservoir randomizes the previous information contained in the wave function and treats scattering processes $H_i(t)$ and $H_i(t-\tau)$ as an independent stochastic processes.

In the non-Markovian theory, the memory effects extend over the time interval τ_c the correlation time of the stochastic processes.

For example, if we assume the simplest form of the non-Markovian correlation function

$$\langle\langle \alpha k|[H_i(t)(\underline{U}_0(\tau)H_i(t-\tau))|\alpha k\rangle\rangle_i$$

as

$$\langle\langle \alpha k|[H_i(t)(\underline{U}_0(\tau)H_i(t-\tau))|\alpha k\rangle\rangle_i = \frac{1}{2\tau_c\tau_\alpha(k)}\exp[-|\tau|/\tau_c] , \quad (62)$$

the intraband relaxation and the dephasing terms are reduced to the results of the Markovian approximation when $t=\infty$.

In the derivation of the ordinary semiconductor Bloch equations, we completely neglect all the kinetic effects of $D_1^{(2)}(t)$ and use the Markovian approximation in the narrowing limit $\tau_r \gg \tau_c$ in the collision operator $C_0^{(2)}(t)\rho(t)$. Here τ_r denotes the macroscopic relaxation time of the system. In this way, we get the familiar semiconductor Bloch equations which describe the particles as free particles in between collisions or interactions with photons. The more general Eqs. (54)–(56) include the effects of the non-Markovian relaxation on the motion of particles between collisions. The interband kinetic equations incorporate additional interference effects between the system-reservoir interaction and the external driving field. When the system is fairly dense, the particle never gets away from the other particles in the system and we cannot really think of the particles as being "in between collisions." Quantum mechanically, the wave function of the particles are smeared out so that there is always some overlap of wave functions and as a result the particle retains some memory of the collisions it has experienced through its correlation with other particles in the system.[35] These memory effects are the characteristics of the quantum kinetic equations. Equations (54) and (55) can be used to describe the quantum transport phenomena and are the generalization of the Boltzmann equations. Equation (56) is the quantum kinetic equation for the interband process and is valid and useful to any time scale. The time-convolutionless nature of these equations will reduce the computation time required for the self-consistent numerical solution of the equations. In future work, we shall use these equations to describe the ultrafast nonlinear optical processes.

IV. SUMMARY

In this paper, we first derived a time-convolutionless equation of motion for a reduced density operator from a quantum Liouville equation for an arbitrary driven system interacting weakly with the stochastic reservoir. Secondly, time-convolutionless quantum kinetic equations for the system of interacting electron-hole pairs near the band edge in semiconductors were obtained from the equation of motion for the reduced density operator. Time-convolutionless quantum kinetic equations incorporate the non-Markovian relaxation (or dephasing) and the renormalized memory effect self-consistently and are in a form convenient for the perturbation expansions in powers of the system-reservoir interaction and the interaction with the external driving field. These equations are valid and useful for any time scale. It was shown that the interference of the driving field and the reservoir modulates the interband polarization and includes the renormalized memory effects.

ACKNOWLEDGMENTS

The author is indebted to Professor Mizuhiko Saeki for valuable comments and supplying Ref. 18. Shun-Lien Chuang contributed to this work actively in developing the density-matrix theory in second quantization. The author also thanks Shun-Lien Chuang for sharing his work prior to publication.

APPENDIX: OPERATOR ALGEBRA

In this section, we prove some useful functional relations among evolution operators (or propagators) defined in Sec. II.

Theorem 1. Let $\underline{V}(t)$ as the projected evolution operator of the system be defined as follows:

$$\underline{V}(t)=\underline{T}\exp\left\{-i\int_0^t d\tau\, QL_s(\tau)Q\right\} , \quad (A1)$$

then

$$\underline{V}(t)=\underline{P}+Q\underline{U}(t)Q . \quad (A2)$$

Proof. By expanding $\underline{V}(t)$ in the Taylor series, we obtain

$$\underline{V}(t)=1-i\int_0^t d\tau\, QL_s(\tau)Q+(-i)^2\int_0^t d\tau\int_0^\tau ds\, QL_s(\tau)QQL_s(s)Q+\cdots$$
$$=1+Q\left\{1-i\int_0^1 d\tau\, L_s(\tau)+(-i)^2\int_0^t d\tau\int_0^\tau ds\, L_s(\tau)L_s(s)+\cdots\right\}Q-Q$$
$$=\underline{P}+Q\underline{U}(t)Q\ .$$

We use the commutativity of Q and $L_s(t)$ and the indempotent property $Q^2=Q$.

Theorem 2. Projected propagators $\underline{H}(t,\tau)$ and $\underline{S}(t,\tau)$ satisfy

$$\underline{H}(t,\tau)=\underline{V}(t)\underline{S}(t,\tau)\underline{V}^{-1}(\tau)\ . \tag{A3}$$

Proof. We differentiate Eq. (8) with respect to obtain

$$\frac{d}{dt}\underline{H}(t,\tau)=-iQL_T(t)Q\underline{H}(t,\tau)$$

with the initial condition $\underline{H}(\tau,\tau)=1$.

On the other hand,

$$\frac{d}{dt}\underline{V}(t)\underline{S}(t,\tau)\underline{V}^{-1}(\tau)=-i\underline{Q}L_s(t)\underline{QV}(t)\underline{S}(t,\tau)\underline{V}^{-1}(\tau)+\underline{V}(t)\{-i\underline{QU}^{-1}(t)L_i(t)\underline{U}(t)\underline{Q}\}\underline{S}(t,\tau)\underline{V}^{-1}(\tau)$$
$$=-i\underline{Q}L_s(t)\underline{QV}(t)\underline{S}(t,\tau)\underline{V}^{-1}(\tau)+\{\underline{P}+\underline{QU}(t)\underline{Q}\}\{-i\underline{QU}^{-1}(t)L_i(t)\underline{U}(t)\underline{Q}\}\underline{S}(t,\tau)\underline{V}^{-1}(\tau)$$
$$=-i\underline{Q}L_s(t)\underline{QV}(t)\underline{S}(t,\tau)\underline{V}^{-1}(\tau)-i\underline{Q}L_i(t)\underline{QQU}(t)\underline{Q}S(t,\tau)\underline{V}^{-1}(\tau)$$
$$=-i\underline{Q}L_s(\tau)\underline{QV}(t)\underline{S}(t,\tau)\underline{V}^{-1}(\tau)-i\underline{Q}L_i(t)\underline{Q}\{\underline{P}+\underline{QU}(t)\underline{Q}\}\underline{S}(t,\tau)\underline{V}^{-1}(\tau)$$
$$=-i\underline{Q}L_T(t)\underline{QV}(t)\underline{S}(t,\tau)\underline{V}^{-1}(\tau)$$

with $\underline{V}(\tau)\underline{S}(\tau,\tau)\underline{V}^{-1}(\tau)=1$. Since any two functions which satisfy the identical differential equation and initial condition must be identical to each other, it is obvious that $\underline{H}(t,\tau)=\underline{V}(t)\underline{S}(t,\tau)\underline{V}^{-1}(\tau)$.

Lemma. It can be shown that

$$\underline{H}(t,\tau)\underline{Q}=\underline{U}(t)\underline{S}(t,\tau)\underline{U}^{-1}(\tau)\underline{Q}\ . \tag{A4}$$

Theorem 3. Evolution operators $\underline{G}(t,\tau)$ and $\underline{R}(t,\tau)$ satisfy

$$\underline{G}(t,\tau)=\underline{U}(\tau)\underline{R}(t,\tau)\underline{U}^{-1}(t)\ . \tag{A5}$$

Proof of this theorem is similar to that of Theorem 2.

Theorem 4. Let $\underline{U}_{\rm ext}(t)$ be the evolution operator of the system in the interaction picture such that

$$\underline{U}(t)=\underline{U}_0(t)\underline{U}_{\rm ext}(t)\ , \tag{A6}$$

where

$$\underline{U}_0(t)=\underline{T}\exp\left\{-i\int_0^t ds\, L_0(s)\right\}$$

is the unperturbed evolution operator of the system. Then

$$\underline{U}_{\rm ext}(t)=\underline{T}\exp\left\{-i\int_0^t ds\, \underline{U}_0^{-1}(s)L_{\rm ext}(s)\underline{U}_0(s)\right\}\ . \tag{A7}$$

Proof. We differentiate $\underline{U}(t)$ with respect to t in order to obtain

$$\frac{d}{dt}\underline{U}(t)=-iL_s(t)\underline{U}(t)$$
$$=\left\{\frac{d}{dt}\underline{U}_0(t)\right\}\underline{U}_{\rm ext}(t)+\underline{U}_0(t)\frac{d}{dt}\underline{U}_{\rm ext}(t)\ .$$

Then we have

$$\frac{d}{dt}\underline{U}_{\rm ext}(t)=-i\underline{U}_0^{-1}(t)L_{\rm ext}(t)\underline{U}_0(t)\underline{U}_{\rm ext}(t)\ . \tag{A8}$$

The formal solution of (A8) is

$$T\exp\left\{-i\int_0^t ds\, \underline{U}_0^{-1}(s)L_{\rm ext}(s)\underline{U}_0(s)\right\}\ .$$

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