Time-convolutionless reduced-density-operator theory of an arbitrary driven system coupled to a stochastic reservoir. II. Optical gain and line-shape function of a driven semiconductor

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In this paper, recently developed time-convolutionless quantum-kinetic equations for electron-hole pairs near the band edge are used to derive the optical gain and the line-shape function of a driven semiconductor taking into account excitonic effects. The equation of motion for the interband pair amplitude is integrated directly assuming the quasiequilibrium or adiabatic approximation. It is shown that the line shape of the optical-gain spectra is Gaussian for the simplest non-Markovian quantum kinetics. On the other hand, the line-shape function becomes Lorentzian, which has been assumed in most practical calculations, in the Markovian limit. It is also shown that the optical gain is enhanced by (1) excitonic effects caused by the attractive electron-hole Coulomb interaction and (2) interference effects (or renormalized memory effects) between the external driving field and the stochastic reservoir of the system. Gain enhancement by the memory effects can be interpreted as the result of the violation of energy conservation on the time scale shorter than the correlation time.

I. INTRODUCTION

This paper is a direct continuation of a preceding one[1] (I) in which the time-convolutionless (TCL) quantum-kinetic equations for interacting electron-hole pairs in band-edge semiconductors are derived from the equation of motion for the reduced density operator for an arbitrary driven system coupled to the stochastic reservoir. Time-convolutionless quantum-kinetic equations generalize the semiconductor Bloch equations to incorporate the non-Markovian relaxation and the renormalization of the memory effects through the interference between the external driving field and the stochastic reservoir of the system. The memory effects arise because the wave functions 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. These memory effects are the characteristics of the quantum-kinetic equations.[2–5] In quantum kinetics, a particle may possess a component of the wave function prior to the collision. As a result, the strict energy conservation may no longer hold for a time interval shorter than the correlation time.

From an application point of view, the optical gain is one of the most important basic properties of optoelectronic devices. For most practical calculations,[6–9] the phenomenological Lorentzian line-shape function for the optical gain, analogous to gas laser theories, are assumed. However, it was pointed out by Yamanish and Lee[10] that the optical spectra calculated with the Lorentzian line-shape function deviate from the experimental results. Especially, a gradual tailing and an unnatural absorption region appears at photon energies lower than the band gap in spontaneous emission and gain spectra, respectively, as long as the Lorentzian line-shape is used. Moreover, the gain coefficient may be underestimated and may sensitively depend on an upper limit in an integral over the electron energy due to the weak convergence of the Lorentzian function with increasing off resonance. Besides, the Urbach absorption tail universally observed in polar semiconductors cannot be explained by the Lorentzian line-shape function.[5] When these issues are set aside, there are many-body effects to be considered in the theoretical description of the optical grain.[11–14] Well-known effects are the reduction of the band gap with increasing carrier density (band-gap renormalization) and the enhancement of the optical transitions due to the attractive electron-hole interaction (Coulomb or excitonic enhancement). 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.[15] Many-body effects such as band-gap renormalization and Coulomb enhancement are not considered in their work.

In this paper, the optical gain and the line-shape function of a driven semiconductor are derived from the TCL quantum-kinetic equations for the interband polarization taking into account many-body effects. This paper can be regarded as an extension of Tomita and Suzuki's work[15] on the system of noninteracting electron-hole plasmas to the interacting electron-hole pairs in the band-edge semiconductors. The optical gain and the line-shape function are studied from first principles starting from the TCL quantum-kinetic equations obtained in I. We simplify the theory by considering the case of quasiequilibrium or adiabatic approximation only. Nonlinearities due to the Coulomb effects are considered in the present theoretical frame. We integrate the equation of motion for the interband pair amplitude directly to obtain the optical gain assuming the quasiequilibrium or adiabatic approximation and discuss the interference effects on the lineshape between the external driving field and the stochastic reservoir of the system.

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II. TIME-CONVOLUTIONLESS QUANTUM-KINETIC EQUATIONS FOR SEMICONDUCTORS

In this section, we summarize the results obtained in I on the time-convolutionless quantum-kinetic equations for the interacting electron-hole pairs in band-edge semiconductors with an external driving field. We consider an arbitrary driven system interacting with the stochastic reservoir and assume that the interaction of the system with its surroundings can be represented by the stochastic Hamiltonian.[16] The Hamiltonian of the total system is assumed to be

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

where $H_0(t)$ is the Hamiltonian of the system, $H_{\text{ext}}(t)$ 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 the stochastic Liouville equation,[16–21]

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

where $L_T(t)=L_0(t)+L_i(t)+L_{\text{ext}}(t)=L(t)+L_{\text{ext}}(t)=L_s(t)+L_i(t)$ is the Liouville superoperator in one-to-one correspondence with the Hamiltonian. In this paper, we use units where $\hbar=1$. The stochastic Hamiltonian $H_i(t)$ may include electron-electron interaction and electron-LO-phonon interaction for both conduction and valence electrons. 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.[22–28] The information of the system is then contained in the reduced density operator $\rho(t)$, which is defined by[16]

$$\rho(t)=\langle P\rho_T(t)\rangle_1 ,$$

where P is the projection operator.

As an initial condition, decoupling of the system and the reservoir for the total density operator is assumed. Perturbation expansions of the generalized collision operator are carried out in powers of the driving field with the lowest-order Born approximation for the interaction of the system with the reservoir.

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 pair amplitude induced by the optical field, are the matrix elements of the reduced density operator and are given by

$$\begin{aligned} n_{ck}(t) &= \rho_{cck}(t) \\ &= \langle ck|\rho(t)|ck\rangle , \end{aligned} \tag{3}$$

$$\begin{aligned} n_{vk}(t) &= \rho_{vvk}(t) \\ &= \langle vk|\rho(t)|vk\rangle , \end{aligned} \tag{4}$$

and

$$\begin{aligned} p_k^*(t) &= \rho_{vck}(t) \\ &= \langle vk|\rho(t)|ck\rangle , \end{aligned} \tag{5}$$

for the two-band model of a semiconductor.

The time-convolutionless quantum-kinetic equations for $n_{ck}(t)$, $n_{vk}(t)$, and $p_k^*(t)$ are (I)

$$\begin{aligned} \frac{\partial}{\partial t}n_{ck}(t) - 2\,\text{Im}&\left\{\left[\mu(k)E_p(t)+\sum_{k'}V(k-k')p_{k'}(t)\right]p_k^*(t)\right\} \\ &-2\int_0^t d\tau\,\text{Re}(\langle\langle ck|\{H_i(t)[\underline{U}_0(\tau)H_i(t-\tau)]\}|ck\rangle\rangle_i)\{n_{ck}(t)-(\rho_0^{(0)})_{cck}(t)\} , \end{aligned} \tag{6}$$

$$\begin{aligned} \frac{\partial}{\partial t}n_{vk}(t) = 2\,\text{Im}&\left\{\left[\mu(k)E_p(t)+\sum_{k'}V(k-k')p_{k'}(t)\right]p_k^*(t)\right\} \\ &-2\int_0^t d\tau\,\text{Re}\{\langle\langle vk|\{H_i(t)[\underline{U}_0(\tau)H_i(t-\tau)]\}|vk\rangle\rangle_i\}\{n_{vk}(t)-(\rho_0^{(0)})_{vvk}(t)\} . \end{aligned} \tag{7}$$

and

$$\begin{aligned} \frac{\partial}{\partial t}p_k^*(t) = {}& i[E_c(k)-E_v(k)]p_k^*(t)+i\left[\mu^*(k)E_p(t)+\sum_{k'}V(k-k')p_{k'}^*(t)\right][n_{ck}(t)-n_{vk}(t)] \\ &-\int_0^t d\tau(\langle\langle vk|\{H_i(t)[\underline{U}_0(\tau)H_i(t-\tau)]\}|vk\rangle\rangle_i \\ &\qquad+\langle\langle ck|\{[\underline{U}_0(\tau)H_i(t-\tau)]H_i(t)\}|ck\rangle\rangle_i)p_k^*(t) \\ &+i\int_0^t d\tau\int_0^\tau ds\,\exp\{-i[E_v(k)-E_c(k)]s\}(\langle\langle vk|\{H_i(t)[\underline{U}_0(\tau)H_i(t-\tau)]\}|vk\rangle\rangle_i \\ &\qquad\qquad+\langle\langle ck|\{(\underline{U}_0(\tau)H_i(t-\tau)]H_i(t)\}|ck\rangle\rangle_i) \\ &\qquad\times\mu^*(k)E_p(t-s)\{(\rho_0^{(0)})_{cck}(t)-(\rho_0^{(0)})_{vvk}(t)\} . \end{aligned} \tag{8}$$

Here, $E_c(k)$, $E_v(k)$ are renormalized single-particle energies given by

$$E_c(k)=E_c^0(k)-\sum_{k'} V(k-k')n_{ck'}^0 \quad (9)$$

and

$$E_v(k)=E_v^0(k)-\sum_{k'} V(k-k')n_{vk'}^0 \ , \quad (10)$$

and we include the band-gap renormalization that reduces the band gap with increasing carrier density.

The last term of (8), $\langle vk|D_1^{(2)}(t)|ck\rangle$ of (I), modulates the interband pair amplitude due to the interference of the driving optical field and the stochastic reservoir of the system and gives the renormalized memory effects. In other words, it describes the effects of the external driving field on the motion of a particle between collisions.

Equations (6)–(8) include the effects of the non-Markovian relaxation on the motion of particles between collisions. The interband kinetic-equation incorporates additional interference effects between the system-reservoir interaction and the external driving field.

III. TIME-CONVOLUTIONLESS INTERBAND QUANTUM KINETICS

In this section, we derive the optical gain and the line-shape function from the TCL quantum-kinetic equations. We simplify the theory by considering the case of quasiequilibrium and steady-state interband polarization only. Nonlinear effects caused by the population modulation, such as spectral hole burning, are ignored in this work. Spectral hole burning for noninteracting electron-hole pairs is studied briefly in the Appendix with the gain enhancement due to the renormalized memory effects taken into account. In order to obtain temporal dynamics of the interband polarization self-consistently, one needs to solve (6)–(8) numerically. We consider the system of interacting electron-hole pairs in semiconductors in the presence of coherent monochromatic radiation. The optical field $E_p(t)$ is given by

$$E_p(t)=E_p e^{-i\omega t}+E_p^* e^{i\omega t} \ . \quad (11)$$

In the rotating wave approximation, we rewrite the interband pair amplitude $p_k^*(t)$ as

$$p_k^*(t)=e^{i\omega t}\underline{p}_k^*(t) \ , \quad (12)$$

for electron-hole pairs in the band-edge semiconductors driven by the coherent optical field.

Substituting (12) into (8) and linearizing the equation of motion by assuming the quasiequilibrium, we get

$$\begin{aligned}\frac{\partial}{\partial t}\underline{p}_k^*(t)=&\,i\Delta_k\underline{p}_k^*(t)+i\left[\mu^*(k)E_p^*+\sum_{k'}V(k-k')\underline{p}_{k'}^*(t)\right][n_{ck}^0-n_{vk}^0]\\ &-\int_0^t d\tau(\langle\langle vk|\{H_i(t)[\underline{U}_0(\tau)H_i(t-\tau)]\}|vk\rangle\rangle_i+\langle\langle ck|\{[\underline{U}_0(\tau)H_i(t-\tau)]H_i(t)\}|ck\rangle\rangle_i)\underline{p}_k^*(t)\\ &+i\int_0^t d\tau\int_0^\tau ds\,\exp\{-i[E_v(k)-E_c(k)]s\}(\langle\langle vk|\{H_i(t)[\underline{U}_0(\tau)H_i(t-\tau)]\}|vk\rangle\rangle_i\\ &\qquad+\langle\langle ck|\{[\underline{U}_0(\tau)H_i(t-\tau)]H_i(t)\}|ck\rangle\rangle_i)\\ &\times\mu^*(k)E_p(t-s)[n_{ck}^0-n_{vk}^0] \ ,\end{aligned} \quad (13)$$

where $\Delta_k=E_c(k)-E_v(k)-\omega$ and n_{ck}^0 and n_{vk}^0 are the quasiequilibrium distribution of electrons in the conduction band and the valence band, respectively.

We assume that the interband pair amplitude follows the temporal variation of the polarization and the field amplitude adiabatically. We introduce functions $g_1(t)$ and $g_2(t,\Delta_k)$ as

$$g_1(t)=\int_0^t d\tau(\langle\langle vk|\{H_i(t)[\underline{U}_0(\tau)H_i(t-\tau)]\}|vk\rangle\rangle_i+\langle\langle ck|\{[\underline{U}_0(\tau)H_i(t-\tau)]H_i(t)\}|ck\rangle\rangle_i) \quad (14a)$$

and

$$g_2(t,\Delta_k)=\int_0^t d\tau\int_0^\tau ds\,\exp\{i\Delta_k s\}(\langle\langle vk|\{H_i(t)[\underline{U}_0(\tau)H_i(t-\tau)]\}|vk\rangle\rangle_i+\langle\langle ck|\{[\underline{U}_0(\tau)H_i(t-\tau)]H_i(t)\}|ck\rangle\rangle_i) \ . \quad (14b)$$

Equation (13) becomes

$$\left[\frac{\partial}{\partial t}-i\Delta_k+g_1(t)\right]\underline{p}_k^*(t)=i\left[\mu^*(k)E_p^*\{1+g_2(t,\Delta_k)\}+\sum_{k'}V(k-k')\underline{p}_{p'}^*(t)\right][n_{ck}^0-n_{vk}^0] \ . \quad (15)$$

Equation (15) can be integrated directly using the integrating factor $h(t)$, which is given by

$$h(t)=\exp\left[\int^t dt'\{-i\Delta_k+g_1(t')\}\right] \ ,$$

using the time-convolutionless nature of the equation.

The result is

$$\underline{p}_k^*(t)=i\int_0^t d\tau \exp\left\{-\int_\tau^t dt'[-i\Delta_k+g_1(t')]\right\}\left[\mu^*(k)E_p^*\{1+g_2(\tau,\Delta_k)\}+\sum_{k'}V(k-k')\underline{p}_{k'}^*(\tau)\right][n_{ck}^0-n_{vk}^0] . \quad (16)$$

Equation (16) is the generalized form of the optical dipole with phase damping.[10] Mathematical manipulations can be simplified considerably by taking the Laplace transformation of (16).

We define the following Laplace transformations:

$$\psi_k(s)=\mathcal{L}\{p_k^*(t)\} , \quad (17a)$$

$$\Xi(s,\Delta_k)=\mathcal{L}\left\{\int_0^t dt'[i\Delta_k-g_1(t')]\right\} , \quad (17b)$$

and

$$G_2(s,\Delta_k)=\mathcal{L}\{g_2(t,\Delta_k)\} , \quad (17c)$$

where $\mathcal{L}\{f(t)\}$ denotes the Laplace transformation of $f(t)$.

We obtain

$$\psi_k(s)=\psi_k^{(0)}(s)Q_k(s) , \quad (18)$$

where

$$\psi_k^{(0)}(s)=i\Xi(s,\Delta_k)\mu^*(k)E_p^*\left[\frac{1}{s}+G_2(s,\Delta_k)\right][n_{ck}^0-n_{vk}^0] , \quad (19)$$

and

$$Q_k(s)=1+\frac{s}{\mu^*(k)E_p^*[1+sG_2(s,\Delta_k)]}\times\sum_{k'}V(k-k')\psi_{k'}^{(0)}(s)Q_{k'}(s) . \quad (20)$$

The vertex function $Q_k(s)$ can be approximated by following Haug and Koch[13]

$$Q_k(s)=\frac{1}{1-q_{1k}(s)} , \quad (21)$$

where

$$q_{1k}(s)=\frac{s}{\mu^*(k)E_p^*[1+sG_2(s,\Delta_k)]}\sum_{k'}V(k-k')\psi_{k'}^{(0)}(s) \quad (22)$$

in the simplest Pade' approximation.

The factor $Q_k(s)$ describes the excitonic enhancement of the interband polarization. The steady-state interband pair amplitude is determined by

$$\begin{aligned}\underline{p}_k^*(\infty)&=\lim_{s\to 0}s\psi_k(s)\\&=\lim_{s\to 0}\frac{s\psi_k^{(0)}(s)}{1-q_{1k}(s)}\\&=i\frac{\Xi(0,\Delta_k)}{1-q_{1k}(0)}\mu^*(k)E_p^*\\&\quad\times[1+g_2(\infty,\Delta_k)][n_{ck}^0-n_{vk}^0] .\end{aligned} \quad (23)$$

The interband polarization P and the susceptibility χ can be expressed through the dipole operator and the interband pair amplitude as

$$P=\frac{1}{V}\mathrm{Tr}\{\mu(k)\underline{p}_k^*(\infty)\} \quad (24)$$

or

$$\varepsilon_0\chi(\omega)=\frac{1}{V}\mathrm{Tr}\left\{i\frac{\Xi(0,\Delta_k)}{1-q_{1k}(0)}|\mu(k)|^2\times[1+g_2(\infty,\Delta_k)][n_{ck}^0-n_{vk}^0]\right\} . \quad (25)$$

The optical gain $g(\omega)$ is[9]

$$\begin{aligned}g(\omega)&=\frac{\omega\mu c}{n_r}\mathrm{Im}\varepsilon_0\chi(\omega)\\&=\frac{\omega\mu c}{n_r}\frac{2}{V}\sum_k\frac{\mathrm{Re}\Xi(0,\Delta_k)}{1-\mathrm{Re}q_{1k}(0)}\\&\quad\times|\mu(k)|^2[1+\mathrm{Re}g_2(\infty,\Delta_k)][n_{ck}^0-n_{vk}^0] ,\end{aligned} \quad (26)$$

with

$$\mathrm{Re}q_{1k}(0)\approx\sum_{k'}V(k-k')\mathrm{Re}\Xi(0,\Delta_{k'})[n_{ck'}^0-n_{vk'}^0] , \quad (27)$$

where μ is the permeability, n_r is the refractive index, c is the speed of light in free space, V is the volume, Tr denotes the trace, and ε_0 is the permittivity of free space. $\chi(\omega)$ is the Fourier component of $\chi(t)$ with $e^{i\omega t}$ dependence. In Eq. (26), $\mathrm{Re}\Xi(0,\Delta_k)$ is the line-shape function that describes the spectral shape of the optical gain in a driven semiconductor. It will be shown that the line-shape function becomes Gaussian for the simplest non-Markovian relaxation and Lorentzian for Markovian relaxation later in this section.

The energy difference $E_c(k)-E_v(k)$ between the electrons in the conduction band and the valence band contains the Coulomb effects which account for the band-gap renormalization. From

$$E_c(k)-E_v(k)=\mathrm{E}_g+\frac{k^2}{2\mathrm{m}}-\sum_{k'}V(k-k')(n_{ck'}^0+1-n_{vk'}^0) , \quad (28)$$

renormalized band-gap energy near the band edge is

$$\begin{aligned}\Delta\mathrm{E}_g&=-\sum_{k'}V(k-k')(n_{ck'}^0+1-n_{vk'}^0)\\&=-\int\frac{d^3k'}{(2\pi)^3}\frac{e^2}{\varepsilon_s}\frac{n_{ck'}^0+1-n_{vk'}^0}{k^2+k'^2-2kk'\cos\theta}\\&=-\int_0^\infty\frac{dk'e^2}{(2\pi)^2\varepsilon_s}\frac{k'}{k}\log\left|\frac{k+k'}{k-k'}\right|(n_{ck'}^0+1-n_{vk'}^0)\\&\approx-\int_0^\infty\frac{dk'e^2}{2\pi^2\epsilon_s}(n_{ck'}^0+1-n_{vk'}^0)\ \text{as}\ k\to 0 ,\end{aligned} \quad (29)$$

where we made use of the limit $(1/x)\log|(1+x)/(1-x)|\rightarrow 2$ as $x\rightarrow 0$. This explains some of recent results which show that the peak gain of a laser at high injection level occurs at a lower-energy level than the spontaneous emission-peak energy, a phenomenon called band-gap renormalization, which arises from the interband correlation due to the Coulomb interaction.

Excitonic effects enhance the optical gain by the Coulomb enhancement factor $Q_k(0)$:

$$Q_k(0)=\frac{1}{1-\int \frac{d^3k' e^2 n^0_{ck'}-n^0_{vk'}}{(2\pi)^3\varepsilon_s k^2+k'^2-2kk'\cos\theta}\mathrm{Re}\Xi(0,\Delta_{k'})}, \tag{30}$$

where m is the reduced mass and ε_s is the static dielectric constant.

Recently, Haug and Koch[13] showed that at room temperature the excitonic enhancement can be neglected for carrier densities above the Mott density but one needs to keep the density-dependent band-gap renormalization. On the other hand, excitonic enhancement has noticeable contribution if the carrier density approaches the Mott density.

The factor $[1+\mathrm{Re}g_2(\infty,\Delta_k)]$ in (26) describes the gain (or line-shape) enhancement due to the interaction between the optical field and the stochastic reservoir of the system. This enhancement of gain is due to the absence of strict energy conservation in the non-Markovian quantum kinetics. It can be shown that $\mathrm{Re}g_2(\infty,\Delta_k)$ vanishes in the Markovian limit. The optical-gain (or line-shape) enhancement by the interference between the optical field and the reservoir is predicted for the first time, to the best of the author's knowledge, in this work for a semiconductor driven by a laser.

We now turn our attention to the study of a line-shape function and its enhancement for the cases of Markovian and non-Markovian relaxations.

A. Markovian limit

In order to analyze the collision term and the interference term in the Markovian limit, we put

$$\langle\langle \alpha k|\{H_i(t)[\underline{U}_0(\tau)H_i(t-\tau)]\}|\alpha k\rangle\rangle_i=\frac{1}{2\tau_\alpha(k)}\delta(|\tau|). \tag{31}$$

Then the dephasing term $g_1(t)$ and the interference term $g_2(t,\Delta_k)$ become

$$g_1(t)=\gamma_{vc}(k)=\gamma_{cv}(k)=\frac{1}{2}\left\{\frac{1}{\tau_v(k)}+\frac{1}{\tau_c(k)}\right\}, \tag{32}$$

and

$$g_2(t,\Delta_k)=0.$$

In the Markovian limit, the interference term $g_2(t,\Delta_k)$ vanishes because in the integration over ds, the upper limit τ is zero and the resulting integral vanishes because of (31). The calculation of the line-shape function is straightforward and is given by the Lorentzian

$$\mathrm{Re}\Xi(0,\Delta_k)=\frac{\gamma_{cv}(k)}{\Delta_k^2+\gamma_{cv}^2(k)}, \tag{33}$$

which is used in most calculations.

B. Simple non-Markovian relaxation

We assume the simplest form of the non-Markovian correlation function[29]

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

as

$$\langle\langle \alpha k|\{H_i(t)[\underline{U}_0(\tau)H_i(t-\tau)]\}|\alpha k\rangle\rangle_i=\frac{1}{2\tau_c\tau_\alpha(k)}\exp\left[-\frac{|\tau|}{\tau_c}\right], \tag{34}$$

where τ_c is the correlation time for the intraband relaxation.

The dephasing term $g_1(t)$ becomes

$$g_1(t)=\frac{\gamma_{cv}(k)}{\tau_c}\int_0^t dt'\exp\left[\frac{t'}{\tau_c}\right]=\gamma_{cv}(k)\left[1-\exp\left[-\frac{t}{\tau_c}\right]\right].$$

We obtain

$$\int_0^t dt' g_1(t')=\gamma_{cv}(k)\left[t+\tau_c\exp\left[-\frac{t}{\tau_c}\right]-\tau_c\right] \tag{35}$$

and

$$\Xi(0,\Delta_k)=\int_0^\infty dt\exp\left\{i\Delta_k t-\gamma_{cv}(k)\times\left[t+\tau_c\exp\left[-\frac{t}{\tau_c}\right]-\tau_c\right]\right\}=\tau_c I_0[-i\Delta_k\tau_c,\gamma_{cv}(k)\tau_c], \tag{36}$$

where

$$I_0(A,B)=\int_0^\infty dt\exp\{-At-B[t+\exp(-t)-1]\}. \tag{37}$$

In general, $I_0(A,B)$ is evaluated by the continued fraction representation.[30] If we expand the argument of the exponential function in (37) up to the second order in t, we get the Gaussian line-shape function:

$$\mathrm{Re}\Xi(0,\Delta_k)=\left[\frac{\tau_c\pi}{2\gamma_{cv}(k)}\right]^{1/2}\exp\left[-\frac{\tau_c\Delta_k^2}{2\gamma_{cv}(k)}\right]. \tag{38}$$

We do not specify the explicit form of $\gamma_{cv}(k)$, in this paper, and the recent calculations[31,32] of the intraband relaxations by several authors can be used in Eq. (38).

The line-shape enhancement (or gain enhancement) due to the interference between the driving field and the stochastic reservoir is described by

$$g_2(\infty,\Delta_k)=\int_0^t d\tau\int_0^\tau ds\,\frac{\gamma_{cv}(k)}{\tau_c}\exp\left[i\Delta_k s-\frac{|\tau|}{\tau_c}\right]$$

$$=\frac{\gamma_{cv}(k)\tau_c}{1-i\Delta_k\tau_c}\ , \tag{39}$$

or

$$\mathrm{Re}\,g_2(\infty,\Delta_k)=\frac{\gamma_{cv}(k)\tau_c}{1+\Delta_k^2\tau_c^2}\ . \tag{40}$$

The gain (or line-shape) enhancement factor at the resonance is

$$1+\mathrm{Re}\,g_2(\infty,\Delta_k)=1+\gamma_{cv}(k)\tau_c\ . \tag{41}$$

The enhancement of gain (or line shape) (41) is due to the interference between the external driving field and the stochastic reservoir of the system and may be caused by the absence of a strict energy conservation in the non-Markovian quantum-kinetic domain. Quantum mechanically, $\langle\langle \alpha k|[H_i(t)[\underline{U}_0(\tau)H_i(t-\tau)]|\alpha k\rangle\rangle_i$ 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 $|\alpha k\rangle$, got scattered at $t-\tau$ by $H_i(t-\tau)$, and went as a free particle for the time interval τ. In the non-Markovian kinetics, the memory effects extend over the time interval τ_c and yield the nontrivial probability amplitude for t in the interval.

Equation (41) can also be deduced from physical intuition only. It is well known that the optical transition satisfies $\Delta k=0$ selection rule for electrons in the conduction and valence bands. Assume an electron at $k=0$ in the conduction band was scattered at $t-\tau$ by the reservoir. For t in the time interval τ_c, an electron still may retain its previous wave function corresponding to the state $k=0$ as a partial wave which decays exponentially with t. When there is an incident photon at $t\ll\tau_c$ to this system, there will be a nonzero probability of transition for this electron to the state $k=0$ in the valence band. As a result, the stimulated emission probability for the state $k=0$ is enhanced by the presence of the memory effect. The probability of the occurrence of enhanced transition is proportional to the number of nonrandomizing scattering events per second, $\gamma_{cv}(k)$, times the time interval τ_c in which the memory effects extend. Equation (41) follows.

Tomita and Suzuki[15] estimated the correlation time τ_c for the non-Markovian relaxation to be on the order of 10 fs using the uncertainty principle. In comparison, typical intraband relaxation time is on the order of 100 fs. The non-Markovian enhancement of optical gain becomes significant as the correlation time increases. For example, when τ_c is 50 fs, the optical gain is predicted to be enhanced by as much as 50%.

In this section, it is shown that the direct integration of the equation of motion for the interband pair amplitude is possible because of the time-convolutionless nature of the equation. In addition, it is shown that the line-shape function is Gaussian and is enhanced at the resonance due to the interference between the driving field and the stochastic reservoir of the system.

IV. SUMMARY

In this paper, recently developed time-convolutionless quantum-kinetic equations for electron-hole pairs near the band edge are used to derive the optical-gain and the line-shape function of a driven semiconductor taking into account the excitonic effects. The equation of motion for the interband pair amplitude is integrated directly assuming the quasiequilibrium or adiabatic approximation. It is shown that the simplest non-Markovian quantum kinetics yields the optical gain with Gaussian line-shape function. On the other hand, the line-shape function becomes Lorentzian, which has been assumed in most practical calculations, in the Markovian limit. It is shown that the optical gain is enhanced by (1) the excitonic effects caused by the attractive electron-hole Coulomb interaction and (2) the interference effects (or renormalized memory effects) between the external driving field and the stochastic reservoir of the system. The enhancement of optical gain by the latter process is caused by the absence of strict energy conservation in the non-Markovian quantum kinetics.

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APPENDIX: SPECTRAL HOLE BURNING OF NONINTERACTING ELECTRON-HOLE PAIRS

In this appendix, a closed expression of the nonlinear gain for noninteracting electron-hole pairs in a driven semiconductor is studied briefly taking into account spectral hole burning. We extend Tomita and Suzuki's work[15] on the gain and spectral hole burning in semiconductors by including the gain enhancement due to the renormalized memory effects. We turn off the Coulomb potential $V(k-k')$ in Eqs. (6)–(8) to obtain closed expressions.

The time-convolutionless quantum-kinetic equations for $n_{ck}(t)$, $n_{vk}(t)$, and $\underline{p}_k^*(t)$ are in the rotating-wave approximation:

$$\frac{\partial}{\partial t}n_{ck}(t)=-2\,\mathrm{Im}\{\mu(k)E_p\underline{p}_k^*(t)\}-g_c(t)[n_{ck}(t)-n_{ck}^0]\ , \tag{A1}$$

$$\frac{\partial}{\partial t}n_{vk}(t)=2\,\mathrm{Im}\{\mu(k)E_p\underline{p}_k^*(t)\}-g_v(t)[n_{vk}(t)-n_{vk}^0]\ , \tag{A2}$$

and

$$\frac{\partial}{\partial t}\underline{p}_k^*(t)=i\Delta_k\underline{p}_k^*(t)+i\mu^*(k)E_p^*[n_{ck}(t)-n_{vk}(t)]-g_1(t)\underline{p}_k^*(t)+i\mu^*(k)E_p^*g_2(t,\Delta_k)[n_{ck}^0-n_{vk}^0] , \tag{A3}$$

where

$$g_c(t)=2\int_0^t d\tau\, \mathrm{Re}(\langle\langle ck|\{H_i(t)[\underline{U}_0(\tau)H_i(t-\tau)]\}|ck\rangle\rangle_i) , \tag{A4}$$

and

$$g_v(t)=2\int_0^t d\tau\, \mathrm{Re}(\langle\langle vk|\{H_i(t)[\underline{U}_0(\tau)H_i(t-\tau)]\}|vk\rangle\rangle_i) . \tag{A5}$$

Equations (A1)–(A3) can be integrated directly using the integrating factors and can be solved self-consistently using the Laplace transformation.

We define the following Laplace transformations:

$$N_{ck}(s)=\mathcal{L}\{n_{ck}(t)\} , \tag{A6}$$

$$N_{vk}(s)=\mathcal{L}\{n_{vk}(t)\} , \tag{A7}$$

$$\Xi_c(x)=\mathcal{L}\{-\int_0^t dt'(g_c(t'))\} , \tag{A8}$$

and

$$\Xi_v(s)=\mathcal{L}\{-\int_0^t dt'(g_v(t'))\} . \tag{A9}$$

After some mathematical manipulations, we obtain

$$N_{ck}(s)=-2\Xi_c(s)|\mu(k)|^2|E_p|^2\mathrm{Re}\Xi(s,\Delta_k)\{N_{ck}(s)-N_{ck}(s)+\mathrm{Re}G_2(s,\Delta_k)[n_{ck}^0-n_{vk}^0]\}+\left[\frac{1}{s}-\Xi_c(s)\right]n_{ck}^0+\frac{1}{s}n_{ck}^0 , \tag{A10}$$

$$N_{vk}(s)=2\Xi_v(s)|\mu(k)|^2|E_p|^2\mathrm{Re}\Xi(s,\Delta_k)\{N_{ck}(s)-N_{ck}(s)+\mathrm{Re}G_2(s,\Delta_k)[n_{ck}^0-n_{vk}^0]\}+\left[\frac{1}{s}-\Xi_v(s)\right]n_{vk}^0+\frac{1}{s}n_{vk}^0 , \tag{A11}$$

and

$$\psi_k(s)=\frac{i\Xi(s,\Delta_k)\mu^*(k)E_p^*\left[\frac{1}{s}+G_2(s,\Delta_k)\right][n_{ck}^0-n_{vk}^0]}{1+2|\mu(k)|^2|E_p|^2[\Xi_c(s)+\Xi_v(s)]\mathrm{Re}\Xi(s,\Delta_k)} . \tag{A12}$$

The steady-state interband pair amplitude is determined by

$$\begin{aligned}\underline{p}_k^*(\infty)&=\lim_{s\to 0} s\psi_k(s)\\&=\frac{i\Xi(0,\Delta_k)\mu^*(k)E_p^*[1+g_2(\infty,\Delta_k)][n_{ck}^0-n_{vk}^0]}{1+2|\mu(k)|^2|E_p|^2[\Xi_c(0)+\Xi_v(0)]\mathrm{Re}\Xi(0,\Delta_k)} .\end{aligned} \tag{A13}$$

The interband polarization P and the susceptibility χ can be expressed through the dipole operator and the interband pair amplitude as

$$P=\frac{1}{V}\mathrm{Tr}\{\mu(k)\underline{p}_k^*(\infty)\} , \tag{A14}$$

or

$$\varepsilon_0\chi(\omega)=\frac{1}{V}\mathrm{Tr}\left\{\frac{i\Xi(0,\Delta_k)|\mu(k)|^2[1+g_2(\infty,\Delta_k)][n_{ck}^0-n_{vk}^0]}{1+2|\mu(k)|^2|E_p|^2[\Xi_c(0)+\Xi_v(0)]\mathrm{Re}\Xi(0,\Delta_k)}\right\} . \tag{A15}$$

The optical gain $g(\omega)$ is

$$\begin{aligned}g(\omega)&=\frac{\omega\mu_c}{n_r}\mathrm{Im}\varepsilon_0\chi(\omega)\\&=\frac{\omega\mu c}{n_r}\frac{2}{V}\sum_k\frac{\mathrm{Re}\Xi(0,\Delta_k)|\mu(k)|^2[1+\mathrm{Re}g_2(\infty,\Delta_k)][n_{ck}^0-n_{vk}^0]}{1+2|\mu(k)|^2|E_p|^2[\Xi_c(0)+\Xi_v(0)]\mathrm{Re}\Xi(0,\Delta_k)}\end{aligned} \tag{A16}$$

Equation (A16) is the nonlinear gain for the noninteracting electron-hole pairs in a band-edge semiconductor driven by a monochromatic laser field. The denominator is the gain suppression due to the spectral hole burning. $\mathrm{Re}\Xi(0,\Delta_k)$ is the line-shape function that describes the spectral shape of optical-gain and spectral hole burning. The factor $\Xi_c(0)+\Xi_v(0)$ is responsible for the population beating through the relaxation processes. The factor $[1+\mathrm{Re}g_2(\infty,\Delta_k)]$ in (A16) describes the gain enhancement due to the interaction between the optical field and the stochastic reservoir of the system. This enhancement of gain is due to the absence of strict energy conservation in the non-Markovian quantum kinetics. The gain enhancement affects the linear gain only. Spectral hole burning is not affected by the interference effects.

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