The theory of non-Markovian optical gain in excited semiconductors

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A reduced description of the dynamics of carriers in excited semiconductors is presented. Firstly, a time-convolutionless equation of motion for the reduced density operator is derived from the microscopic Liouville equation for a driven system coupled to the stochastic reservoir by applying a projection operator method. Secondly, the quantum kinetic equations for interacting electron-hole pairs near band-edge in semiconductors under an external optical field are obtained from the equation of motion for the reduced density operator. The non-Markovian optical gain of a driven semiconductor is derived including the many-body effects. Plasma screening and excitonic effects are taken into account using an effective Hamiltonian in the time-dependent Hartree-Fock approximation. It is shown that the line shape of optical-gain spectra is Gaussian for the simplest non-Markovian quantum kinetics, and the optical gain is enhanced by the excitonic effects caused by the attractive electron-hole Coulomb interaction and the interference effects (renormalized memory effects) between the external driving field and the stochastic reservoir of the system.

I. INTRODUCTION

From a device fundamental point of view, the optical gain is one of the most important basic properties of the semiconductor lasers. For most practical calculations [1-4], phenomenological Lorentzian line shape function for the optical gain, analogous to gas laser theories, are assumed. However, it was pointed out [5-7] that the optical spectra calculated with the Lorentzian line shape function deviate considerably from the experimental results, especially, below the band gap region. Besides, the many-body effects need to be considered in the theoretical description of the optical gain [8-11]. 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).

On the other hand, advances in the theory of ultrafast optics [12-15] revealed non-Markovian relaxation kinetics of the electrons and holes in driven semiconductors. In these nonequilibrium kinetics, the system has the memory effects on a very short time scale and the equations of the motion for the system have time-convolution forms of integral kernels which are responsible for the memory effects. In previous work [16], the author extended the work [17] of Saeki on stochastic Liouville equation for a weakly driven system to derive a time-convolutionless (TCL) equation for a reduced density operator of an arbitrary driven system coupled to the stochastic reservoir and obtained time-convolutionless quantum kinetic equations and unscreened optical dielectric functions for the system of interacting electron-hole pairs under arbitrary external optical field. These equations generalize of the semiconductor Bloch equations [18-20] 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. Tomita and Suzuki studied the density matrix theory of optical gain for non-interacting electron-hole pairs in semiconductors including non-Markovian intraband relaxation and showed that the non-Markovian relaxation enhances both linear and nonlinear gain [21]. Many-body effects such as band-gap renormalization and Coulomb enhancement were not considered in their work. Here, we obtain non-Markovian optical gain and lineshape function of a semiconductor laser by including the many-body and the renormalized memory effects from the TCL quantum kinetic equations for the driven semiconductors. Plasma screening and excitonic effects are taken into account using an effective Hamiltonian in the time-dependent Hartree-Fock approximation. Nonlinearities due to the Coulomb effects are considered in the present theoretical frame.

II. THEORY

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. The Hamiltonian of the total system is assumed to be

$$\begin{aligned} H_T(t) &= H_0(t) + H_1(t) + H_{ext}(t) \\ &= H(t) + H_{ext}(t) \\ &= H_s(t) + H_1(t), \end{aligned} \qquad (1)$$

where $H_0(t)$ is the Hamiltonian of the system, $H_{ext}(t)$ the interaction of the system with the external driving field, and $H_1(t)$ 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

$$\frac{d\rho_T(t)}{dt} = -i\,[\,H_T(t),\, \rho_T(t)\,]$$

$$= -i\, L_T(t)\, \rho_T(t), \qquad (2)$$

where $L_T(t) = L_0(t) + L_1(t) + L_{ext}(t) = L(t) + L_{ext}(t) = L_S(t) + L_1(t)$, is the Liouville super operator in one-to-one correspondence with the Hamiltonian. In this paper, we use unit where $\hbar = 1$. The stochastic Hamiltonian $H_1(t)$ may include the electron-electron interaction and electron-LO phonon interaction for both conduction and valence electrons which would be responsible for the intraband relaxation or optical dephasing appears as correlation functions of $H_1(t)$. 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 [18,19]. Plasma screening can be taken into account by using an effective Hamiltonian in the time-dependent Hartree-Fock approximation. The information of the system is then contained in the reduced density operator obtained by eliminating the dynamical variables of the reservoir from the total density operator using projection operator [16, 17]. The screening can be described in a self-consistent way [20] by replacing the unscreened potential V(k) by a screened one, $V_s(k)$ which has a reduced interaction strength at the long distance and single particle energies $E_c(k)$, $E_v(k)$ by $E_c^{sc}(k)$, $E_v^{sc}(k)$. The replacement must be done in the original Hamiltonian for the system H_0 in eq. (1). In the following, we include the screening in the kinetic equations by replacing $V(k) \rightarrow V_s(k)$, and $E_c(k)$, $E_v(k) \rightarrow E_c^{sc}(k)$, $E_v^{sc}(k)$.

The non diagonal interband matrix element $p_k^*(t)$ which describes the interband pair amplitude induced by the optical field, is the matrix elements of the reduced density operator and satisfy the following time-convolutionless quantum kinetic equations [22]:

$$\frac{\partial}{\partial t} p_k^*(t) = i\,[E_c^{sc}(k) - E_v^{sc}(k)]\, p_k^*(t)$$

$$+\, i\,[\,\mu^*(k)E_p(t) + \sum_{k'} V_s(k-k')\, p_{k'}^*(t)]\,[\,n_{ck}(t) - n_{vk}(t)]$$

$$-\int_0^t d\tau\, \{ \langle\langle vk \mid [\, H_1(t)\,(\underline{U}_0(\tau) H_1(t-\tau))] \mid vk \rangle\rangle_1$$

$$+ \langle\langle ck \mid [\,(\underline{U}_0(\tau) H_1(t-\tau)) H_1(t)] \mid ck \rangle\rangle_1 \}\; p_k^*(t)$$

$$+ i \int_0^t d\tau \int_0^\tau ds \exp\{-i[E_v^{sc}(k) - E_c^{sc}(k)]s\} \{ \langle\langle vk | [H_1(t)(U_0(\tau)H_1(t-\tau))] | vk \rangle\rangle_1$$

$$+ \langle\langle ck | [(U_0(\tau)H_1(t-\tau)) H_1(t)] | ck \rangle\rangle_1 \}$$

$$\times \mu^*(k) E_p(t-s) \{ (\rho_0^{(0)})_{cck}(t) - (\rho_0^{(0)})_{vvk}(t) \}, \quad (3)$$

where $V_s(k)$ is the screened Coulomb potential, $U_0(t) = T \exp\{-i\int_0^t ds\ L_0(s)\}$ is the unperturbed evolution operator of the system with the time-ordering operator T, $\mu(k)$ is the dipole moment, $\rho_0^{(0)}(t) = U_0(t)\ \rho(0)$, $\rho(0)$ is the initial condition for the reduced density operator, and $\langle \cdots\cdots \rangle_1$ is the average over the stochastic process $H_1(t)$. The Coulomb term $\sum_{k'} V_s\ (k-k')\ p_{k'}(t)$ is responsible for the excitonic effects. Here $E_c^{sc}(k)$, $E_v^{sc}(k)$ are renormalized single particle energies given by

$$E_c^{sc}(k) = E_c^0(k) - \sum_{k'} V_s(k-k')\, n_{ck'}^0 ,$$

and $$E_v^{sc}(k) = E_v^0(k) + \sum_q [V_s(q) - V(q)] - \sum_{k'} V_s(k-k')\, n_{vk'}^0,$$

where the term $\sum_q [V_s(q) - V(q)]$ is the Coulomb-hole part of the single-particle energy renormalization which is the change of electrostatic energy (or self-energy) due to the presence of the electron-hole plasma. Here, we employed the two-band model for the semiconductor and introduce two short-handed notations $|ck\rangle$ and $|vk\rangle$ such that

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

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

The last term of (3) 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 word, it describes the effects of the external driving field on the motion of a particle between collisions.

Now, we derive the optical gain and the line shape function from the TCL quantum kinetic equations. We simplify the theory considerably by considering the case of quasi-equilibrium and steady state interband polarization only. Nonlinear effects caused by the population modulation such as spectral hole burning are ignored in this work. 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

$$E_p(t) = E_p e^{-i\omega t} + E_p^* e^{i\omega t} . \tag{4}$$

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

$$p_k^*(t) = e^{i\omega t} \underline{p}_k^*(t) , \tag{5}$$

for electron-hole pairs in the band-edge semiconductors driven by the coherent light field. We introduce functions $g_1(t)$ and $g_2(t,\Delta_k)$ as

$$g_1(t) = \int_0^t d\tau \{ \langle\langle vk \,|\, [H_1(t) (\underline{U}_0(\tau) H_1(t-\tau))] \,|\, vk \rangle\rangle_1 + \langle\langle ck \,|\, [(\underline{U}_0(\tau) H_1(t-\tau)) H_1(t)] \,|\, ck \rangle\rangle_1 \}, \tag{6a}$$

and

$$g_2(t,\Delta_k) = \int_0^t d\tau \int_0^\tau ds \exp\{i \Delta_k s\} \{ \langle\langle vk \,|\, [H_1(t) (\underline{U}_0(\tau) H_1(t-\tau))] \,|\, vk \rangle\rangle_1 + \langle\langle ck \,|\, [(\underline{U}_0(\tau) H_1(t-\tau)) H_1(t)] \,|\, ck \rangle\rangle_1 \}. \tag{6b}$$

Here $\Delta_k = E_c^{sc}(k) - E_v^{sc}(k) - \omega$. Then

$$\underline{p}_k^*(t) = i \int_0^t d\tau \exp[- \int_\tau^t dt' (- i \Delta_k + g_1(t'))] \times [\mu^*(k) E_p^* \{1+g_2(\tau,\Delta_k)\} + \sum_{k'} V_s(k-k') \underline{p}_{k'}^*(\tau)] [n_{ck}^0 - n_{vk}^0]. \tag{7}$$

To derive Eq. (7) from Eq. (3), we assumed that the interband pair amplitude follows the temporal variation of the polarization and the field amplitude adiabatically and suppressed the dynamics of the particle distributions (or assumed the particle distribution is determined mostly by injection) by replacing $n_{\alpha k}(t) \to n_{\alpha k}^0$ where n_{ck}^0 and n_{vk}^0 are the quasi-equilibrium distribution of electrons in the conduction band and the valence band, respectively..

Equation (7) is the generalized form of the optical dipole with phase-damping[5]. Mathematical manipulations can be simplified considerably by taking the Laplace transformation of (7). In Eqs. (6)-(7), $g_1(t)$ is the optical dephasing which is the temporal decay of the interband polarization. We define the following Laplace transformations:

$$\psi_k(s) \quad = \mathcal{L}\{ \, \underline{p}_k^*(t) \, \}, \tag{8a}$$

$$\Xi\,(s, \Delta_k) \;=\; \mathcal{L}\{ \int_0^t dt' \, (\, i\, \Delta_k - g_1(t')\,)\}, \tag{8b}$$

and $$G_2(s, \Delta_k) \;=\; \mathcal{L}\{ \, g_2(t,\Delta_k)\} \tag{8c}$$

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

The steady state interband pair amplitude is determined by

$$\begin{aligned} \underline{p}_k^*(\infty) \;&=\; \lim_{s\to 0} \, s\,\psi_k(s) \\ &= \lim_{s\to 0} \frac{s\psi_k^{(o)}(s)}{1 - q_{1k}(s)} \\ &= i\,\frac{\Xi\,(0,\,\Delta_k)}{1 - q_{1k}(0)}\; \mu^*(k)E_p^*\,[\,1 \;+\; g_2(\infty,\Delta_k)\,][\,n_{ck}^{o} - n_{vk}^{o}\,], \end{aligned} \tag{9}$$

after some mathematical manipulations

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

$$P \;=\; \frac{1}{V}\,\mathrm{Tr}\,\{\;\mu(k)\,\underline{p}_k^*(\infty)\;\} \tag{10}$$

or $$\varepsilon_0\,\chi\,(\omega) = \frac{1}{V}\,\mathrm{Tr}\left\{ i\,\frac{\Xi\,(0,\Delta_k)}{1 - q_{1k}(0)}\; |\mu(k)|^2\;[1 \,+\, g_2(\infty,\Delta_k)][n_{ck}^{o} - n_{vk}^{o}] \right\}. \tag{11}$$

The optical gain $g(\omega)$ is [4]

$$g(\omega) = \frac{\omega\mu c}{n_r} \operatorname{Im} \varepsilon_0 \chi(\omega)$$

$$= \frac{\omega\mu c}{n_r} \frac{2}{V} \sum_k \frac{\operatorname{Re} \Xi\,(0,\Delta_k)}{1-\operatorname{Re} q_{1k}(0)} |\mu(k)|^2 [1 + \operatorname{Re} g_2(\infty,\Delta_k)][n_{ck}^0 - n_{vk}^0] \tag{12}$$

with $$\operatorname{Re} q_{1k}(0) \approx \sum_{k'} V_s(k-k')\operatorname{Re} \Xi\,(0,\Delta_{k'})[n_{ck'}^0 - n_{vk'}^0] \tag{13}$$

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 equation (12), $\operatorname{Re} \Xi\,(0,\Delta_k)$ 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 in later this section. The denominator [1- $\operatorname{Re} q_{1k}(0)$] describes the gain enhancement due to the excitonic effects caused by the attractive Coulomb interaction

The factor (1 + $\operatorname{Re} g_2(\infty,\Delta_k)$) in (12) 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 a strict energy conservation in the non-Markovian quantum kinetics. It can be shown that $\operatorname{Re} g_2(\infty,\Delta_k)$ 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 in this work to the best of the author's knowledge.

We now turn out attention to the study of line shape function and its enhancement for the case of non-Markovian relaxation. We assume the simplest form of the non-Markovian correlation function [23] $\langle\langle \alpha k \,|\, [\, \underline{H}_1(t)\, (\underline{U}_0(\tau)\underline{H}_1(t-\tau))] \,|\, \alpha k \rangle\rangle_i$ as

$$\langle\langle \alpha k \,|\, [\, \underline{H}_1(t)\, (\underline{U}_0(\tau)\underline{H}_1(t-\tau)) \,|\, \alpha k \rangle\rangle_i$$

$$= \frac{1}{2\tau_c\tau_\alpha(k)} \exp\left[-\frac{|\tau|}{\tau_c}\right], \tag{14}$$

where τ_c is the correlation time for the intraband relaxation.

We obtain

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

$$= \tau_c\ I_0\ (\ -i\ \Delta_k \tau_c,\ \gamma_{cv}(k)\tau_c\ ) \tag{15}$$

where $$I_0\ (A,B) = \int_0^{\infty} dt \exp\ [\ -At\ -B(\ t + \exp\ (-t)\ -1\ )]. \tag{16}$$

In general I_0 (A,B) is evaluated by the continued fraction representation [24]. If we expand the argument of the exponential function in (16) up to the second order in t (weak modulation limit), we get the Gaussian line shape function:

$$\mathrm{Re}\ \Xi\ (0,\Delta_k) = \sqrt{\frac{\tau_c\ \pi}{2\gamma_{cv}(k)}} \exp\left(\ -\ \frac{\tau_c \Delta_k^2}{2\gamma_{cv}(k)}\right). \tag{17}$$

When $\tau_c = 0$ (strong modulation limit), Eq. (16) yields the Markovian lineshape given by

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

which is Lorentzian used in most calculations.

Here $$\gamma_{vc}(k) = \gamma_{cv}(k)$$

$$= \frac{1}{2}\ \{\ \frac{1}{\tau_v(k)} + \frac{1}{\tau_c(k)}\ \},$$

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

$$1 + \mathrm{Re}\ g_2(\infty,\Delta_k) = 1 + \frac{\gamma_{cv}(k)\tau_c}{1\ + \Delta_k^2 \tau_c^2}. \tag{18}$$

The gain (or line shape) enhancement factor at resonace ($\Delta_k = 0$) is

$$1 + \mathrm{Re}\, g_2(\infty,\Delta_k) = 1 + \gamma_{cv}(k)\tau_c \,. \tag{19}$$

Gain enhancement is due to the renormalized memory effects and may be caused by the absence of a strict energy conservation on very time scale as compared with the correlation time of the system governed by non-Markovian quantum kinetics. The probability that the enhanced transition occurs is proportional to the number of non-randomizing scattering events per second, $\gamma_{cv}(k)$, times the time interval τ_c for which the memory effects extend. Non-Markovian enhancement of optical gain becomes significant as the correlation time increases. For example, when τ_c is 50 fs, the peak value of optical gain at $\Delta_k = 0$ is predicted to be enhanced by as much as 50% for the typical intraband relaxation time of 100 fs.

The correlation time τ_c and the intraband relaxation $\gamma_{cv}(k)$ are related to each other by the frequency fluctuations and the their relation is described as follows:

Substituting $\tau = 0$ in Eq. (15), we get

$$<< \alpha k \mid H_i^2(t) \mid \alpha k >>_i = \frac{1}{2\tau_c \tau_\alpha(k)} \,. \tag{20}$$

Then, from Eq. (20) and integrating Eq. (14) over the time t, we obtain

$$\frac{\gamma_{cv}(k)}{\tau_c} = << ck \mid H_i^2(t) \mid ck >>_i + << vk \mid H_i^2(t) \mid vk >>_i \,, \tag{21}$$

where the right half side of eq. (21) the total intraband frequency fluctuation. We assume that the intraband relaxation $\gamma_{cv}(k)$ used in the line shape functions is mostly due to elastic or randomizing collisions, i.e., Markovian processes. The correlation time is then inversely proportional to the energy transfer in the intraband frequency fluctuations.

III. SUMMARY

In this paper, the time-convolutionless equation of motion for the interband pair amplitude is integrated directly to obtain the optical gain of semiconductor lasers. Plasma screening and excitonic effects are taken into account using an effective Hamiltonian in the time-dependent Hartree-Fock approximation. Time-convolutionless quantum kinetic equations incorporate the non-Markovian relaxation (or dephasing) and the renormalized memory effects 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. 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 a strict energy conservation in the non-Markovian quantum kinetics.

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