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Kyoto University
TIME-CONVOLUTIONLESS REDUCED-DENSITY-OPERATOR THEORY OF 
HIGHLY EXCITED SEMICONDUCTORS

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In this work, a reduced description of the dynamics of carriers in excited semiconductors is developed. Especially, the optical gain and the line shape function of a quantum-well is studied taking into account the valence-band mixing, non-Markovian relaxation and the many-body effects. Conventional gain spectra calculated with the Lorentzian line shape function show two anomalous phenomena: unnatural absorption region below the band-gap energy and mismatch of the transparency point in the gain spectra with the Fermi-level separation, the latter suggesting that the carriers and the photons are not in thermal (or quasi) equilibrium. It is shown that the non-Markovian gain model with many-body effects removes the two anomalies associated with the Lorentzian line shape function with the proper choice of the correlation time.

In previous works 1,2, the author has derived 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. TCL 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 the component of the wave function prior to the collision. As a result, the strict energy conservation may no longer hold for the 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 the optoelectronic devices. Conventional theoretical calculations3-6 of the optical gain of semiconductor lasers are usually based on the density matrix formalism with a phenomenological damping term which gives the Lorentzian line shape function for the optical gain. However, it was pointed out by Yamanishi and Lee 7 and by others 8-10 that the optical gain spectra calculated with the Lorentzian line shape function deviate from the experimental results. Especially, an anomalous absorption region appears at photon energies below the band-gap in the gain spectra as long as the Lorentzian line shape is used. Moreover, the detailed balance between absorption and emission of photons requires that the transparency point in the gain spectra coincide with the Fermi (or quasi Fermi) -level energy.10,11 In general, the gain spectra with the Lorentzian do not satisfy the detailed balance condition, either. In addition, most of the previous work 12,13 on many-body effects on the optical gain also assumed Markovian (or Lorentzian) line shape functions to describe the intraband relaxation processes. In this letter, we present a model for the optical gain of a quantum-well laser taking into account the non-Markovian relaxation, many-body effects, and the valence-band mixing employing the 6x6 Luttinger-Kohn Hamiltonian. The optical gain and the line shape function of the quantum well under an external optical field are derived from recently developed time-convolutionless quantum-kinetic equations 1,2 for electron-hole pairs near the band edge. These equations are the generalization of the semiconductor Bloch equations by incorporating the non-Markovian relaxation. Many-body effects such as band-gap renormalization and excitonic enhancement are included by taking into account the Coulomb interaction in the Hartree-Fock approximation. In this work, the plasma screening of the Coulomb interaction is not considered and this makes the calculation of the many-body effects to be overestimated. Unnormalized single-particle energies are obtained using the
multiband effective approximation, i.e., a parabolic band structure for electrons and a 6x6 Luttinger-Kohn model \(^{14}\) for holes. The latter includes the spin-orbit (SO) split-off band coupling effects on the valence-band structure. It was shown \(^{14}\) that the account of the SO coupling would be particularly important for the semiconductor with narrow spin-orbit (SO) split-off band splitting, the heterostructures with relatively large valence-band-edge discontinuity, and the strained-layers.

The non-diagonal interband matrix element \(p_{k}(t)\) which describes the interband pair amplitude induced by the optical field, is given by \(^{2}\)

\[
\frac{\partial}{\partial t} p_{k}(t) = i [E_{c}(k) - E_{v}(k)] p_{k}(t) + i [\mu^{*}(k)E_{p}(t) + \sum_{k'} V(k-k') p_{k'}(t)] [\eta_{ck}(t) - \eta_{vk}(t)]
\]

\[
+ \int_{0}^{t} d\tau \{ <\eta_{ck} | [H_{c}(t) (U_{o}(\tau)H_{c}(t - \tau))] | \eta_{vk} >_{1} >_{1} p_{k}(\tau)
\]

\[
+ i \int_{0}^{t} ds \exp \{-i [E_{c}(k) - E_{c}(k')] s\} \{ <\eta_{ck} | [H_{c}(t) (U_{o}(\tau)H_{c}(t - \tau))] | \eta_{vk} >_{1} >_{1} \}
\]

\[
\times \mu^{*}(k)E_{p}(t - s) \{ (\rho_{0}^{(0)})_{ckk'}(t) - (\rho_{0}^{(0)})_{vk}(t) \}, \quad (1)
\]

where \(V(k)\) is the unscreened Coulomb potential, \(U_{o}(t) = T \exp \{-i \int_{0}^{t} L_{o}(s)\) is the unperturbed evolution operator of the system with the time-ordering operator \(T\), \(\mu(k)\) is the dipole moment, \(\rho_{0}(0) = U_{o}(t) \rho(0), \rho(0)\) is the initial condition for the reduced density operator, \(<\ldots>_{1}\) is the average over the stochastic process \(H_{c}(t)\), and \(\eta_{ck}(t), \eta_{vk}(t)\) are the nonequilibrium distributions for electrons in the conduction band and in the valence band, respectively. The Coulomb term \(\sum_{k} V(k-k') p_{k'}(t)\) is responsible for the excitonic effects and \(E_{c}(k), E_{v}(k)\) are renormalized single particle energies.

The optical gain \(g(\omega)\) is obtained after some lengthy mathematical manipulations and is given by \(^{1,2}\)

\[
g(\omega) = \frac{\omega_{0} n_{r} c}{n_{r}} \frac{2}{V} \sum_{k} \frac{Re \Xi(0,\Delta_{k})}{1 - Re q_{k}(0)} |\mu(k)|^{2} [1 + Re g_{2}(\omega,\Delta_{k})]^{2} <\eta_{ck} - \eta_{vk} >_{1} \quad (2)
\]

with \(Re q_{k}(0) = \sum_{k'} V_{s}(k-k')Re \Xi(0,\Delta_{k}) <\eta_{ck} - \eta_{vk} >_{1} \quad (3)\)

where \(\mu\) is the permeability, \(n_{r}\) is the refractive index, \(c\) is the speed of light in free space, \(V\) is the volume, and \(\varepsilon_{0}\) is the permittivity of free space. In equation \((2)\), \(Re \Xi(0,\Delta_{k})\) is the line shape
function that describes the spectral shape of the optical gain in a driven semiconductor and $\Delta_k = E_c(k) - E_{v}(k) - \omega$. The denominator $[1 - \text{Re} q_1(0)]$ describes the gain enhancement due to the excitonic effects caused by the attractive Coulomb interaction and the factor $(1 + \text{Re} g_2(\infty, \Delta_k))$ describes the gain (or line shape) enhancement due to the interaction between the optical field and the stochastic reservoir of the system.

It was shown $^2$ that the line shape of the gain spectra is Gaussian for the simplest non-Markovian quantum kinetics:

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

(4)

where $\tau_c$ is the correlation time for the intraband relaxation.

Fig.1 We plot the calculated optical gain spectra for the three cases of: (i) Markovian model with Lorentzian line shape function (dashed line), (ii) non-Markovian model (dotted line), and (iii) non-Markovian model with excitonic enhancement (solid line) for the correlation time $\tau_c = 20$ fs.
As a numerical example, we have calculated the band-structure, the band-gap renormalization (BGR), and the optical gain spectra of a lattice-matched 60 Å In\textsubscript{0.53}Ga\textsubscript{0.47}As quantum well surrounded by InP outer barriers. In Fig. 1, we plot the calculated optical gain spectra for the three cases of: (i) Markovian model with Lorentzian line shape function (dashed line), (ii) non-Markovian model (dotted line), and (iii) non-Markovian model with excitonic enhancement (solid line) for the correlation time $\tau = 20$ fs. The intraband relaxation time of 0.1 ps and the carrier density of $3 \times 10^{18}$ cm\textsuperscript{-3} are assumed throughout the calculation and the renormalization of the band-gap energy is taken care of in all three cases.

In the absence of spectral broadening the optical gain spectra are related to the spontaneous emission spectra from the detailed balance between absorption and emission of photons. One of the remarkable feature of this relation is that there is a transparency point in the gain spectra which coincide with the Fermi (or quasi-Fermi) - level separation that suggests the carriers and the photons are in thermal (or quasi) equilibrium. For the carrier density of $3 \times 10^{18}$ cm\textsuperscript{-3} the Fermi-level separation is 0.8752 eV for the quantum-well structure. Then the optical gain spectra calculated with the Lorentzian line shape function have two anomalies: unnatural absorption region below the band-gap energy and mismatch of the transparency point of the gain with the Fermi-level separation, the latter suggests that the carriers and the photons are not in thermal (or quasi) equilibrium. It is seen that the two anomalies associated with the Lorentzian line shape are removed in the non-Markovian model with many-body effects. The interband excitonic enhancement causes the peak gain to increase from 607.1 cm\textsuperscript{-1} to 858.9 cm\textsuperscript{-1} for $\tau = 20$ fs. It is interesting to see that the detailed balance between absorption and emission of photons is nearly satisfied in the high energy tail or the large time in the temporal domain even in non-Markovian dynamics.

In summary, the optical gain of a quantum-well laser is calculated taking into account the valence-band mixing, the non-Markovian relaxation and the many-body effects. The valence-band structure calculations include the spin-orbit (SO) split-off band coupling which turns out to have significant effects on the band-structure, density-of-states, dipole moments, and the band-gap renormalization. The gain spectra calculated with the Lorentzian line shape function show two anomalies: unnatural absorption region below the band-gap energy and mismatch of the transparency point in the gain spectra with the Fermi-level separation, the latter suggesting that the carriers and the photons are not in thermal (or quasi) equilibrium. We have shown that the non-Markovian gain model with many-body effects removes the two anomalies associated the Lorentzian line shape function.

References