Importance of the microscopic effects on the linewidth enhancement factor of quantum cascade lasers

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Abstract: Microscopic density matrix analysis on the linewidth enhancement factor (LEF) of both mid-infrared (mid-IR) and Terahertz (THz) quantum cascade lasers (QCLs) is reported, taking into account of the many body Coulomb interactions, coherence of resonant-tunneling transport and non-parabolicity. A non-zero LEF at the gain peak is obtained due to these combined microscopic effects. The results show that, for mid-IR QCLs, the many body Coulomb interaction and non-parabolicity contribute greatly to the non-zero LEF. In contrast, for THz QCLs, the many body Coulomb interactions and the resonant-tunneling effects greatly influence the LEF resulting in a non-zero value at the gain peak. This microscopic model not only partially explains the non-zero LEF of QCLs at the gain peak, which observed in the experiments for a while but cannot be explicitly explained, but also can be employed to improve the active region designs so as to reduce the LEF by optimizing the corresponding parameters.

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References and links


1. Introduction

Quantum cascade lasers (QCLs) are unipolar laser sources relying on intersubband transitions in multiple quantum well systems. The light emission can be tuned across the mid-infrared (mid-IR, from 3 to 20 μm) and Terahertz (THz, from 1.2 to 5 THz, or 60 to 250 μm) ranges of the electromagnetic spectrum. As compact and coherent radiation sources, they have received considerable attentions since their first demonstrations [1, 2].

Linewidth enhancement factor (LEF, α) plays an important role in determining the optical emission linewidth of the laser systems [3] and the frequency responses [4]. It is defined as the ratio of the carrier-induced variation of the real and imaginary parts of the susceptibility, and describes the amplitude-phase coupling [3]. Owing to this coupling, a variation of the intensity of optical field will induce an excess perturbation of the phase of a laser mode, which influences the performance of lasers e.g. causes the laser linewidth increases well beyond the Schawlow-Townes limit. Typical value of LEF in interband semiconductor diode lasers is about 2-7 [5], resulting from the asymmetric differential gain spectrum caused by two bands associated with the laser transition with opposite curvature in the k-space. In contrast, due to the intersubband transition characteristics, QCLs are expected to have a narrow and symmetric gain spectrum, and hence, according to the Kramers-Kronig relation, resulting in a zero LEF at the peak gain wavelength predicted by Faist et al [2]. However, the currently reported experiments [6–11] have demonstrated that the LEFs in both mid-IR and THz QCLs are not zero, although they are much smaller than those of diode lasers. For example, a LEF of up to –0.5 and 0.5 at the gain peak was reported at a lasing wavelength of 8.22 μm [6] and ~116 μm (~2.55 THz) [7], respectively. In addition, a strong dependence of the LEF on frequency detuning was observed, and a large value of up to –2 was reported for distributed feedback (DFB) mid-IR QCLs [10].

Although several experimental works have been carried out in studying the LEF of QCLs, few theoretical investigations on LEF has been reported [11] and the reasons behind the observed non-zero LEF at the gain peak has not been completely understood for both mid-IR and THz QCLs. In the previous work, the non-zero LEF was investigated in mid-IR QCLs by considering the refractive index change due to the device self-heating and the transitions not involved in the laser action based on the experiments and the macroscopic theoretical analysis [11]. The results show that device self-heating is the dominate factor compared with the other
factors. In addition to the device self-heating effect, other mechanisms such as many body Coulomb interactions, coherence of resonant tunneling (RT) and non-parabolicity can cause the non-zero LEF of QCLs at the gain peak, which cannot be considered in the macroscopic picture. Furthermore, a theoretical analysis on the LEF of THz QCLs is lacked. Since the active region structures of THz QCLs are different from those of mid-IR QCLs, they shall show different characteristic of non-zero LEF. To disclose more physical underlining mechanisms of non-zero LEF value in both mid-IR and THz QCLs, a microscopic model is required.

Recently, we have developed a microscopic density matrix model to examine the optical gain of THz QCLs demonstrating that the many body effects, which lead to renormalization of band structure and Rabi frequency, have significant modifications to the gain profile [12]. In this paper, we extend this microscopic model to further investigate the role of many body Coulomb interactions, coherence of resonant tunneling and non-parabolicity on the LEF of both mid-IR and THz QCLs. The results show that, for mid-IR QCLs, both the many body Coulomb interactions and non-parabolicity contribute to the non-zero LEF among these three factors. Furthermore, the $\alpha$-spectrum is blue-shifted as the injection current increases, which may explain the recent experimental result of $\alpha$-value increases with the bias. In contrast, for THz QCLs, both the many body Coulomb interactions and the coherence of resonant tunneling greatly influence the LEF deviating from zero at the gain peak. Our results demonstrate the importance of the microscopic characteristics on the LEF of QCLs, and partially explain the discrepancy between the experimental results and the previous models.

2. LEF in mid-IR QCLs

2.1. The microscopic model

Fig. 1. Schematic conduction band diagram of a two-phonon resonance gain region designed at 60 kV/cm in the “tight-binding” scheme [13]. The coupling between the periods, achieved by resonant tunneling ($\Omega_{\text{RT}}$ is the coupling strength), is shown through the injector barrier. The layer sequence of the structure, in Angstrom, and starting from the injection barrier, is as follows: 40/25/15/74/11/60/34/39/11/34/11/34/12/37/17/41. In$_{0.53}$Ga$_{0.47}$As barrier layers are in bold, In$_{0.53}$Ga$_{0.47}$As well layers are in roman, and $n$-doped layers ($2.5 \times 10^{17}$ cm$^{-3}$) are underlined.

For mid-IR QCLs, in order to make a comparison with experiments, we use the active region with the three-well vertical design which has been used in the LEF measurement experiment [6, 13]. This structure is also very similar with the one used for another LEF experiment in [9]. Figure 1 shows the conduction band diagram and magnitude squared envelope wave functions of this design in a “tight-binding” scheme. The coupling between the periods,
achieved by tunneling ($\Omega_{51}$ is the coupling strength), is shown through the injector barriers. The energy states within one period are coupled through scattering processes, but at the injector barrier, the transport is modeled by tunneling. In order to conveniently treat the many body effects, we derive the dynamic equations of motion in the second quantized representation. The Hamiltonian of the system of mid-IR QCL in Fig. 1, which characterizes the electron-light coupling, the tunneling effects, free electrons and electron-electron Coulomb interactions, can be written as

$$H = -\sum_{j=1,3,5} \sum_{k} (\mu_{5j} E b_{j,k} b_{j,k}^\dagger + c.c.) - \sum_{k} \left[ \frac{(\Delta_{5j}/2)}{2} b_{5,k}^\dagger b_{5,k} + c.c. \right] + \sum_{j=1,3,5} \sum_{k} \epsilon_{j,k} b_{j,k}^\dagger b_{j,k} + \sum_{k} \frac{1}{2} \sum_{q} V_{q}^{uvv} b_{j,k}^\dagger b_{j,k}^\dagger b_{j,k} b_{j,k}^\dagger,$$

where $E(t) = \xi(t) / 2 e^{-i\omega t} + c.c.$ is the laser field (\xi is the slowly varying complex electric field amplitude, $\omega_j$ is the laser frequency). $\mu_{5j}$ is the electron charge times the dipole matrix element of laser transition between energy level $j$ and 5, $\Delta_{5j}$ is the injection coupling strengths. $\epsilon_{j,k}$ is the $j$th subband energy, $k$ is the in-plane wave vector. $V_{q}^{uvv}$ is the two dimensional screening Coulomb matrix element [12, 14]. The parasitic coupling between levels 1 and 4 is neglected, which is a reasonably good approximation for mid-IR QCLs [15]. The relaxation of electrons in level 3 into level 1 is characterized by an effective scattering rate.

Using the semiclassical laser theory and Maxwell equations, the gain $G$ and carrier-induced refractive index change $\delta n$ are given by

$$G = -\frac{2\epsilon_0}{\epsilon_0 n c V_m \xi} \text{Im} \left[ \sum_{k} (\mu_{55} P_{55,k} + \mu_{5j} P_{5j,k}) \right],$$

$$\delta n = \frac{\mu}{\epsilon_0 n V_m \xi} \text{Re} \left[ \sum_{k} (\mu_{55} P_{55,k} + \mu_{5j} P_{5j,k}) \right],$$

where $\epsilon_0$ is the vacuum permittivity, $n$ is the refractive index, $c$ is the light speed in vacuum, and $V_m$ is the volume of one period of active region. $p_{q,k} = \langle b_{q,k}^\dagger b_{q,k} \rangle$ and $n_{q,k} = \langle b_{q,k}^\dagger b_{q,k} \rangle$ are the slowing varying polarization and electron occupation, respectively. The details of the derivation of dynamic equations for polarization and electron occupation, considering the many-body Coulomb interaction, coherence of resonant-tunneling transport and non-parabolicity, can be found in Appendix A.

The linewidth enhancement factor $\alpha$ can be obtained by the ratio of the change in the real part of the refractive index change $\delta n$ to the change in the gain $G$ with respect to the carrier density $N_0$ [14]

$$\alpha = -2 \frac{\omega_j}{c} \frac{d}{dN_0} \left[ \frac{dG}{dN_0} \right].$$

2.2. Results and discussions

Figure 2 shows the $\alpha$-spectrum at different biases calculated from the microscopic model including the many body Coulomb interactions, coherence of resonant-tunneling transport and non-parabolicity. The points shown in the figure indicate the positions of the gain peak. In contrast to the macroscopic calculations of zero LEFs the value of $\alpha$-spectrum at gain peak cannot be neglected due to the asymmetry of gain spectrum caused by the interplay of many body interactions, non-parabolicity and tunneling effects. The obtained LEF is around $-0.22.$

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The sign of the $\alpha$-value at gain peak reflects the asymmetry of gain spectrum relative to the lasing central frequency $\omega_\lambda$. The experimental $\alpha$-value of this active region design at gain peak is around $-0.5$ [6]. The absolute experimental value is 0.28 larger than our simulation result of $-0.22$. This discrepancy can be attributed to the refractive index change due to device self-heating effect demonstrated in [11], which needs a more complicated calculation on the thermal effects and is not taken into account in our simulations. Therefore, the microscopic contributions induced by many body interaction, non-parabolicity and coherence of resonant tunneling and refractive index change due to device self-heating are all important to determine the LEF of QCLs. Moreover, although the LEF at gain peak is smaller than 1, the LEF shows a large value as the lasing frequency is slightly away from the central frequency. This means that LEF strongly depends on the frequency detuning. Therefore, for the waveguide structure with grating, a laser will show a relatively large LEF at lasing frequency. In addition, for DFB-QCLs, the emission wavelengths will be shifted to longer wavelengths as the injection current increases [16], the value of LEF at the operation wavelength will rise as the external bias increases. Because the $\alpha$-spectrum is blue-shifted as the external bias increases seen from Fig. 2, the LEF will increase as the injection current rises. This could explain the experimental observations in a QCL in [9], where the active region is very similar with the one used in this paper (see Fig. 1), that the LEF increases with the injection current. It is noted that the measured LEF in [9] does not include the contribution from the device self-heating effect due to self-mixing measurement technique used in this experiment.

![Fig. 2. Linewidth enhancement factor (LEF) including many body Coulomb interactions, coherence of resonant tunneling transport and non-parabolicity for different biases at 100 K. The points indicate the values of LEF at the gain peak.](image)

Figure 3 shows the details of $\alpha$-parameter at the gain peak position, the 0.18 $\mu$m blue-shift and red-shift positions relative to the peak under different biases computed from the microscopic model “many body + non-parabolicity” (considering both many body and non-parabolicity effects), the microscopic free-carrier model (considering both free carriers and non-parabolicity but neglecting the renormalization of band structure and Rabi frequency), the macroscopic density-matrix model with and without including the resonant-tunneling effect, respectively. As shown in Fig. 3(a), the many body Coulomb interaction, coherence of resonant tunneling and non-parabolicity all induce a finite value of LEF at the gain peak. Furthermore, by the comparison of these contributions to LEF, the many body Coulomb interaction and non-parabolicity parameter have more important effects on the LEF at gain peak. This is because the many body Coulomb interaction and non-parabolicity all tend to distort the shape of gain spectrum, as shown in Fig. 3(b), where the non-parabolicity can more greatly modify the gain spectrum as compared with Coulomb interaction.
The non-parabolicity in In$_{0.53}$Ga$_{0.47}$As quantum wells is higher than that in GaAs quantum wells. Therefore, the gain spectrum in mid-IR QCLs can be more greatly influenced by the non-parabolicity. If the non-parabolicity (proportional to the ratio of the effective mass at the upper laser level and the lower laser level) slightly increases, the LEF will increase according to Fig. 4(a). The increase is attributed to the influence of non-parabolicity on symmetry of gain spectrum, as shown in Fig. 4(b). Therefore, according to the above simulations, it is
expected that, for similar structures, mid-IR QCL emitting at a shorter wavelength and with a higher non-parabolicity has a larger $\alpha$-value.

### 3. LEF in THz QCLs

![Conduction band diagram](image)

Fig. 5. Conduction band diagram of a four level resonant-phonon THz QCL with a diagonal design at 12.3 kV/cm in the “tight-binding” scheme. $\Omega_{41}$, $\Omega_{23}$ and $\Omega_{31}$ are the injection, extraction and parasitic coupling strength, respectively. The thickness in angstrom of each layer is given as 49/88/27/82/42/160 starting from the injector barrier. The barriers Al$_{0.15}$Ga$_{0.85}$As are indicated in bold fonts. The widest well is doped at $3 \times 10^{10}$ cm$^{-2}$.

The present best temperature performance of THz QCLs is obtained by using resonant-phonon (RP) design [17] at an operation temperature of up to ~200 K. We consider this design in this paper. Figure 5 shows the conduction band diagram in the “tight-binding” scheme. The details of this structure can be found in [12]. The Hamiltonian of this system can be expressed as

$$
H = -\sum_{k} (\mu E b_{k}^{\dagger} b_{k} + c.c.) - \sum_{k} \left[ (\Delta_{nn}/2) b_{k}^{\dagger} b_{k} + c.c. \right] - \sum_{k} \left[ (\Delta_{nn}/2) b_{k}^{\dagger} b_{k} + c.c. \right] - \sum_{k} \left[ (\Delta_{nn}/2) b_{k}^{\dagger} b_{k} + c.c. \right]
$$

$$
- \sum_{k} \sum_{j} \sum_{i} \sum_{k} e_{ij} b_{k}^{\dagger} b_{k} b_{j} + \frac{1}{2} \sum_{k} \sum_{i} \sum_{k} V_{ijkl}^{\alpha} b_{k}^{\dagger} b_{k} b_{l} b_{l}^{\dagger}.
$$

The optical gain and the refractive index change are written as

$$
G = \frac{2\omega_{0}}{\varepsilon_{0} n c V_{m}^{\omega}} \text{Im} \left( \sum_{k} \mu_{sk} \tilde{P}_{sk}^{a} \right),
$$

$$
\delta n = \frac{\mu}{\varepsilon_{0} n V_{m}^{\omega}} \text{Re} \left( \sum_{k} \mu_{sk} \tilde{P}_{sk}^{a} \right).
$$

Similar to Eq. (4), we can obtain the LEF of THz QCLs.
Fig. 6 LEF including many body Coulomb interactions, coherence of resonant-tunneling transport and non-parabolicity for different biases at 100 K. The points indicate the values of LEF at the gain peak. The simulation parameters can be found in [12].

Figure 6 shows the $\alpha$-spectrum at different biases calculated from the microscopic model including the many body Coulomb interactions, coherence of resonant-tunneling transport and non-parabolicity. The points indicate the positions of the gain peak. Similarly, the value of $\alpha$-spectrum of THz QCLs at gain peak cannot be neglected due to the asymmetry of gain spectrum caused by the interplay of many-body interaction, non-parabolicity and resonant-tunneling effects. Its value is around $-0.7$ when electric pumping does not exceed the designed bias. The sign of $\alpha$-value at gain peak reflects the asymmetry of gain spectrum relative to the lasing central frequency $\omega_\lambda$. It is noted that, when the bias goes above the designed value and becomes 13.3 kV/cm, the LEF becomes $-0.3$. This is mainly attributed to the symmetric changes of gain spectrum with the extraction detuning due to tunneling effects i.e. the energy splitting due to the coupling between the lower laser level and the extraction level, as shown in Fig. 5 in [12]. It has shown that the peak position and the lineshape of gain spectrum strongly follow the variations of extraction detuning. When the extraction detuning changes to a positive value from a negative one, the peak frequency is redshifted. Therefore, the absolute value of LEF is reduced. In addition, although the LEF at gain peak is smaller than 1, the LEF shows a large value as the lasing frequency is slightly away from the central frequency, which means that LEF of THz also strongly depends on the frequency detuning.

Figure 7 shows the details of $\alpha$-parameter at frequencies of gain peak, 0.2 THz red-shift and 0.2 THz blue-shift relative to the peak position under different biases, computed from the microscopic model with “many body + non-parabolicity”, microscopic free-carrier model, the macroscopic models with and without resonant tunneling, respectively. According to the macroscopic models with and without resonant tunneling, the coherence of resonant tunneling contributes to a significant increase of the LEF at gain peak. Since tunneling exhibits an increasing broadening and modification for gain spectrum when injection level (level 1) and upper laser level (level 4), and lower laser (level 3) and extraction level (level 2) are in resonance simultaneously, the absolute $\alpha$ value rises with the increasing bias in the regime of negative injection and extraction detunings according to macroscopic model with resonant tunneling. Once the operation bias exceeds the designed bias, the sign of LEF is changed due to the variation of the symmetry of gain spectrum relative to the lasing central frequency $\omega_\lambda$. Furthermore, the non-parabolicity, in contrast to mid-IR QCLs, can only induce a slight influence on the LEF according to the comparisons between free-carrier model and macroscopic one with resonant tunneling, but the many body Coulomb interaction causes a large variation of LEF at gain peak with the comparison of the model “many-body + non-
parabolicity” and free-carrier one due to its strong modifications to gain spectrum, as shown in Fig. 8 (The Coulomb interaction include the Hartree-Fock, dephasing and scattering contributions, more details can be seen in [12]). Overall, the non-zero LEF of THz QCLs is mainly due to the combined impacts from the Coulomb interaction and coherence of resonant-tunneling effects.

4. Conclusion

In conclusion, our study shows that the interplay of the many body interactions, coherence of resonant-tunneling effects and non-parabolicity for both mid-IR and THz QCL play an important role on the non-zero LEF at gain peak. A strong dependence of the LEF on the lasing frequency detuning is observed. The results show that, for mid-IR QCLs, the many body interactions and non-parabolicity all play an important key role in non-zero LEF at gain peak, where non-parabolicity has a more significant influence. In contrast, the many body Coulomb interactions and coherence of resonant tunneling have a significant impact on the
LEF of THz QCLs, but the non-parabolicity only induces a little effect. Although the simulated LEF at gain peak is smaller than the measured value, our microscopic model can well explain some experimental observations e.g. the increase of LEF with injection current for DFB QCLs. The discrepancy between the experimental value and the proposed theoretical models can be attributed to the refractive index change due to device self-heating, which should also be considered in the future explorations.

Appendix A

According to the following Hershberger equation and the anticommutations relations of fermionic operator [14]

\[
\frac{dO}{dt} = \frac{i}{\hbar} [H, O],
\]

\[
[b_{i,k}, b_{j,k}^\dagger] = \delta_{i,j} \delta_{\alpha,\beta}, \quad [b_{i,k}, b_{j,k}^\dagger] = [b_{i,k}^\dagger, b_{j,k}^\dagger] = 0,
\]

where \( O \) is operator, \( H \) is the Hamiltonian. Due to the Coulomb interaction, the result is an infinite hierarchy of coupled differential equations. The hierarchy describes the correlation effect in the Coulomb potential. The first order correlation is induced by the Hartree-Fock contributions, which results in band structure and Rabi frequency renormalizations. Scattering and dephasing contributions cause the second order correlation in the Coulomb potential, and so on. In this paper, we only include the Hartree-Fock contributions, and dephasing and scattering contributions at the level of a relaxation-rate approximation. Then one can get the following equations of motion for the slowing varying polarization \( p_{\alpha,k} = \{b_{\alpha,k}, b_{\alpha,k}^\dagger\} \) and electron occupation \( n_{\alpha,k} = \langle b_{\alpha,k}^\dagger b_{\alpha,k} \rangle \) in the rotating-wave approximation

\[
\frac{dp_{\alpha,k}}{dt} = -\gamma_{5\alpha} p_{5\alpha,k} - \frac{\tilde{e}_{\alpha,k}^5}{\hbar} p_{5\alpha,k} + i\tilde{\Omega}_{5\alpha} (n_{5,k} - n_{\alpha,k}) - i\tilde{\Omega}_{5\alpha} p_{4\alpha,k} - i\tilde{\Omega}_{5\alpha}^\dagger p_{5\alpha,k},
\]

\[
\frac{dp_{\alpha,k}}{dt} = -\gamma_{5\alpha} p_{5\alpha,k} - \frac{\tilde{e}_{\alpha,k}}{\hbar} - \omega_\alpha p_{\alpha,k} - i\tilde{\Omega}_{\alpha} (n_{\alpha,k} - n_{\alpha,k}) + i\tilde{\Omega}_{\alpha} p_{\alpha,k} + i\tilde{\Omega}_{\alpha}^\dagger p_{\alpha,k},
\]

\[
\frac{dp_{\alpha,k}}{dt} = -\gamma_{5\alpha} p_{5\alpha,k} - \frac{\tilde{e}_{\alpha,k}}{\hbar} - \omega_\alpha p_{\alpha,k} - i\tilde{\Omega}_{\alpha} (n_{\alpha,k} - n_{\alpha,k}) + i\tilde{\Omega}_{\alpha} p_{\alpha,k} + i\tilde{\Omega}_{\alpha}^\dagger p_{\alpha,k},
\]

\[
\frac{dp_{\alpha,k}}{dt} = -\gamma_{5\alpha} p_{5\alpha,k} - \frac{\tilde{e}_{\alpha,k}}{\hbar} - \omega_\alpha p_{\alpha,k} - i\tilde{\Omega}_{\alpha} (n_{\alpha,k} - n_{\alpha,k}) + i\tilde{\Omega}_{\alpha} p_{\alpha,k} + i\tilde{\Omega}_{\alpha}^\dagger p_{\alpha,k},
\]

\[
\frac{dp_{\alpha,k}}{dt} = -\gamma_{5\alpha} p_{5\alpha,k} - \frac{\tilde{e}_{\alpha,k}}{\hbar} - \omega_\alpha p_{\alpha,k} - i\tilde{\Omega}_{\alpha} (n_{\alpha,k} - n_{\alpha,k}) + i\tilde{\Omega}_{\alpha} p_{\alpha,k} + i\tilde{\Omega}_{\alpha}^\dagger p_{\alpha,k},
\]

\[
\frac{dn_{\alpha,k}}{dt} = i(\tilde{\Omega}_{\alpha} p_{\alpha,k} - \tilde{\Omega}_{\alpha}^\dagger p_{\alpha,k}^\dagger) - \gamma_T [n_{\alpha,k} - f_{\alpha,k} (\mu_{\alpha}, T_{\alpha})] - \gamma_T [n_{\alpha,k} - f_{\alpha,k} (\mu_{\alpha}, T_{\alpha})],
\]

\[
\frac{dn_{\alpha,k}}{dt} = i(\tilde{\Omega}_{\alpha} p_{\alpha,k} - \tilde{\Omega}_{\alpha}^\dagger p_{\alpha,k}^\dagger) + i(\tilde{\Omega}_{\alpha} p_{\alpha,k} - \tilde{\Omega}_{\alpha}^\dagger p_{\alpha,k}^\dagger) + \gamma_T [n_{\alpha,k} - f_{\alpha,k} (\mu_{\alpha}, T_{\alpha})] - \gamma_T [n_{\alpha,k} - f_{\alpha,k} (\mu_{\alpha}, T_{\alpha})].
\]
\[
\frac{dn_{i,k}}{dt} = i(\tilde{\Omega}_{i,k} p_{i,k}^\dagger - \tilde{\Omega}_{i,k} p_{i,k}) - \gamma_i \left[ n_{i,k} - f_{i,k} (\mu_i, T_i) \right] - \sum_{j \neq i} \gamma_{ij} \left[ n_{i,k} - f_{i,k} (\mu_j, T_j) \right],
\]

(18)

\[
\frac{dn_{j,k}}{dt} = i(\tilde{\Omega}_{j,k} p_{j,k}^\dagger - \tilde{\Omega}_{j,k} p_{j,k}) - \gamma_j \left[ n_{j,k} - f_{j,k} (\mu_j, T_j) \right] - \sum_{j' \neq j} \gamma_{jj'} \left[ n_{j,k} - f_{j,k} (\mu_{j'}, T_{j'}) \right]
\]

(19)

where

\[
\gamma_{ij} = \sum_{k'} \left( V_{ij,k'}^\text{intr} n_{i,k'} - V_{ij,k'}^\text{inter} n_{j,k'} \right) f_{i,k'}^\text{av},
\]

(20)

\[
\tilde{\Omega}_{i,j} = \frac{\hbar^2 \epsilon_{i,j}}{2 m^*} + \frac{1}{\hbar} \sum_{k,k'} V_{i,j,k} n_{i,k} - \frac{2}{\hbar} V_{i,j,k}^\text{phon} \sum_k p_{i,k},
\]

(21)

\[
\tilde{\Omega}_{j,i} = \frac{\Delta_{ij}}{2 \hbar} + \frac{1}{\hbar} \sum_{k,k'} V_{j,i,k}^\text{pol} n_{j,k} - \frac{2}{\hbar} V_{j,i,k}^\text{phon} \sum_k p_{j,k},
\]

(22)

where \( \gamma_{ij} \) is the dephasing rate associated with energy levels \( i \) and \( j \). \( \gamma_j \) is the intrasubband electron-electron scattering rate at level \( j \). \( \gamma_{ij} \) is the combined electron-electron and electron-phonon scattering rate between levels \( i \) and \( j \). \( \gamma_{ij} \) is the spontaneous emission rate. \( T_j \) is the electron temperature at level \( j \), \( T_i \) is the lattice temperature. \( f_{i,k} \) is the Fermi-Dirac distribution with chemical potential \( \mu_i \) at level \( j \). The chemical potentials and temperatures are determined by electron number conservation and energy conservation, which are described in details in [12]. The influence of the subband dispersion, namely the nonparabolicity, is represented by using the effective mass of electrons \( m^* \). For subband \( j \), we have \( \epsilon_{j,k} = \epsilon_j + \hbar^2 k^2 / 2m^*_j \). For our mid-IR structure, we estimate that \( m^*_j / m^*_i = 1.28 \) [18, 19].

**Appendix B**

Similarly, one can obtain the following equations of motion for the polarization \( p_{i,k} = \langle b_{i,k}^\dagger b_{i,k} \rangle \) and electron occupation \( n_{i,k} = \langle b_{i,k}^\dagger b_{i,k} \rangle \) for THz QCLs

\[
\frac{dp_{i,k}}{dt} = -\gamma_{i,p} p_{i,k} - i \frac{\tilde{\epsilon}_{i,k}}{\hbar} p_{i,k} - i \tilde{\Omega}_{i,k} (n_{i,k} - n_{i,k}) - i \tilde{\Omega}_{i,k} p_{2i,k} - i \tilde{\Omega}_{i,k} p_{3i,k},
\]

(23)

\[
\frac{dp_{i,k}}{dt} = -\gamma_{i,p} p_{i,k} - i \frac{\tilde{\epsilon}_{i,k}}{\hbar} p_{i,k} - i \tilde{\Omega}_{i,k} (n_{i,k} - n_{i,k}) - i \tilde{\Omega}_{i,k} p_{3i,k} + i \tilde{\Omega}_{i,k} p_{4i,k},
\]

(24)

\[
\frac{dp_{j,k}}{dt} = -\gamma_{j,p} p_{j,k} - i \frac{\tilde{\epsilon}_{j,k}}{\hbar} p_{j,k} - i \tilde{\Omega}_{j,k} (n_{j,k} - n_{j,k}) + i \tilde{\Omega}_{j,k} p_{3j,k} + i \tilde{\Omega}_{j,k} p_{4j,k},
\]

(25)

\[
\frac{dp_{j,k}}{dt} = -\gamma_{j,p} p_{j,k} - i \frac{\tilde{\epsilon}_{j,k}}{\hbar} p_{j,k} - i \tilde{\Omega}_{j,k} (n_{j,k} - n_{j,k}) + i \tilde{\Omega}_{j,k} p_{3j,k} - i \tilde{\Omega}_{j,k} p_{2j,k},
\]

(26)

\[
\frac{dp_{2i,k}}{dt} = -\gamma_{i,p} p_{2i,k} - i \frac{\tilde{\epsilon}_{2i,k}}{\hbar} p_{2i,k} + i \tilde{\Omega}_{2i,k} p_{2i,k} + i \tilde{\Omega}_{2i,k} p_{3i,k} - i \tilde{\Omega}_{2i,k} p_{4i,k},
\]

(27)

\[
\frac{dp_{2i,k}}{dt} = -\gamma_{i,p} p_{2i,k} - i \frac{\tilde{\epsilon}_{2i,k}}{\hbar} p_{2i,k} + i \tilde{\Omega}_{2i,k} p_{2i,k} + i \tilde{\Omega}_{2i,k} p_{3i,k} + i \tilde{\Omega}_{2i,k} p_{4i,k},
\]

(28)
\[
\frac{dn_{i,k}}{dt} = -i(\Omega_0 n_{i,k} - \Omega_{it} p_{it,k}) - i(\bar{\Omega}_0 n_{it,k} - \bar{\Omega}_{it} p_{it,k}) - \gamma_i [n_{i,k} - f_{i,k} (\mu_{i,T})] - \gamma_{it} n_{i,k},
\]
(29)

\[
\frac{dn_{k}}{dt} = -i(\Omega_0 p_{k} - \Omega_{it} p_{it,k}) - i(\bar{\Omega}_0 p_{it,k} - \bar{\Omega}_{it} p_{it,k}) - \gamma_i [n_{k} - f_{i,k} (\mu_{i,T})] + \gamma_{it} n_{i,k} - i(\bar{\Omega}_{it} n_{i} - \bar{\Omega}_{it} n_{i,k}),
\]
(30)

\[
\frac{dn_{k}}{dt} = -i(\Omega_0 p_{k} - \Omega_{it} p_{it,k}) - \gamma_i [n_{k} - f_{i,k} (\mu_{i,T})] - \gamma_{it} [n_{i,k} - f_{i,k} (\mu_{i,T})].
\]
(31)

\[
\frac{dn_{i,k}}{dt} = -i(\Omega_0 p_{i,k} - \Omega_{it} p_{it,k}) - \gamma_i [n_{i,k} - f_{i,k} (\mu_{i,T})] + \gamma_{it} n_{i,k} - i(\bar{\Omega}_{it} p_{i} - \bar{\Omega}_{it} p_{i,k}).
\]
(32)

where

\[
\bar{\epsilon}_{s_{i,k}} = \epsilon_{s_{i,k}} - \sum_{k \neq k} (V_{s_{i,k}}^{\text{vso}} n_{s_{i,k}} - V_{s_{i,k}}^{\text{vso}} n_{s_{i,k}}) + \sum_{k \neq k} (n_{s_{i,k}} - n_{s_{i,k}}) \lambda_{s_{i,k}}^{\text{vso}},
\]

\[
\bar{\Omega}_0 = \frac{\mu \xi}{2\hbar} + \frac{1}{\hbar} \sum_{k \neq k} V_{s_{i,k}}^{\text{sst}} n_{s_{i,k}} - \frac{2}{\hbar} V_{s_{i,k}}^{\text{sst}} \sum_{k} p_{s_{i,k}},
\]

\[
\bar{\Omega}_{it} = \frac{\Delta \Omega}{2\hbar} + \frac{1}{\hbar} \sum_{k \neq k} V_{s_{i,k}}^{\text{sst}} n_{s_{i,k}} - \frac{2}{\hbar} V_{s_{i,k}}^{\text{sst}} \sum_{k} p_{s_{i,k}}.
\]

Since levels (1', 4) and (2, 3) are coherently coupled by the tunneling, the coherences corresponding to the levels (1', 3), (2, 4) and (1', 2) have a time-harmonic character due to the time-harmonic (3, 4) coherent coupling. Therefore, we try to look for solutions in the form of

\[
p_{4i,k} = p_{4i,k}^{(0)} + p_{4i,k}^{(1)} e^{-i\omega t}, \quad p_{3i,k} = p_{3i,k}^{(0)} + p_{3i,k}^{(1)} e^{-i\omega t},
\]

\[
p_{23,k} = p_{23,k}^{(0)} + p_{23,k}^{(1)} e^{-i\omega t}, \quad p_{21,k} = p_{21,k}^{(0)} + p_{21,k}^{(1)} e^{-i\omega t},
\]

where \(p_{ij,k}^{(0)}\) is the static tunneling induced coherence, and \(p_{ij,k}^{(1)}\) is the laser-induced coherence [20].

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