Theoretical investigation on intrinsic linewidth of quantum cascade lasers

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**NANYANG
TECHNOLOGICAL
UNIVERSITY**

**Theoretical investigation on intrinsic linewidth
of quantum cascade lasers**

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School of Electrical and Electronic Engineering

2014

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Summary

Quantum cascade lasers (QCLs) are unipolar laser sources relying on intersubband transitions in coupled 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. Since their invention in 1994 for mid-IR QCLs and in 2002 for THz QCLs, respectively, these lasers have undergone tremendous improvement, and have become probably the most prominent coherent light sources in the mid-IR and THz spectral ranges. However, many important applications of mid-IR and THz coherent light sources, *e.g.* spectroscopy and high-speed free-space data communication, are greatly influenced by intrinsic laser fluctuations and noises. Although intrinsic linewidth and noise in semiconductor diode lasers have been widely investigated, theoretical and experimental studies in noise dynamics and linewidth of QCLs have only attracted interests recently. This field is still in its infancy. Since the operational principle of QCLs is totally different from that of semiconductor diode lasers (resonant tunneling effect and coherent interactions in QCLs are unique owing to the intersubband transitions), models on noise and linewidth investigations of diode lasers cannot be directly applied to QCLs. New physical models must be developed on both noise dynamics and linewidth for QCLs.

This thesis discusses many aspects of intrinsic linewidth and noises of QCLs, and it is divided into three main parts. Since optical gain spectrum can greatly influence the intrinsic linewidth of lasers, in the first part, we report the study on optical gain of QCLs. The optical gain of QCLs is investigated based on a developed *microscopic* density-matrix (DM) model. Intersubband semiconductor-Bloch equations are established by incorporating many-body Coulomb interaction, non-parabolicity and coherence of resonant-tunneling transport effects in a quantitative way. The calculations demonstrate the importance of many-body interaction, non-parabolicity and resonant-tunneling transport on optical gain spectrum of mid-IR and THz QCLs. The results show that the gain peak in frequency calculated by the developed *microscopic* DM model is closer to the experimentally measured lasing frequency compared with the existing *macroscopic* DM model. In addition, the dependence of optical gain of THz and mid-IR QCLs on device parameters such as the injection and extraction coupling strengths, energy detuning and doping density are also systematically studied in details. This model provides a comprehensive picture of optical properties of THz and mid-IR QCLs and potentially enables a

more accurate and faster prediction of the device performance *e.g.* the laser linewidth enhancement factor and current characteristics.

In the second part of the thesis, we report the intrinsic linewidth of QCLs caused by the spontaneous emission, thermal photon and fundamental thermodynamic fluctuation. The linewidth induced by spontaneous emission and thermal photon is analytically derived on the basis of the quantum mechanical Langevin equation. It differs from the traditional rate equation model for the laser linewidth calculation of diode lasers in that the dynamics of coherent interaction and resonant-tunneling effects are considered. Results show that the coupling strength and the dephasing rate associated with resonant tunneling strongly affect the linewidth of THz QCLs in the incoherent resonant-tunneling transport regime but only induce little influence in the coherent regime. The dynamics of coherent interaction and resonant-tunneling transport show negligible effects on the linewidth calculation of mid-IR QCLs due to strong coupling in resonant tunneling. The fundamental thermodynamic noise and linewidth broadening is investigated based on the Green function analysis and the *Van Vliet-Fassett* theory. The results show that the fundamental frequency noise caused by intrinsic temperature fluctuations is prominent in the low frequency range (below a few kHz) and is sensitive to the temperature, heat conductivity and the thickness of the active region/substrate. Specifically, for THz QCLs, considering only the refractive index variation caused by the current-induced device self-heating, we calculate the linewidth broadening to be only around 1 Hz, which is comparable to the predicted value of 3 Hz caused by spontaneous emission and thermal photon in high power THz QCLs. For mid-infrared QCLs, this frequency noise leads to a linewidth broadening from 14.74 Hz to 62.02 Hz as the temperature increases from 200 K to 400 K. When the microscopic features of the refractive index variations associated with the intersubband gain transition, the self-heating-induced thermal expansion and energy level broadening in mid-IR QCLs are considered, an estimation shows that the linewidth broadening increases greatly by a factor of more than 4 times.

In the final part, microscopic density matrix analysis on the linewidth enhancement factor (LEF) of QCLs is reported by 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, both the many body Coulomb interaction and non-parabolicity contribute to the non-zero LEF. In

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1 Introduction

1.1 Introduction and background

The first semiconductor laser was reported in 1962 by using homojunctions [1]. But these lasers were operated at cryogenic temperature with a high threshold current density. Until the introduction of the concept of heterojunction in 1970s [2], the low-threshold and room-temperature semiconductor laser was invented due to the improvement of both carrier and optical confinement. In early 1980s, quantum well and multiple quantum wells lasers were developed at room temperature with a much lower threshold current density and somehow wavelength tunability to some extent [3]. The quantum well is a two-dimensional system which confines the motion of carriers in the growth direction. Due to the high power, high efficiency, small size, high reliability and wavelength tunability, quantum well semiconductor lasers have been widely used in our modern life. The high-performance semiconductor lasers are typically operated in the visible and near-infrared wavelengths below 1.5 μm due to the limit of bandgap of materials. However, in the last a few decades, there was an increasing demand on the spectral applications of mid-infrared (mid-IR) and terahertz (THz) wavelength ranges e.g. mid-IR trace-gas absorption spectroscopy and THz imaging [4-9]. The breakthrough occurred in 1994, a novel class of semiconductor laser, called quantum cascade laser (QCL) [10], emerged. The active region of QCLs is made up of repeating multi-quantum wells of nanometric thickness and constructed as superlattice structures. They overthrow the key working principles of conventional semiconductor lasers, in that they do not involve a transition of the electrons from the conduction band into the valence band but depend on intersubband transitions between quantized energy levels within the conduction band [5]. Therefore, their emission wavelength can be typically engineered across the mid-IR (3-24 μm) [5, 11-13] and THz (1.2-5 THz, or 60-250 μm) [14-16] regions by changing the thickness of quantum wells and barriers. In this section, we firstly demonstrate the differences of intersubband and interband transitions, then present the development of the mid-IR and THz QCLs.

1.1.1 Intersubband vs. interband transitions

QCLs are the unipolar devices. The operational principles of QCLs are typically different from the conventional diode laser. In this section, we will briefly describe the main differences between intersubband and interband lasers. This will help us to better understand the operating

principle of QCLs as compared to diode lasers. Figure 1 - 1 schematically shows the interband and intersubband transitions in a quantum well. For the interband transition, the optical radiation takes places by the recombination of electrons within conduction band and holes within valence band. Therefore, an interband laser is a bipolar device. The emission wavelength is inherently determined by the band gap E_g of the quantum well material, which is typically of the order of $1.24 \mu\text{m}$ ($\sim 1 \text{ eV}$), and can hardly be designed based on interband transition. Moreover, due to the opposite curvature of two bands associated with the laser transition in the k -space, the gain spectrum is broad and asymmetric. Therefore, an interband laser has a larger linewidth enhancement factor which will cause a large linewidth broadening of laser field. In addition, the nonradiative transition lifetime is typically long and order of nanoseconds due to the large energy separation (larger than one phonon energy) between these two bands in interband lasers. This will induce a different intrinsic linewidth characteristic from the QCLs.

In contrast, as shown in Fig. 1 - 1 (b), the intersubband transition occurs within conduction band and only electrons involve laser transition. Thus, an intersubband laser is a unipolar device in nature. The emission wavelength is determined by the thickness of quantum wells, and theoretical limitation depends on band offset. The present emission wavelength of intersubband lasers covers from the mid-IR ($3\text{-}24 \mu\text{m}$) to THz ($1.2\text{-}5 \text{ THz}$, or $60\text{-}250 \mu\text{m}$) ranges. The intersubband transition has an atomic-like joint density of states and the two subbands associated with laser transitions have the same curvature, therefore an intersubband laser has a narrow and symmetric gain linewidth resulting in a very small linewidth enhancement factor and thus a narrower intrinsic linewidth. In addition, the intersubband transition has a much smaller nonradiative emission lifetime (order of picoseconds) which is three orders of magnitude shorter than that of interband transition. This fast nonradiative relaxation in intersubband transition results from the fast intersubband polar longitudinal-optical (LO) phonon scattering mechanism. Therefore, the population inversion is limited by this small subband lifetime. In order to achieve enough optical gain for lasing, the intersubband laser is typically made of multiple cascaded structures. A trade off is that this short fast relaxation of carriers allows high frequency modulation without relaxation oscillations.

Fig. 1 - 1 Schematic of interband and intersubband transitions in a quantum well. The right diagrams show the energy dispersion curves in in-plan momentum space. (a) Interband transition from conduction band to valence band. E_{fc} and E_{fv} are the Fermi-Dirac distribution in conduction band and valence band. (b) Intersubband transition within conduction band. E_f is the Fermi-Dirac distribution.

1.1.2 Mid-infrared quantum cascade lasers

The concept of intersubband transition for light amplification was firstly proposed by Kazarinov and Suris in 1971 in a superlattice structure [17]. The superlattice, first described by Esaki and Tsu in 1970 [18], is formed by a periodic modulation of the conduction band edge in a repeated quantum well and barrier. After few years in 1989, Helm *et al* firstly observed the intersubband emission through resonant tunneling in a superlattice [19]. In order to achieve the lasing, many proposals are put forward [20-23]. Until 1994, the first successful intersubband laser, named as quantum cascade laser, was invented by Faist *et al* [10]. The emission wavelength is 4.2 μm . The device operated in pulse and required a cryogenic cooling. Figure 1 - 2 shows the conduction band profile and squared magnitude wavefunctions. The lasing transition

takes place between level 3 and level 2 and it is diagonal in real space. The energy separation between level 2 and level 1 equals the LO-phonon resonance energy so as to fast depopulate the electrons of subband 2. Since the energy separation between level 3 and level 2 is designed to be larger than one LO-phonon resonance energy, the relaxation of electrons from level 3 to level 2 is low due to the large in-plane momentum exchange [10]. Thus the population inversion between level 3 and level 2 is achieved. The electrons are injected into level 3 and extracted from level 1 through a doped quasi-classical multi-well superlattice region formed by the digitally graded alloy. By cascading N periods, the extracted electron from level 1 can be reinjected into the level 3 and recycling N times. Thus one electron can emit N photons, which allows for the quantum efficiency larger than one.

Fig. 1 - 2 Conduction band profile and squared magnitude wavefunctions of the first intersubband laser (named as quantum cascade laser) in a semiconductor superlattice. The emission wavelength is 4.3 μm . The figure is reproduced from Ref. [10].

Since the invention, QCLs have become the important mid-IR coherent source and shows an impressive development by improving the active region, e. g. bound-to-continuum and two-phonon resonance scheme [24, 25], and waveguide designs. The emission wavelength covers from 3-24 μm [5]. Most of wavelength can operate with continuous-wave (cw) mode at room temperature. The optical output power of order of 100 mW is available for cw operation [24]. Single-model distributed-feedback QCLs have been also demonstrated [26]. Nowadays, mid-IR QCLs become a well-developed coherent laser system.

1.1.3 Terahertz quantum cascade lasers

It is more difficult to achieve the terahertz lasing in quantum cascade structure than obtain mid-infrared lasing. There are two primary reasons. One is the difficulty of achieving a population inversion. Since the subband separation between the upper laser and lower laser levels is much smaller, it will be a challenge to selectively inject electrons into upper laser level but not lower laser level. It is also difficult to selectively depopulate the lower laser level while still maintaining a large upper state population for THz QCLs design. The other is the challenge to obtain a low-loss waveguide for such a long wavelength due to free-carrier absorption. The first THz QCL was demonstrated by Kohler in 2002 [15]. The structure used a chirped superlattice and a low-loss semi-insulating surface Plasmon waveguide. The device operated at 4.4 THz and pulsed operation was observed up to a heat-sink temperature of 50 K. Figure 1 - 3 shows the active region design and mode pattern. The chirped superlattice forms a large inter-miniband radiative dipole matrix element due to vertical transition and a greater spread of the wavefunctions in real space [15]. The optical transition occurs between level 2 and level 1. The electrons in the ground state g is injected into upper laser level 2 via resonant tunneling. The fast depopulation of electrons in level 1 is achieved by the electron-electron scattering and resonant tunneling within the miniband, thus the population inversion is achieved. The semi-insulating surface Plasmon waveguide is used by growing the active region of QCLs on a semi-insulating substrate and making the bottom highly doped contact layer much thinner. Since the mode overlap with n-doped regions is small, the free carrier loss is reduced.

Fig. 1 - 3 (a) Active region design for the first THz QCLs. (b) Mode pattern for semi-insulating surface Plasmon waveguide. The figure is reproduced from Ref. [15].

Since the invention, THz QCLs go through a fast development. The main task and challenge are to achieve the room-temperature operation [14]. The use of metal-metal waveguide has provided near unity confinement of resonant optical field [14]. Furthermore, till now, three designs for multiple quantum well active regions have been successfully used for THz QCLs (Fig. 1 - 4): chirped superlattice, band to continuum, resonant phonon and indirect pumping (or scattering-assisted injection). The first successful THz QCLs used the chirped superlattice designs, and have been described above. The bound-to-continuum design was shortly used for THz QCLs after the first THz QCL was demonstrated by Scalari *et al* in 2003 [27]. The structure showed the pulsed operation up to 100 K. The bound-to-continuum (BTC) design also uses the miniband to depopulate the electrons in lower laser level, but the upper level state is a more localized state. Therefore, in BTC, the optical transition is somehow diagonal. This can reduce the nonradiative scattering of the upper laser level into the miniband and also help reduce the coupling between the injector with lower laser level. The second successful design is based on resonant-phonon (RP) depopulation mechanism, which was firstly demonstrated by Williams *et al* in 2003 [28]. This RP design presently has been widely used. It is basically different from the above mentioned two designs. The depopulation of electrons in lower laser level does not depend on the miniband, but is achieved by resonant tunneling and LO phonon scattering. In this design, the lower laser level is in resonant with the extraction level in the adjacent quantum well. The extraction level state is designed to spread over few quantum wells and then emit LO-phonon in the phonon well [14]. The RP depopulation scheme via LO-phonon scattering is fast and can reduce the thermal backfilling into upper laser level due to spatial separation between the laser and phonon wells. The upper laser level is spatially separated from the injector level, which induce a reduce overlap with the injector level. Therefore, electrons can be effectively injected into upper laser level and the parasitic relaxation is minimized. This RP design for THz QCLs has exhibited the best temperature performance among other designs. The maximum operating temperature up to 200 K at pulsed mode was achieved in 2012 [29].

Figure 1 - 4 (d) shows the new-developed active region scheme *i.e.* indirect pumping (or scattering-assisted injection) for THz QCLs [30-35]. Since in THz QCLs, the energy separation between the upper and the lower laser levels is relatively small ($\sim 4\text{-}20$ meV), it is difficult to selectively inject electrons only into the upper laser level. Therefore, the injector barrier in resonant-tunneling injection QCLs is relatively thick so that off-resonant tunneling into states

other than the upper laser level is suppressed. Typically, the width of the injector barrier is chosen such that the anti-crossing splitting of the injection state and the upper laser level $2\hbar\Omega_{iu}$ (Ω_{iu} is the Rabi frequency of the coupled levels) is smaller than $\hbar\Delta\nu_{scatt}$, i.e. the FWHM linewidth due to scattering [36]. The disadvantage of a small Ω_{iu} and consequently a weak coupling between the injector and the upper laser level is the inefficient injection of electrons into the upper laser level. Based on this resonant-tunneling injection scheme, the maximum operating temperature of THz QCLs turned out to be limited by an empirical relation $T_{max} \sim \hbar\omega / \kappa_B$ (ω is the laser frequency, κ_B is the Boltzmann constant) [14, 37]. The indirect pumping scheme has recently managed to surpass this empirical limit [30-33]. Such indirect pumping scheme can be traced back as early as in 2001 by Scamarcio et al [38]. In this scheme, as shown in Fig. 1 - 4 (d), electrons are injected into the upper laser level by resonantly emitting phonons from the injector level. In this case, off resonant tunneling from the injector level to the lower laser level is well suppressed. Therefore, it is expected that the injector barriers can be kept thinner than those in the resonant injection scheme, since a pronounced tunneling through the injector barrier does not broaden the laser transition. The indirect pumping scheme may therefore offer a venue for high-performance THz QCLs.

Fig. 1 - 4 Schematics for successful THz QCLs active region design. (a) Chirped superlattice. (b) Bound to continuum. (c) Resonant phonon. (d) Indirect pumping (or scattering-assisted injection).

The present best operating temperature performance is up to ~200 K for THz QCLs [29]. For higher temperature operation, the main challenge comes from the thermal activated LO phonon scattering between the upper laser and lower laser level, which can greatly reduce the population inversion. Additionally, due to the closely spaced subbands, it is also a challenge to suppress the parasitic transport channels from the injector levels into other undesired levels. Such parasitic current will be enhanced at higher temperature. The endeavor to achieve THz QCLs operating above the temperature accessible by thermoelectric cooler is still ongoing.

1.2 Motivations

Since the birth of “quantum cascade laser”, it has been applied into many areas e.g. including but not limited to: trace-gas absorption spectroscopy, optical free-space data communication, remote sensing and imaging, and local oscillators for submillimeter-wave astronomy [12, 14, 26, 37, 39-48]. For these applications, a narrow-linewidth, single-mode, coherent and compact source is highly desired due to their very stable frequency. Understanding of the frequency stability of QCLs is important and advantageous for applications.

Like all other lasers, QCLs can easily suffer from various unwanted disturbance (so-called noise). The noise can induce the linewidth broadening and frequency instability, and therefore degrade performance of a laser. The linewidth of a laser is determined by the combined effects of various intrinsic noises together with various external noises. External noise factors such as mechanical vibrations, external environmental temperature variations and bias-current fluctuations can be potentially totally removed by various frequency-stabilization techniques, such as phase locking techniques [49-52]. Intrinsic noise, e.g. spontaneous emission, thermal photon and fundamental temperature fluctuation, is inherent to the laser system and it is impossible to reduce them unless the parameters of the laser system are adjusted. Therefore, investigation on intrinsic fluctuations induced linewidth broadening becomes very important to determine the ultimate sensitivity of QCLs system.

1.3 Challenges

Study of intrinsic linewidth has been widely reported for conventional semiconductor diode lasers [53-58]. However, QCLs are different from semiconductor diode lasers in many important ways which can have a significant impact on their noise properties.

Firstly, electron transport in diode laser takes places mainly by scattering process. But electron transport across the whole active region in QCLs depends on both resonant tunneling and scattering between states. It is well known that the resonant tunneling exhibits an significant impact on the electrical and optical properties of QCLs [36]. Therefore, the linewidth investigation should take into account this critical tunneling feature of QCLs. As an example, in conventional semiconductor lasers the carrier density in two energy states of laser transitions does not rises beyond its threshold value, hence, the linewidth broadening will not be influenced by the nonradiative recombination processes beyond threshold. However, in QCLs, the electron densities in the upper and lower laser level do not clamp at threshold, but keep increasing as the external bias exceeds its threshold value [5]. As a result, nonradiative processes influence significantly the laser linewidth even at high bias currents. Hence noise models of conventional semiconductor lasers cannot be directly applied into QCLs due to their different operational principles. New model needs to be developed specifically for QCLs.

Secondly, QCLs have a more complicated waveguide structure from the conventional semiconductor lasers, the feature of fundamental thermal noise, which has a great dependence on the waveguide structure, can be different from that of conventional semiconductor lasers, and should be carefully investigated.

Finally, because QCL is a complicated many-body system, in addition to the critical transport mechanism i.e. resonant tunneling, the various scattering and Coulomb interactions further complicate the electron transports. These characteristics require a quantitative insight into the details of the nonequilibrium carrier dynamics and energy relaxation as well as dephasing processes. The combined effects of many body interaction, nonparabolicity and coherence of resonant tunneling can have a significant influence on the optical properties of QCLs, and thus the linewidth enhancement factor.

The understanding of noise and linewidth of QCLs is still in its infancy, we aim to systematically investigate and demonstrate the intrinsic linewidth of QCLs with the consideration of the special electron transport characteristics so as to help us to optimize the system parameters for narrow-linewidth operation in this work.

1.4 Objectives and methodologies

We aim to develop theoretical models to solve the above mentioned challenges in QCLs, which also constitutes the main objectives of this thesis:

- (1). Study the effects of the critical electron transport characteristics on the optical properties of QCLs. In this thesis, these effects are theoretically evaluated by incorporating electron-electron interaction, nonparabolicity and coherence of resonant tunneling transport in a quantitative way based on the density matrix theory. Based on these analyses, their influences on intrinsic linewidth are investigated. Specifically, the effects of resonant-tunneling transport and dynamics of coherent interaction on intrinsic linewidth caused by spontaneous emission and thermal photon are investigated based on quantum Langevin equations.
- (2). Study the fundamental thermal frequency noise and linewidth broadening caused by the intrinsic temperature fluctuations. We conduct the analytical derivations based on the Green function analysis and the *Van Vliet-Fassett* theory.
- (3). Investigate the linewidth enhancement factor in QCLs why such factor is macroscopically theoretically predicted to be zero but experimentally measured to be non-zero. We develop the microscopic density matrix model to analyze the linewidth enhancement factor taking account of the many body Coulomb interaction, coherence of resonant-tunneling transport and non-parabolicity.

1.5 Thesis overview

This thesis presents a comprehensive theoretical analysis on the intrinsic linewidth of both mid-IR and THz QCLs. The thesis is organized into following chapters: Chapter 2 introduces the theoretical foundation for the QCLs so as to better understand the operation principle. Chapter 3 investigates the optical gain by incorporating many-body Coulomb interaction, nonparabolicity and coherence of resonant tunneling transport in a quantitative way based on the density matrix theory. Optical gain is one of fundamental properties that govern operation of quantum cascade lasers and is the bridge of amplitude-phase coupling (*i.e.* linewidth enhancement factor). The calculations demonstrate the importance of these parameters on optical properties, especially the optical gain spectrum, of THz and mid-IR QCLs. Chapter 4 analyzes the intrinsic linewidth caused by spontaneous emission and thermal photon on the basis of the quantum Langevin approach. The model includes the effects of resonant tunneling and coherent interactions on linewidth. Chapter 5 theoretically investigates the fundamental thermal frequency noise and linewidth broadening caused by intrinsic temperature fluctuations in both THz and mid-IR QCLs. The analytical derivation is based on the Green function analysis and the *Van Vliet-Fassett*

theory. Chapter 6 reports the microscopic density matrix analysis on the linewidth enhancement factor (LEF) of both mid-IR and THz QCLs, taking account of the many body Coulomb interaction, coherence of resonant-tunneling transport and non-parabolicity. A non-zero LEF at the gain peak is obtained due to these combined effects. Chapter 7 summarizes the works and provides some proposals for future works.

2 Theoretical foundations of intersubband transitions

2.1 Introduction

Quantum cascade laser (QCL) is a unipolar device, which is different from the conventional diode laser. In this chapter some basic theory and modeling necessary for designing the active region and understanding the operation principle of quantum cascade lasers are presented. We only consider the GaAs/Al_xGa_{1-x}As and In_xAl_{1-x}As/In_xGa_{1-x}As material system, and these materials are lattice-matched, hence the effects of strain on energy band and wavefunction are neglected. The chapter starts by presenting a more quantitative discussion of electronic states in heterostructures, followed by an analysis of the intersubband relaxation mechanisms. The final part of the chapter is devoted to a review of the most important transport mechanism *i.e.* resonant tunneling of electrons in QCLs.

2.2 Electronic states in heterostructures

A QCL is composed of multiple quantum wells and barriers with nano-scaled size. Its active region is a planar semiconductor heterostructure. Due to the quantum confinement, localized and quantized electron subbands along the growth direction are created within the conduction band edge profile.

The electron wavefunction can be described within the envelope function approximation as [59]

$$\Psi(\mathbf{r}) = F(\mathbf{r})U_0(\mathbf{r}) \quad (2.1)$$

where $U_0(\mathbf{r})$ is the Bloch state wavefunction at the band minimum, and $F(\mathbf{r})$ is the envelope function, which satisfies the following effective mass Schrodinger equation as

$$\left[-\frac{\hbar^2 \nabla_{\parallel}^2}{2m^*(z)} - \frac{\hbar^2}{2} \frac{\partial}{\partial z} \frac{1}{m^*(z)} \frac{\partial}{\partial z} + V(z) \right] F(\mathbf{r}) = EF(\mathbf{r}) \quad (2.2)$$

where $m^*(z)$ is the spatially varying effective mass and $V(z)$ is the conduction band edge profile including applied external electric field and local electric field due to space charge. In order to solve the above equation, the envelope function is firstly expressed as

$$F(\mathbf{r}) = \frac{1}{\sqrt{S}} e^{i\mathbf{k}_{\parallel} \cdot \mathbf{r}_{\parallel}} \varphi_n(\mathbf{k}_{\parallel}, z) \quad (2.3)$$

where S is the normalization area. Then we need to solve the following equation

$$\left[-\frac{\hbar^2}{2} \frac{\partial}{\partial z} \frac{1}{m^*(z)} \frac{\partial}{\partial z} + V(z) + \frac{\hbar^2 \mathbf{k}_{\parallel}^2}{2m^*(z)} \right] \varphi_n(\mathbf{k}_{\parallel}, z) = E_n(\mathbf{k}_{\parallel}) \varphi_n(\mathbf{k}_{\parallel}, z) \quad (2.4)$$

where $\mathbf{k}_{\parallel}$ is the in-plane wavevector, and n is the index of subband. By neglecting the coupling between the in-plan and z directions, which is usually small, equation (2.4) becomes

$$\left[-\frac{\hbar^2}{2} \frac{\partial}{\partial z} \frac{1}{m^*(z)} \frac{\partial}{\partial z} + V(z) \right] \varphi_n(z) = E_n \varphi_n(z) \quad (2.5)$$

where the total energy is given as

$$E_n(\mathbf{k}_{\parallel}) = E_n + \frac{\hbar^2 \mathbf{k}_{\parallel}^2}{2m^*} \quad (2.6)$$

where m^* is the effective mass in quantum wells.

In a QCL, the active region is usually optimized in such a way that electrons are forced to motion in the lower subband minima [59]. In this case, the in-plane motion of electrons around the conduction minima is only small extended, so that the band non-parabolicity can be neglected in most cases. However, this approximation can be improved by expanding the energy dispersion relation [60]. On the basis of this consideration, the energy band non-parabolicity is introduced through an energy dependent effective mass [59]

$$m^*(E) = m^*(0) [1 + \gamma(E - V)] \quad (2.7)$$

where $m^*(0)$ is the effective mass at the bottom of the conduction band, and γ is the non-parabolicity coefficient, which is.

$$\gamma = [1 - m^*(0)/m_0]^2 / E_g \quad (2.8)$$

where E_g is the band gap.

In Eq. (2.5), we included the effects of applied external electric field and local electric field due to space charge on potential. These effects can be described by using Poisson's equation as

$$\frac{d}{dz} \left[\varepsilon(z) \frac{d}{dz} \Phi(z) \right] = -\rho(z) \quad (2.9)$$

where $\Phi(z)$ is the electrostatic potential, $\varepsilon(z)$ is the spatially varying permittivity and $\rho(z)$ is the charge density. The solution of Poisson's equation shows us the total potential $V(z) = V_{c,0}(z) - e\Phi(z)$, where $V_{c,0}(z)$ is the intrinsic conduction band profile. By the self-consistent solution of the Schrodinger and Poisson's equations using the iterative method, the conduction band profile and wavefunctions of a QCL can be obtained.

2.3 Intersubband radiative and non-radiative transition

2.3.1 Intersubband radiative transition

The following section reviews the intersubband radiative transition, and the treatment is standard [61, 62].

The light-matter interaction is the core of an optical device. Quantum mechanically, this interaction can be described by the Hamiltonian

$$H' = -\frac{e}{m^*} \mathbf{A} \cdot \mathbf{p} \quad (2.10)$$

where p is momentum and m^* is the effective mass of quantum wells and band non-parabolicity is neglected. A is the Lorentz-gauge vector potential and is given by

$$\mathbf{A} = \sqrt{\frac{\hbar}{2\varepsilon\omega_q V}} \hat{\mathbf{e}}_{\mathbf{q},\eta} \left[a_{\mathbf{q},\eta} e^{i\mathbf{q}\cdot\mathbf{r}} + a_{\mathbf{q},\eta}^\dagger e^{-i\mathbf{q}\cdot\mathbf{r}} \right] \quad (2.11)$$

where V is the volume of the cavity, $\hat{\mathbf{e}}_{\mathbf{q},\eta}$ is the polarization vector of light with wavevector $\mathbf{q}$ at frequency ω_q . $\eta=1, 2$ is the polarization state. $a_{\mathbf{q},\eta}$ and $a_{\mathbf{q},\eta}^\dagger$ are the creation and annihilation operators of photon, respectively.

According to Fermi's Golden Rule, the intersubband radiative transition from initial state $|i, m_{\mathbf{q},\eta}\rangle$ to final state $|f, n_{\mathbf{q},\eta}\rangle$ within conduction subbands is given as

$$W_{i \rightarrow f} = \frac{2\pi}{\hbar} \left| \langle f, n_{\mathbf{q},\eta} | H' | i, m_{\mathbf{q},\eta} \rangle \right|^2 \delta(E_f(\mathbf{k}_f) - E_i(\mathbf{k}_i) \pm \hbar\omega_q) \quad (2.12)$$

where $n_{\mathbf{q},\eta}$ and $m_{\mathbf{q},\eta}$ are photon numbers in each mode.

By inserting Eq. (2.11) into Eq. (2.12), the transition matrix element becomes

$$\left| \langle H'_{i \rightarrow f} \rangle \right|^2 = \left(\frac{e^2 \hbar}{2m^* \varepsilon \omega_q V} \right) \left[m_{\mathbf{q},\eta} \delta_{m_{\mathbf{q},\eta}-1, n_{\mathbf{q},\eta}} \left| \langle f | e^{i\mathbf{q}\cdot\mathbf{r}} \hat{\mathbf{e}}_{\mathbf{q},\eta} \cdot \mathbf{p} | i \rangle \right|^2 + (m_{\mathbf{q},\eta} + 1) \delta_{m_{\mathbf{q},\eta}+1, n_{\mathbf{q},\eta}} \left| \langle f | e^{-i\mathbf{q}\cdot\mathbf{r}} \hat{\mathbf{e}}_{\mathbf{q},\eta} \cdot \mathbf{p} | i \rangle \right|^2 \right] \quad (2.13)$$

The first term represents the optical absorption, and second term includes both stimulated emission (portioned to $m_{\mathbf{q},\eta}$) and spontaneous emission (independent on optical field) processes.

In real space, the projection of initial $|i, m_{\mathbf{q},\eta}\rangle$ and final $|f, n_{\mathbf{q},\eta}\rangle$ states can be expressed as by using the transverse and longitudinal envelop function approximation

$$\langle \mathbf{r} | i \rangle = \frac{1}{\sqrt{S}} e^{i\mathbf{k}_{\parallel,i} \cdot \mathbf{r}_{\parallel}} \varphi_i(z) \quad (2.14)$$

$$\langle \mathbf{r} | f \rangle = \frac{1}{\sqrt{S}} e^{i\mathbf{k}_{\parallel,f} \cdot \mathbf{r}_{\parallel}} \varphi_f(z) \quad (2.15)$$

Assuming the cell periodic part rotates more rapidly than the envelope part, we can adopt the dipole approximation $e^{i\mathbf{q} \cdot \mathbf{r}} \approx 1$ [63]. Then the dipole interaction between the conduction subbands can be expressed as

$$\begin{aligned} \langle f | e^{\pm i\mathbf{q} \cdot \mathbf{r}} \hat{\mathbf{e}}_{\mathbf{q},\eta} \cdot \mathbf{p} | i \rangle &\approx \langle f | \hat{\mathbf{e}}_{\mathbf{q},\eta} \cdot \mathbf{p} | i \rangle \\ &= (\hat{\mathbf{e}}_{\mathbf{q},\eta} \cdot \mathbf{k}_{\parallel,i}) \frac{\hbar}{S} \int d\mathbf{r}_{\parallel} e^{i(\mathbf{k}_{\parallel,i} - \mathbf{k}_{\parallel,f}) \cdot \mathbf{r}_{\parallel}} \int dz \varphi_f^*(z) \varphi_i(z) \\ &\quad - \hat{\mathbf{z}} \frac{i\hbar}{S} \int d\mathbf{r}_{\parallel} e^{i(\mathbf{k}_{\parallel,i} - \mathbf{k}_{\parallel,f}) \cdot \mathbf{r}_{\parallel}} \int dz \varphi_f^*(z) \frac{\partial \varphi_i(z)}{\partial z} \end{aligned} \quad (2.16)$$

Because the envelope functions between φ_i and φ_j are orthogonal, the first integral equals to zero. Therefore, we achieve a well-known intersubband selection rule for dipole interaction: only transitions with the optical field polarized along the growth direction are permitted. Then interaction is

$$\langle f | e^{\pm i\mathbf{q} \cdot \mathbf{r}} \hat{\mathbf{e}}_{\mathbf{q},\eta} \cdot \mathbf{p} | i \rangle \approx (\hat{\mathbf{e}}_{\mathbf{q},\eta} \cdot \hat{\mathbf{z}}) \delta \langle \varphi_f | p_z | \varphi_i \rangle \quad (2.17)$$

where Eq. (2.17) can rewritten in terms of unperturbed Hamiltonian H_0 . Assuming the perturbed Hamiltonian H' can be expressed in terms of dipole interaction, then, for unperturbed Hamiltonian H_0 , we have

$$\frac{i}{\hbar} [H_0, z] = \frac{p_z}{m^*} \quad (2.18)$$

So we obtain

$$\langle f | p_z | i \rangle = \frac{im^*}{\hbar} (E_f - E_i) \langle \varphi_f | z | \varphi_i \rangle \quad (2.19)$$

where the overlap integral $\langle \varphi_f | z | \varphi_i \rangle$ is the dipole matrix element $z_{i \rightarrow f}$. By inserting Eq. (2.19) into Eq. (2.13) and Eq. (2.12), we get the following spontaneous and stimulated emission rates per unit volume at a given photon mode

$$W_{i \rightarrow f}^{sp} \Big|_{\text{mode}} = \frac{\pi e^2 \omega_{\mathbf{q}}}{\varepsilon V} |\hat{\mathbf{e}}_{\mathbf{q},\eta} \cdot \hat{\mathbf{z}}|^2 |z_{i \rightarrow f}|^2 \delta(E_f - E_i + \hbar \omega_{\mathbf{q}}) \quad (2.20)$$

$$W_{i \rightarrow f}^{st} \Big|_{\text{mode}} = \frac{\pi e^2 \omega_{\mathbf{q}}}{\varepsilon V} m_{\mathbf{q},\eta} |\hat{\mathbf{e}}_{\mathbf{q},\eta} \cdot \hat{\mathbf{z}}|^2 |z_{i \rightarrow f}|^2 \delta(E_f - E_i + \hbar \omega_{\mathbf{q}}) \quad (2.21)$$

2.3.1.1 Spontaneous emission rate

In order to get the total spontaneous emission rate, the sum over all the photon modes and polarization should be calculated. Provided that the cavity dimensions of a QCL is much larger than the emission wavelength, the mode number in a differential volume $d^3 \mathbf{q}$ can be expressed as

$$\rho(\mathbf{q}) d^3 \mathbf{q} = \frac{d^3 \mathbf{q}}{8\pi^3/V} = \frac{q^2 dq \sin \theta d\theta d\phi V}{8\pi^3} \quad (2.22)$$

Then the total spontaneous emission rate is obtained as by the integral of Eq. (2.20) over all the cavity modes

$$\begin{aligned} W_{i \rightarrow f}^{sp} &= \frac{e^2 |z_{i \rightarrow f}|^2}{8\hbar \pi^2 \varepsilon} \int_0^\infty \int_0^\pi \int_0^{2\pi} \left(\frac{n\omega_{\mathbf{q}}}{c} \right)^3 \sin^3 \theta \delta(E_f - E_i + \hbar \omega_{\mathbf{q}}) d\hbar \omega d\theta d\phi \\ &= \frac{e^2 n \omega_0^3}{3\pi \varepsilon_0 \hbar c^3} |z_{i \rightarrow f}|^2 = \frac{e^2 n \omega_0^3}{6\pi m^* \varepsilon_0 c^3} f_{i \rightarrow f} \end{aligned} \quad (2.23)$$

where n is the index of refraction with frequency $\omega_0 = (E_f - E_i)/\hbar$, and c is the light speed in a vacuum. Here, the spontaneous emission rate is expressed as the oscillator strength $f_{i \rightarrow f}$

$$f_{i \rightarrow f} = \frac{2m^* (E_f - E_i)}{\hbar^2} |z_{i \rightarrow f}|^2 \quad (2.24)$$

The oscillator strength is proportional to the optical gain and characterizes the strength of a transition. It satisfies the Thomas-Reiche-Kuhn sum rule if the spatial dependence of effective mass and non-parabolicity are not considered [59]

$$\sum_{f \neq i} f_{i \rightarrow f} = 1 \quad (2.25)$$

The modification to this rule due to spatially dependent effective mass and non-parabolicity can be found in Ref. [64].

A typical spontaneous emission time of a QCL is on the order of microseconds, which is much longer than the nonradiative lifetime (it is on the order of picoseconds). Therefore, the spontaneous emission has no influence on transition and transport of electrons. It is also the reason that quantum cascade lasers have a much smaller intrinsic linewidth than that of diode laser.

2.3.1.2 Stimulated emission rate

Equation (2.21) expressed the stimulated emission rate with a single photon mode. This equation can help us calculate the stimulation emission rate induced by an incident monochromatic wave with frequency ν .

By considering the homogeneous broadening of transition, we can replace the Dirac delta-function with the following Lorentzian lineshape function

$$\gamma(\nu) = \frac{(\Delta\nu/2\pi)}{(\nu - \nu_0)^2 + (\Delta\nu/2)^2} \quad (2.26)$$

where $\Delta\nu$ is the full-width half-maximum linewidth and is the function of initial lifetime τ_i , final state lifetime τ_f and pure dephasing time T_2^*

$$\Delta\nu = \frac{1}{\pi} \left(\frac{1}{2\tau_i} + \frac{1}{2\tau_f} + \frac{1}{T_2^*} \right) \quad (2.27)$$

By incorporating the intensity of incident wave $I_\nu = cm_\ell h\nu_\ell / nV$ with photon number m_ℓ in the mode frequency ν_ℓ , the stimulated emission rate becomes

$$W_{i \rightarrow f}^{st} = \frac{3|z_{i \rightarrow f}|^2}{|r_{i \rightarrow f}|^2} \frac{\lambda^2 I_\nu}{8\pi h \nu n^2 \tau_{i \rightarrow f}^{sp}} \gamma(\nu) \sim \frac{3\lambda^2 I_\nu}{8\pi h \nu n^2 \tau_{i \rightarrow f}^{sp}} \gamma(\nu) \quad (2.28)$$

2.3.2 Nonradiative transition

The population inversion is a necessary condition for the light emission in a laser. For QCLs, we can design the active region to allow the scattering rate of electrons to satisfy this condition. Therefore, in addition to radiative transition, a thorough understanding of the non-radiative transition is essential for optimizing the design and improving the performance of QCLs. There

are two kinds of non-radiative transition mechanism i.e. polar longitudinal optical (LO) phonon scattering and electron-electron (e-e) scattering. In this section, we will review the theoretical aspects of these two kinds of scattering. The treatment in this section is directly followed the discussion given by Williams [62].

2.3.2.1 Polar longitudinal optical phonon scattering

The polar longitudinal optical (LO) phonon scattering is a dominate relaxation channel and dephasing sources of decoherence for the electron transport in a QCL when the energy separation of conduction subband is larger than the LO phonon energy. Electron-LO phonon scattering can occurs both between intersubbands and within intrasubbands. The electron-LO phonon scattering rate from the initial state $|i\rangle$ to final state $|f\rangle$ through an interaction potential H can be expressed by the Fermi's Golden Rule

$$W_{i \rightarrow f}(\mathbf{k}_i, \mathbf{k}_f) = \frac{2\pi}{\hbar} \left| \langle f, \mathbf{k}_f | H | i, \mathbf{k}_i \rangle \right|^2 \delta(E_f(\mathbf{k}_f) - E_i(\mathbf{k}_i) \pm \hbar\omega_{LO}) \quad (2.29)$$

where the electron-phonon interaction Hamiltonian H is

$$H = \sum_{\mathbf{q}} \left[\alpha(q) (e^{i\mathbf{q} \cdot \mathbf{r}} b_{\mathbf{q}} + e^{-i\mathbf{q} \cdot \mathbf{r}} b_{\mathbf{q}}^\dagger) \right] \quad (2.30)$$

$b_{\mathbf{q}}$ and $b_{\mathbf{q}}^\dagger$ are the creation and annihilation operators of electron, respectively. $\alpha(q)$ is the Frohlich electron-LO phonon interaction strength. Considering the bulk phonon approximation, this strength is given by

$$|\alpha(q)|^2 = \frac{\hbar\omega_{LO}}{2} \frac{e^2}{q^2} \left(\frac{1}{\epsilon_\infty} - \frac{1}{\epsilon_0} \right) \quad (2.31)$$

where ϵ_∞ and ϵ_0 are the high and static frequency permittivity. Then the matrix element of phonon absorption (-) and emission (+) becomes

$$\left| \langle f, \mathbf{k}_f | H | i, \mathbf{k}_i \rangle \right|^2 = \frac{\hbar e^2 \omega_{LO}}{2V} \left(\frac{1}{\epsilon_\infty} - \frac{1}{\epsilon_0} \right) \frac{1}{q_z^2 + q_{\parallel}^2} |A_{i \rightarrow f}(q_z)|^2 \delta_{\mathbf{k}_i, \mathbf{k}_f \mp \mathbf{q}_{\parallel}} (n_{\omega_{LO}} + 1/2 \mp 1/2) \quad (2.32)$$

where q_z and $q_{\parallel}$ is the z -direction and in-plan (x - y plane) phonon wavevectors. $n_{\omega_{LO}}$ is the Bose-Einstein distribution of phonon. $\delta_{\mathbf{k}_i, \mathbf{k}_f \mp \mathbf{q}_{\parallel}}$ ensures in-plane momentum conservation and the form factor

$$A_{i \rightarrow f}(q_z) = \int_{-\infty}^{+\infty} dz \varphi_f^*(z) \varphi_i^*(z) e^{\pm q_z z} \quad (2.33)$$

The electron-LO phonon scattering satisfies the in-plane momentum and energy conservation between the initial and final states, as shown in Fig. 2 - 1. Without the consideration of non-parabolicity, they are, respectively

$$q_{\parallel}^2 = |\mathbf{k}_i - \mathbf{k}_f|^2 = k_f^2 + k_i^2 - 2k_i k_f \cos \theta \quad (2.34)$$

$$k_f^2 = k_i^2 + \frac{2m^* (E_f(0) - E_i(0) \mp \hbar \omega_{LO})}{\hbar^2} \quad (2.35)$$

Fig. 2 - 1 (a) Schematics for the intersubband electron-LO phonon emission scattering. (b) The relation between the initial wavevector k_i , final wavevector k_f and phonon wavevector q in in-plan momentum.

By the integral of Eq. (2.29) over the final states k_f , the total electron-LO phonon scattering from the initial states k_i becomes

$$W_{i \rightarrow f}^{abs}(\mathbf{k}_i) = \frac{m^* e^2 \omega_{LO}}{8\pi \hbar^2} \left(\frac{1}{\epsilon_{\infty}} - \frac{1}{\epsilon_0} \right) n_{LO} \int_0^{2\pi} d\theta B_{i \rightarrow f}(q_{\parallel}) \quad (2.36)$$

$$W_{i \rightarrow f}^{abs}(\mathbf{k}_i) = \frac{m^* e^2 \omega_{LO}}{8\pi \hbar^2} \left(\frac{1}{\epsilon_{\infty}} - \frac{1}{\epsilon_0} \right) (n_{LO} + 1) \int_0^{2\pi} d\theta B_{i \rightarrow f}(q_{\parallel}) \quad (2.37)$$

where

$$B_{i \rightarrow f}(q_{\parallel}) = \int_{-\infty}^{+\infty} dz \int_{-\infty}^{+\infty} dz' \varphi_f^*(z) \varphi_i(z) \varphi_i^*(z') \varphi_f(z') \frac{1}{q_{\parallel}} e^{-q|z-z'|} \quad (2.38)$$

The total scattering time $\tau_{i \rightarrow f}$ between subbands can be gotten by the average of all possible initial states

$$\frac{1}{\tau_{i \rightarrow f}} = \frac{\int_0^\infty dE_k f_i(E_k) W_{i \rightarrow f}(E_k)}{\int_0^\infty dE_k f_i(E_k)} \quad (2.39)$$

where $E_k = \hbar^2 \mathbf{k}_\parallel^2 / 2m^*$ and $f_i(E_k)$ is the Fermi-Dirac distribution.

When the transition energy E_{if} is lower than the LO Phonon energy E_{LO} , the total scattering time $\tau_{i \rightarrow f}$ can be approximated using the thermally active expression [65]

$$\frac{1}{\tau_{i \rightarrow f}} \approx W_{i \rightarrow f}^{hot} \exp\left(\frac{E_{if} - E_{LO}}{k_B T}\right) \quad (2.40)$$

where $W_{i \rightarrow f}^{hot}$ is the scattering rate when LO-phonon scattering is energetically allowed.

2.3.2.2 Electron- electron scattering

Electron-electron (e-e) interaction is another important scattering mechanism. It can destroy the coherence of resonant tunneling and reduces the lifetime of electrons in the upper laser level. A theoretical calculation of the e-e scattering rate is complicated and a complete description needs many-body theorem. Nevertheless, Smet [66] analyzed the e-e scattering using the static single band approximation. According to this approximation, the unscreened matrix element from the initial electron state $|i\rangle$ to final state $|f\rangle$ is expressed as

$$H'_{i,j \rightarrow f,g}(\mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_f, \mathbf{k}_g) = \frac{2\pi e^2}{S\epsilon} A_{i,j \rightarrow f,g}(q_\parallel) \delta(\mathbf{k}_f + \mathbf{k}_g - \mathbf{k}_i - \mathbf{k}_j) \quad (2.41)$$

Where $q_\parallel$ and $A_{i,j \rightarrow f,g}$ are

$$q_\parallel = |\mathbf{k}_i - \mathbf{k}_f| \quad (2.42)$$

$$A_{i,j \rightarrow f,g}(q_\parallel) = \int_{-\infty}^{+\infty} dz \int_{-\infty}^{+\infty} dz' \phi_i(z) \phi_f^*(z) \phi_j(z') \phi_g^*(z') e^{-q|z-z'|} \quad (2.43)$$

Then the total scattering rate for an electron in state $|i\rangle$ is gotten by

$$\begin{aligned} W_{i,j \rightarrow f,g}(\mathbf{k}_i) = & \frac{e^4}{2\pi\hbar\epsilon^2} \int d^2\mathbf{k}_j \int d^2\mathbf{k}_f \int d^2\mathbf{k}_g \frac{|A_{i,j \rightarrow f,g}(q_\parallel)|^2}{\epsilon_{sc}^2(q_\parallel, T) q_\parallel^2} f_{j,k} (1 - f_{f,k}) (1 - f_{g,k}) \\ & \times \delta(E_f(\mathbf{k}_f) + E_g(\mathbf{k}_g) - E_i(\mathbf{k}_i) - E_j(\mathbf{k}_j)) \delta(\mathbf{k}_f + \mathbf{k}_g - \mathbf{k}_i - \mathbf{k}_j) \end{aligned} \quad (2.44)$$

Averaging over all possible initial state, the e-e scattering time $\tau_{i \rightarrow f}$ is given as

$$\frac{1}{\tau_{i \rightarrow f}} = \frac{\int_0^\infty dE_k f_i(E_k) W_{i,j \rightarrow f,g}(E_k)}{\int_0^\infty dE_k f_i(E_k)} \quad (2.45)$$

2.4 Resonant tunneling transport

Resonant tunneling is the most important and critical transport mechanism in quantum cascade lasers. The injection of electrons into upper laser level and extraction of electrons from the lower laser level is dependent on resonant tunneling. A full and well understanding of this transport mechanism is very important in design and optimization of a QCL for the high performance operation. It can help us determine the relevant parameters that control this process. The importance and role of coherent and incoherent transport of tunneling in QCLs have attracted a great attention in both theoretical and experimental area of research [36, 67-73]. In the section below I will review some basic aspects for the investigation of tunneling transport in QCLs.

2.4.1 General description

In order to demonstrate the importance of coherent transport in QC structures, we consider a simple superlattice, as shown in Fig. 2 - 2. In the coupled quantum wells, we assume that only two energy levels in each well, levels 1 and 2, are involved in the transport processes under the applied electric field. In semiclassical model, as illustrated in Fig. 2 - 2(a), the transport is described by in- and out- scattering from certain stationary energy states based on Boltzmann-like equations. The entire superlattice is dealt with as a single quantum mechanical system and has a well-defined Hamiltonian. All the subband energy and wavefunctions are eigenvalues and eigenstates of this Hamiltonian. In this model, the electron transport takes place via various scattering between these states as described by the Fermi's Golden Rule. No coherent interaction among these states is taken into account. This semiclassical model assumes that the wavefunctions of electrons are spatially extended. Under resonant bias conditions, the ground states 1' in one quantum well is aligned with the excited level of 2 of the adjacent quantum well. A spatially extended doublet with the symmetric wavefunction $|S\rangle$ with a lower energy as well as the asymmetric wavefunction $|A\rangle$ with higher energy is formed by the coupling of these two energy levels. The energy separation of the doublet at resonance is known as the anticrossing gap Δ_0 . Therefore, the transport of electrons through the barrier is instantaneous, and there is no

obstruction from the barrier and no tunneling time between these two adjacent quantum wells due to using sets of delocalized states. The whole transport time is determined by the scattering of electrons from the energy level $2'$ into doublet $|S\rangle$ and $|A\rangle$ or equivalently from the energy level doublet $|S\rangle$ and $|A\rangle$ into 1 . As a result, the current density at resonant bias is independent of the barrier thickness. This is only valid when the dephasing is very weak, and is thereby unphysical for real devices.

In contrast, as shown in Fig. 2 - 2(b), the localized basis states are used in tight-binding resonant tunneling model. In this model, the localized states $1'$ and 2 are eigenstates of left quantum well and right quantum well, respectively. At an initial time, the electron wavepacket resides at in the energy level $1'$. As time evolves, this wavepacket oscillates across the barrier at the Rabi oscillation frequency $\Delta_0/\hbar$. If there is no pure dephasing, this oscillation will be damped only by the intersubband scattering between the energy level 1 and 2 . The main bottleneck of the electron transport is again determined by the intersubband scattering although electrons transport across the barrier within a finite time (half of the oscillation period). However, when the dephasing, which indeed exists and is caused by the various intersubband and intrasubband scattering e.g. electron-phonon scattering and interface roughness scattering, is taken into account, the case is different. Since the dephasing scattering can greatly damp the Rabi oscillation, the time evolution of the electron wavepacket from energy level $1'$ into level 2 is no longer oscillatory. The bottleneck of the electron transport becomes the tunneling barrier. In this case, the simple semiclassical will be no longer a good approximation to investigate the electric and optical properties of a QCL system. It is experimentally verified that the measured current density is very sensitive to the thickness of injector barrier, and the simulation band semiclassical model tends to overestimate the current density and gain of QCLs [74, 75].

Fig. 2 - 2 Difference between semi-classical and coherent picture of coupled quantum wells. (a) Schematic for semiclassical model for resonant tunneling through a barrier. A spatially extended doublet with the symmetric wavefunction $|S\rangle$ with a lower energy as well as the asymmetric wavefunction $|A\rangle$ with higher energy is formed by the coupling of these two energy levels. (b) Schematic for tight-binding resonant tunneling model with interaction strength $\Delta_0/2$. The localized states $1'$ and 2 are eigenstates of left quantum well and right quantum well, respectively.

2.4.2 Density matrix model

The semiclassical is insufficient to describe coherent phenomena. To describe the time evolution and phase coherence of states, an efficient approach is the density matrix formalism [76, 77]. The density matrix model describes the statistical distribution of quantum states of a system and does not care about exact details of the individual electron states.

For an arbitrary electron wavefunction

$$|\varphi\rangle = \sum_n c_n |\phi_n\rangle \quad (2.46)$$

where ϕ_n is the basis wavefunctions of the Hamiltonian and c_n is

$$c_n = \langle \phi_n | \varphi \rangle \quad (2.47)$$

Then the density operator ρ is defined as in the form of a projection operator

$$\rho = |\varphi\rangle\langle\varphi| = \sum_m \sum_n c_n^* c_m |\phi_m\rangle\langle\phi_n| \quad (2.48)$$

For an ensemble of electrons, the density matrix elements are defined as the ensemble average as

$$\rho_{nm} = \langle \phi_m | \rho | \phi_n \rangle = \langle c_n^* c_m \rangle \quad (2.49)$$

The diagonal elements ρ_{ii} provide the information of the probability of finding the particles in state $|i\rangle$ and are state populations. The off-diagonal elements ρ_{ij} correspond to the average degree of coherence between states i and j . By taking the time derivative of the density operator, the time evolution of the density matrix is described as

$$i\hbar \frac{\partial \rho}{\partial t} = i\hbar \frac{\partial |\varphi\rangle}{\partial t} \langle \varphi| + i\hbar |\varphi\rangle \frac{\partial \langle \varphi|}{\partial t} = [\rho, H] \quad (2.50)$$

where H is the Hamiltonian of the system.

For example, for the two-level system, as shown in Fig. 2 - 2(b), we consider resonant tunneling from state 1' into state 2 with tight-binding coupling $\Delta_0/2$. The equation of motion in form of density matrix can be written as

$$\frac{\partial}{\partial t} \begin{pmatrix} \rho_{11} & \rho_{12} \\ \rho_{21} & \rho_{22} \end{pmatrix} = \left[\begin{pmatrix} \rho_{11} & \rho_{12} \\ \rho_{21} & \rho_{22} \end{pmatrix}, H \right] + \frac{\partial \rho}{\partial t} \Big|_{relax} \quad (2.51)$$

where $\partial \rho / \partial t|_{relax}$ is the phenomenological relaxation processes such as dephasing and population lifetimes. It is

$$\frac{\partial \rho}{\partial t} \Big|_{relax} = \begin{pmatrix} \frac{\rho_{11}}{\tau} & \frac{\rho_{12}}{\tau_{\parallel}} \\ \frac{\rho_{21}}{\tau_{\parallel}} & \frac{\rho_{22}}{\tau} \end{pmatrix} \quad (2.52)$$

H is

$$H = \begin{pmatrix} \langle \phi_1 | H | \phi_1 \rangle & \langle \phi_1 | H | \phi_2 \rangle \\ \langle \phi_2 | H | \phi_1 \rangle & \langle \phi_2 | H | \phi_2 \rangle \end{pmatrix} = \begin{pmatrix} E_{11} & -\Delta_0/2 \\ -\Delta_0/2 & E_{22} \end{pmatrix} \quad (2.53)$$

The tight-binding density matrix model can be a useful tool in evaluating the effect of wavefunction localization on transport through the barrier and has been widely used for the design and optimization of QCLs.

2.5 Conclusions

In this chapter, the basic theory of electron transition and transport in QCLs are presented. The Schrödinger equation for MQWs heterostructure is shown followed by the analysis of intersubband radiative and nonradiative transition. In QCLs, the electron-electron and electron-LO phonon interaction dominate the scattering processes of electrons. Finally, the critical

transport mechanism is evaluated. The effects of wavefunction localization on electron through the injector and extractor barriers are very important in simulating the quantum cascade lasers.

3 Optical gain of quantum cascade lasers: microscopic analysis

3.1 Introduction

As one of important coherent mid-IR [10] radiation sources, room-temperature continuous-wave operation of mid-IR QCLs have been achieved in the $\sim 3\text{-}14\text{ }\mu\text{m}$ wavelength range. However, due to the challenges on building up enough optical gain in long-wavelength emission devices above cryogenic temperatures [14], THz QCLs with wavelengths covering from 60 to 300 μm , are still operated below room temperatures. The best temperature performance has been obtained at $\sim 200\text{ K}$ using a resonant-phonon (RP) design [29]. Further improvement on the existing designs requires a better understanding of effects of the electron transport on optical properties, e.g. optical gain, of THz QCLs. Optical gain is one of fundamental properties that govern operation of quantum cascade lasers. In addition to the obvious interest in understanding the nature of optical gain of QCLs, they are important due to their relationship to amplitude-phase coupling factor (*i.e.* linewidth enhancement factor). The linewidth enhancement factor, which causes the laser intrinsic linewidth to increase well beyond the Schawlow-Townes formula, is well-determined by the lineshape of optical gain spectrum. They are also important due to their relationship to the laser gain nonlinearity [78]. For example, the gain nonlinearity dictates limitations to the modulation bandwidth. Furthermore, to design QCLs for expanding range of applications e.g. designing the active region to achieve broadband spectrum for applications of spectroscopy and atmospheric chemistry, it is necessary to be able to predict their gain spectra accurately. However, the usual approach studying the optical gain of QCLs by analogy with a two-level atomic system is inadequacy. In quantum cascade lasers, the optical transition takes place between quantized intersubband levels in multiple quantum well structure. The population inversion between the upper laser and lower laser levels is achieved by selective tunneling processes at operating bias, which is strongly different from that in the two-level atomic system. The coherence of resonant tunneling can greatly influence the electron transport in quantum cascade structures. Moreover, the various scattering and many-body interactions further complicate the electron transports. All these characteristics require a quantitative insight into the details of the nonequilibrium carrier dynamics and energy relaxation as well as dephasing processes. In this chapter, we presented a microscopic analysis on the optical gain of QCLs. This microscopic study is conducted by the well-defined density matrix model with the consideration

of many body Coulomb interaction, non-parabolicity and coherence of resonant tunneling. The analysis can help us understand the relative importance of these different factors on the electron transports in QCLs. It is also the theoretical foundation of following chapters.

3.2 Microscopic density matrix model: effects of many body, non-parabolicity and resonant tunneling

3.2.1 General description

Up to now, several useful theoretical models, *e.g.* Monte Carlo [74, 79-82], nonequilibrium Green's function [30, 35, 70, 83, 84] and simplified density-matrix [36, 71, 85] models, have been developed to predict the optical properties and electron transports of QCLs. Although Monte Carlo and nonequilibrium Green's functions analyses show good agreement between theories and experiments in some aspects, implementations of these two models are difficult, requiring intensive numerical computations. Alternatively, the simplified density-matrix model is simple in the analysis and requires much less computation load, while still captures the essentials of coherent effects such as the electron resonant-tunneling (RT) transport. It has been shown as one of the most promising candidates for the study of QCLs. This model is in essence a set of rate equations but including electron distributions and coherent dynamics in different subbands. Electrons in each subband are assumed to behave the same, regardless of their kinetic energies. Therefore, this model describes the optical properties and electron transport from a *macroscopic* point of view, while the *microscopic* phenomena *e.g.* the electron dynamics in the in-plane k -space are neglected. Moreover, the present experiments and theoretical predictions have shown the limitations of this macroscopic model. The gain peak frequency calculated by the macroscopic model is overestimated compared to the experimentally measured lasing frequency [29]. Although coherence effects of RT transport can be described in the macroscopic simplified density-matrix model [36], another aspect that needs to be considered for a more accurate calculation of optical properties and electron transport is the electron-electron Coulomb interaction.

Direct numerical treatment of many-body Coulomb interaction is complex and hence is often handled at the level of the Hartree-Fock approximation [86]. In this case, the set of motion equations, in terms of the diagonal and off-diagonal elements of the reduced single-particle density matrix, are well-known as the Hartree-Fock semiconductor Bloch equations which treat

Coulomb effects via bandstructure and Rabi frequency renormalizations [87]. For mid-IR QCLs, the effects of many-body Coulomb interactions on the optical properties and electron transports were considered but the role of coherence of resonant-tunneling transport was neglected [88]. For THz QCLs, many-body effects on population dynamics were investigated [89], but only the injection coupling was considered while the extraction coupling was neglected. Dupont *et al* [85] have demonstrated the importance of extraction coupling on optical properties according to the macroscopic simplified density-matrix model but without considering the microscopic properties such as the many-body Coulomb interactions. Since many-body Coulomb interactions result in bandstructure and Rabi frequency renormalizations, and Coulomb-induced subband coupling, they are expected to play an important role in QCLs. However, the effects of electron-electron Coulomb interactions on optical gain of QCLs have not yet been reported. In addition, the subband dispersion with different subband effective masses (commonly referred to as non-parabolicity) is known to be important for modifications to gain spectrum. Thus, effects of the non-parabolicity on subband electrons distribution and transition energy also needs to be included. The purpose of this chapter is to study the dependence of the intersubband gain spectrum on the electron-electron Coulomb interactions, non-parabolicity, resonant tunneling and laser device parameters. The device parameters include energy detuning, injection and extraction coupling strength and doping density.

In this section, we extend the simplified density-matrix model to include the many-body interactions derived from electron Hamiltonian in the second quantization. It not only takes into account of coherent effects in the electron transport through injector and extractor barriers by resonant tunneling, but also distributions of kinetic energy of electrons and many-body effects based on intersubband semiconductor-Bloch equations. The non-parabolicity effect is also approximately considered. The results show that the gain peak frequency calculated by the many-body model is more close to the experimentally measured lasing frequency compared with existing simplified density-matrix model. The proposed model provides a comprehensive picture of optical properties of QCLs, not only enhancing our in-depth understanding of optical gain, but also enabling an accurate prediction of the device performances *e.g.* linewidth enhancement factor. More importantly, this model has a low computational load which can greatly simplify the optimization process of active region designs of QCLs compared to other full quantum mechanical models.

3.2.2 Optical gain in THz QCLs

3.2.2.1 Intersubband semiconductor Bloch equations

Currently, the highest temperature operation of THz QCL is achieved by using resonant-tunneling injection design with three quantum wells in each period [29]. We consider the same design in this paper. Figure 3 - 1 shows the conduction band diagram and magnitude squared envelope wave functions of this design in “tight-binding” scheme, where the injector barrier and extractor barrier are made sufficiently thick to allow one period to be separated into two regions [36, 85, 90], *i.e.* the active region and the injector region (see Fig. 3 - 1). The energy states within either the active region or the injector region are coupled by scattering processes, but energy states from different regions are coupled by tunneling. This localized wave function analysis defined by each region allows us to investigate the effects of resonant tunneling on the electron transport and gain spectrum.

Fig. 3 - 1 Conduction band diagram and magnitude squared envelope wave functions of a four level resonant-phonon THz QCL with a diagonal design in the “tight-binding” scheme. Ω_{41} , Ω_{23} and Ω_{31} are the injection, extraction and parasitic coupling strength (unit: Hz/rad), respectively. The external electric field is 12.3 kV/cm. The radiative transition is from $4 \rightarrow 3$, and depopulation of the lower laser level is via $3 \rightarrow 2$ (RT) and $(2 \rightarrow 1)$ longitudinal optical phonon scattering. The thickness in angstrom of each layer is given as 49/88/27/82/42/160 starting from the injector barrier. The barriers are indicated in bold fonts. The widest well is doped at $3 \times 10^{10} \text{ cm}^{-2}$.

In order to conveniently treat the many-body problem, we derive the dynamical equations of motion in the second quantized representation. The Hamiltonian of the system can be divided into four parts

$$H = H_{el} + H_{rt} + H_0 + H_{Coul} \quad (3.1)$$

The first two terms

$$\begin{aligned} H_{el} + H_{rt} = & -\sum_{\mathbf{k}} (\mu E b_{4,\mathbf{k}}^\dagger b_{3,\mathbf{k}} + c.c) - \sum_{\mathbf{k}} [(\Delta_{41'}/2) b_{1',\mathbf{k}}^\dagger b_{4,\mathbf{k}} + c.c] \\ & - \sum_{\mathbf{k}} [(\Delta_{23}/2) b_{3,\mathbf{k}}^\dagger b_{2,\mathbf{k}} + c.c] - \sum_{\mathbf{k}} [(\Delta_{31'}/2) b_{1',\mathbf{k}}^\dagger b_{3,\mathbf{k}} + c.c] \end{aligned} \quad (3.2)$$

is the Hamiltonian for electron-light coupling and the tunneling effects, respectively. The resonant tunneling terms are written by a close analogy with the electron light term, which is similar to the density matrix model firstly proposed by Kazarinov et al [91]. $b_{j,\mathbf{k}}^\dagger$ ($b_{j,\mathbf{k}}$) is the creation (annihilation) operator of the electron state in subband j with wavevector $\mathbf{k}$. μ is the electron charge times the dipole matrix element of laser transition. $\Delta_{41'}$, Δ_{23} and $\Delta_{31'}$ are the injection, extraction and parasitic coupling strengths, which can be derived by a simple tight-binding approach (see Appendix A) [92].

The last two terms are [93]

$$H_0 + H_{Coul} = \sum_{j,\mathbf{k}} \varepsilon_{j,\mathbf{k}} b_{j,\mathbf{k}}^\dagger b_{j,\mathbf{k}} + \frac{1}{2} \sum_{uvv'u'}^{1,2,3,4} \sum_{\mathbf{k}\mathbf{k}'\mathbf{q}} V_{\mathbf{q}}^{uvv'u'} b_{u,\mathbf{k}+\mathbf{q}}^\dagger b_{v,\mathbf{k}'-\mathbf{q}}^\dagger b_{v',\mathbf{k}'} b_{u',\mathbf{k}} \quad (3.3)$$

which describe free electrons and electron-electron Coulomb interactions, respectively. $\varepsilon_{j,\mathbf{k}}$ is the j th subband energy, $\mathbf{k}$ is the in plane wave vector, the $V_{\mathbf{q}}^{uvv'u'}$ is the screening Coulomb matrix element which reads

$$V_{\mathbf{q}}^{uvv'u'} = \frac{e^2}{2A\varepsilon_0\varepsilon_r\varepsilon(q)q} \int dz \int \varphi_u(z) \varphi_{u'}(z) e^{-q|z-z'|} \varphi_v(z') \varphi_{v'}(z') dz' \quad (3.4)$$

where A is quantum well area, ε_r is background dielectric constant, and $q = |\mathbf{k} - \mathbf{k}'|$. The near-resonant screening is taken into account approximately via the dielectric function $\varepsilon(q)$ calculated by the single subband screening model [86, 94]. The Coulomb matrix elements are difficult to evaluate numerically, but they can be simplified according to the approaches proposed in Refs. [95, 96] without loss of high accuracy, which is presented in Appendix B.

The equations of motion for the polarizations $p_{ij,\mathbf{k}} = \langle b_{i,\mathbf{k}}^\dagger b_{j,\mathbf{k}} \rangle$ and electron occupation $n_{i,\mathbf{k}} = \langle b_{i,\mathbf{k}}^\dagger b_{i,\mathbf{k}} \rangle$, where the bracket $\langle \dots \rangle$ indicates an expectation value, can be derived by using the following Heisenberg equation and the anticommutations relations of fermionic operator [87]

$$\frac{dO}{dt} = \frac{i}{\hbar} [H, O] \quad (3.5)$$

$$[b_{i,\mathbf{k}}, b_{j,\mathbf{k}'}^\dagger]_+ = \delta_{ij,\mathbf{k}\mathbf{k}'}, \quad [b_{i,\mathbf{k}}, b_{j,\mathbf{k}'}]_+ = [b_{i,\mathbf{k}}^\dagger, b_{j,\mathbf{k}'}^\dagger]_+ = 0 \quad (3.6)$$

where O is operator, H is the Hamiltonian. However, due to the Coulomb interaction terms in Eq. (3.3), 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 bandstructure and Rabi frequency renormalizations. Scattering and dephasing contributions cause the second order correlation in the Coulomb potential, and so on [86, 88, 97]. In this chapter, we only include the Hartree-Fock contributions and dephasing and scattering contributions at the level of a relaxation-rate approximation (see appendix C). After a lengthy algebra, we obtain the following Bloch equations of density matrix elements for the polarizations $p_{ij,\mathbf{k}}$ and electron occupation $n_{i,\mathbf{k}}$ within the frame of rotating-wave approximation as

$$\frac{dp_{34,\mathbf{k}}}{dt} = -\gamma_{34p} p_{34,\mathbf{k}} - i \frac{\tilde{\mathcal{E}}_{43,\mathbf{k}}}{\hbar} p_{34,\mathbf{k}} - i \tilde{\Omega}_0 (n_{4,\mathbf{k}} - n_{3,\mathbf{k}}) + i \tilde{\Omega}_{41'}^\dagger p_{31',\mathbf{k}} - i \tilde{\Omega}_{23}^\dagger p_{24,\mathbf{k}} - i \tilde{\Omega}_{31'}^\dagger p_{41',\mathbf{k}} \quad (3.7)$$

$$\frac{dp_{41',\mathbf{k}}}{dt} = -\gamma_{41'p} p_{41',\mathbf{k}} - i \frac{\tilde{\mathcal{E}}_{1'4,\mathbf{k}}}{\hbar} p_{41',\mathbf{k}} - i \tilde{\Omega}_{41'} (n_{1,\mathbf{k}} - n_{4,\mathbf{k}}) - i \tilde{\Omega}_0^\dagger p_{31',\mathbf{k}} + i \tilde{\Omega}_{31'}^\dagger p_{34,\mathbf{k}} \quad (3.8)$$

$$\frac{dp_{23,\mathbf{k}}}{dt} = -\gamma_{23p} p_{23,\mathbf{k}} - i \frac{\tilde{\mathcal{E}}_{32,\mathbf{k}}}{\hbar} p_{23,\mathbf{k}} - i \tilde{\Omega}_{23} (n_{3,\mathbf{k}} - n_{2,\mathbf{k}}) + i \tilde{\Omega}_0^\dagger p_{24,\mathbf{k}} + i \tilde{\Omega}_{31'}^\dagger p_{21',\mathbf{k}} \quad (3.9)$$

$$\frac{dp_{31',\mathbf{k}}}{dt} = -\gamma_{31'p} p_{31',\mathbf{k}} - i \frac{\tilde{\mathcal{E}}_{1'3,\mathbf{k}}}{\hbar} p_{31',\mathbf{k}} - i \tilde{\Omega}_0^\dagger p_{41',\mathbf{k}} + i \tilde{\Omega}_{41'}^\dagger p_{34,\mathbf{k}} - i \tilde{\Omega}_{23}^\dagger p_{21',\mathbf{k}} - i \tilde{\Omega}_{31'} (n_{1',\mathbf{k}} - n_{3,\mathbf{k}}) \quad (3.10)$$

$$\frac{dp_{24,\mathbf{k}}}{dt} = -\gamma_{24p} p_{24,\mathbf{k}} - i \frac{\tilde{\mathcal{E}}_{42,\mathbf{k}}}{\hbar} p_{24,\mathbf{k}} + i \tilde{\Omega}_0^\dagger p_{23,\mathbf{k}} + i \tilde{\Omega}_{41'}^\dagger p_{21',\mathbf{k}} - i \tilde{\Omega}_{23}^\dagger p_{34,\mathbf{k}} \quad (3.11)$$

$$\frac{dp_{21',\mathbf{k}}}{dt} = -\gamma_{21'p} p_{21',\mathbf{k}} - i \frac{\tilde{\mathcal{E}}_{1'2,\mathbf{k}}}{\hbar} p_{21',\mathbf{k}} + i \tilde{\Omega}_{41'}^\dagger p_{24,\mathbf{k}} - i \tilde{\Omega}_{23}^\dagger p_{31',\mathbf{k}} + i \tilde{\Omega}_{31'}^\dagger p_{23,\mathbf{k}} \quad (3.12)$$

$$\begin{aligned} \frac{dn_{4,\mathbf{k}}}{dt} = & -i(\tilde{\Omega}_0^\dagger p_{34,\mathbf{k}} - \tilde{\Omega}_0 p_{34,\mathbf{k}}^\dagger) - i(\tilde{\Omega}_{41'} p_{41',\mathbf{k}}^\dagger - \tilde{\Omega}_{41'}^\dagger p_{41',\mathbf{k}}) - \gamma_4 [n_{4,\mathbf{k}} - f_{4,\mathbf{k}}(\mu_{4,e}, T_{4,e})] \\ & - \gamma_{43} [n_{4,\mathbf{k}} - f_{4,\mathbf{k}}(\mu_{43}, T_l)] - \gamma_{sp} n_{4,\mathbf{k}} \end{aligned} \quad (3.13)$$

$$\begin{aligned} \frac{dn_{3,\mathbf{k}}}{dt} = & -i(\tilde{\Omega}_0 p_{34,\mathbf{k}}^\dagger - \tilde{\Omega}_0^\dagger p_{34,\mathbf{k}}) - i(\tilde{\Omega}_{23}^\dagger p_{23,\mathbf{k}} - \tilde{\Omega}_{23} p_{23,\mathbf{k}}^\dagger) - \gamma_3 [n_{3,\mathbf{k}} - f_{3,\mathbf{k}}(\mu_{3,e}, T_{3,e})] \\ & - \gamma_{43} [n_{3,\mathbf{k}} - f_{3,\mathbf{k}}(\mu_{43}, T_l)] + \gamma_{sp} n_{4,\mathbf{k}} - i(\tilde{\Omega}_{31'} p_{31',\mathbf{k}}^\dagger - \tilde{\Omega}_{31'}^\dagger p_{31',\mathbf{k}}) \end{aligned} \quad (3.14)$$

$$\frac{dn_{2,\mathbf{k}}}{dt} = -i(\tilde{\Omega}_{23}^\dagger p_{23,\mathbf{k}}^\dagger - \tilde{\Omega}_{23} p_{23,\mathbf{k}}) - \gamma_2 [n_{2,\mathbf{k}} - f_{2,\mathbf{k}}(\mu_{2,e}, T_{2,e})] - \gamma_{21'} [n_{2,\mathbf{k}} - f_{2,\mathbf{k}}(\mu_{21'}, T_l)] \quad (3.15)$$

$$\begin{aligned} \frac{dn_{1',\mathbf{k}}}{dt} = & -i(\tilde{\Omega}_{41'}^\dagger p_{41',\mathbf{k}}^\dagger - \tilde{\Omega}_{41'} p_{41',\mathbf{k}}^\dagger) - \gamma_{1'} [n_{1',\mathbf{k}} - f_{1',\mathbf{k}}(\mu_{1',e}, T_{1,e})] - \gamma_{21'} [n_{1',\mathbf{k}} - f_{1',\mathbf{k}}(\mu_{21'}, T_l)] \\ & -i(\tilde{\Omega}_{31'}^\dagger p_{31',\mathbf{k}}^\dagger - \tilde{\Omega}_{31'} p_{31',\mathbf{k}}^\dagger) \end{aligned} \quad (3.16)$$

with

$$\tilde{\mathcal{E}}_{uv,\mathbf{k}} = \mathcal{E}_{u,\mathbf{k}} - \mathcal{E}_{v,\mathbf{k}} - \sum_{\mathbf{k}' \neq \mathbf{k}} (V_{\mathbf{k}-\mathbf{k}'}^{uuuu} n_{u,\mathbf{k}'} - V_{\mathbf{k}-\mathbf{k}'}^{vvvv} n_{v,\mathbf{k}'}) + \sum_{\mathbf{k}' \neq \mathbf{k}} (n_{u,\mathbf{k}'} - n_{v,\mathbf{k}'}) V_{\mathbf{k}-\mathbf{k}'}^{uvuv} \quad (3.17)$$

$$\tilde{\Omega}_0 = \frac{\mu_{\tilde{c}}}{2\hbar} + \frac{1}{\hbar} \sum_{\mathbf{k}' \neq \mathbf{k}} V_{\mathbf{k}-\mathbf{k}'}^{4334} p_{43,\mathbf{k}'} - \frac{2}{\hbar} V_0^{4343} \sum_{\mathbf{k}'} p_{43,\mathbf{k}'} \quad (3.18)$$

$$\tilde{\Omega}_{uv} = \frac{\Delta_{uv}}{2\hbar} + \frac{1}{\hbar} \sum_{\mathbf{k}' \neq \mathbf{k}} V_{\mathbf{k}-\mathbf{k}'}^{uvvu} p_{uv,\mathbf{k}'} - \frac{2}{\hbar} V_0^{uvuv} \sum_{\mathbf{k}'} p_{uv,\mathbf{k}'} \quad (3.19)$$

where γ_{ijp} is the dephasing rate associated with energy levels i and j . γ_j is the intrasubband electron-electron scattering rate at level j , γ_{ij} is the combined electron-electron and electron-phonon scattering rate between levels i and j . The lifetime of the upper laser level as a function of temperature is approximated in the form of $\gamma_{43} = \gamma_{430} \exp[\hbar(\omega_{LO} - \omega_\lambda)/k_b T_{4e}]$ where γ_{430} is the raw LO phonon scattering rate when the upper-state electrons can sufficiently emit LO phonons and ω_{LO} is the energy of LO phonon. γ_{sp} is the spontaneous emission rate. $T_{j,e}$ is the electron temperature at level j , T_l is the lattice temperature. $f_{j,\mathbf{k}}$ is the Fermi-Dirac distribution with chemical potential $\mu_{j,e}$ at level j . The chemical potentials and temperatures are determined by electron number conservation and energy conservation [87]. The terms with coefficients $V_{\mathbf{k}-\mathbf{k}'}^{uuuu}$ refer to the exchange self-energy, $V_{\mathbf{k}-\mathbf{k}'}^{uvvu}$ to the excitonic enhancement, and V_0^{uvuv} to the depolarization. Processes corresponding to these contributions are shown in Fig. 3 - 2. The influence of the subband dispersion, namely the nonparabolicity, is represented by using effective mass of electrons m^* . For subband j , we have $\varepsilon_{j,\mathbf{k}} = \varepsilon_j + \hbar^2 \mathbf{k}^2 / 2m_j^*$. For our structure, the calculation shows that $m_1^* \approx 0.0670m_0$, $m_2^* \approx m_3^* \approx 0.0723m_0$ and $m_4^* \approx 0.0743m_0$ (m_0 is the free electron mass) according to Ekenberg's model [98].

Fig. 3 - 2 Graphical representations of the Coulomb interactions between the subbands u and v (from the left to the right): exchange self-energy, excitonic enhancement and depolarization. The exchange shift induces a renormalization of the free-carrier contributions; the excitonic contribution causes a renormalization of the electron-light interaction; the depolarization contribution is another renormalization of the electron-light interaction with a very different origin.

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

$$\begin{aligned} p_{41',\mathbf{k}} &= p_{41',\mathbf{k}}^{(0)}, \quad p_{34,\mathbf{k}} = p_{34,\mathbf{k}}^{(0)} + \tilde{p}_{34,\mathbf{k}} e^{-i\omega_2 t}, \quad p_{31',\mathbf{k}} = p_{31',\mathbf{k}}^{(0)} + \tilde{p}_{31',\mathbf{k}} e^{-i\omega_2 t}, \\ p_{23,\mathbf{k}} &= p_{23,\mathbf{k}}^{(0)}, \quad p_{24,\mathbf{k}} = p_{24,\mathbf{k}}^{(0)} + \tilde{p}_{24,\mathbf{k}} e^{-i\omega_2 t}, \quad p_{21',\mathbf{k}} = p_{21',\mathbf{k}}^{(0)} + \tilde{p}_{21',\mathbf{k}} e^{-i\omega_2 t}. \end{aligned} \quad (3.20)$$

where $p_{ij,\mathbf{k}}^{(0)}$ is the static tunneling induced coherence, and $\tilde{p}_{ij,\mathbf{k}}$ is the laser-induced coherence [85].

In the above semiconductor Bloch equations, the relaxation-rate approximation is employed to calculate dephasing and scattering contributions [99], because a full kinetic treatment with dephasing and scattering terms based on the Boltzmann equation requires length computational time. In this relaxation-rate approximation, the dephasing contributions on the polarizations is treated as

$$\frac{dp_{ij,\mathbf{k}}}{dt} = -\gamma_{ijp} p_{ij,\mathbf{k}} \quad (3.21)$$

where γ_{ijp} denotes an effective dephasing rate.

As for scattering contributions, the influence of scattering on the electron distributions is treated by the relaxation of a given population distribution $n_{u,\mathbf{k}}$ to a quasi-equilibrium Fermi-Dirac distribution $f_{u,\mathbf{k}}(T_i, \mu_i)$ with temperature T_i and chemical potential μ_i as $\gamma_u (n_{u,\mathbf{k}} - f_{u,\mathbf{k}})$ at

energy level i . The two main scattering contributions are considered in THz QCLs i.e. electron-electron scattering and electron-phonon scattering.

For intrasubband scattering, we only consider the electron-electron scattering due to the much smaller intrasubband electron-phonon scattering rate. The electron distribution of each subband (e.g. subband u) is driven towards Fermi-Dirac distribution by this intrasubband scattering at the corresponding electron temperature $T_{u,e}$ and chemical potential $\mu_{u,e}$. The actual value of electron temperature and chemical potential can be determined by the conditions of particle and energy conservation

$$\sum_{\mathbf{k}} n_{u,\mathbf{k}} = \sum_{\mathbf{k}} f_{u,\mathbf{k}}(\mu_{u,e}, T_{u,e}) \quad (3.22)$$

$$\sum_{\mathbf{k}} \varepsilon_{u,\mathbf{k}} n_{u,\mathbf{k}} = \sum_{\mathbf{k}} \varepsilon_{u,\mathbf{k}} f_{u,\mathbf{k}}(\mu_{u,e}, T_{u,e}) \quad (3.23)$$

As to the intersubband scattering, two contributions are considered. Firstly, for the intersubband electron-phonon scattering between levels u and v , energy is dissipated from the electrons to the lattice. In this case, electrons of these two energy levels relax to share a Fermi-Dirac distribution with lattice temperature T_l and chemical potential $\mu_{uv,l}$. The chemical potential can be determined by the particle conservation

$$\sum_{\mathbf{k}} \sum_{i=u,v} n_{i,\mathbf{k}} = \sum_{\mathbf{k}} \sum_{i=u,v} f_{i,\mathbf{k}}(\mu_{uv,l}, T_l) \quad (3.24)$$

In case of the intersubband electron-electron scattering between levels u and v , electrons of these two energy levels are driven to share a Fermi-Dirac distribution with electron temperature $T_{uv,e}$ and chemical potential $\mu_{uv,e}$. According to the particle and energy conservation, it has

$$\sum_{\mathbf{k}} \sum_{i=u,v} n_{i,\mathbf{k}} = \sum_{\mathbf{k}} \sum_{i=u,v} f_{i,\mathbf{k}}(\mu_{uv,e}, T_{uv,e}) \quad (3.25)$$

$$\sum_{\mathbf{k}} \sum_{i=u,v} \varepsilon_{i,\mathbf{k}} n_{i,\mathbf{k}} = \sum_{\mathbf{k}} \sum_{i=u,v} \varepsilon_{i,\mathbf{k}} f_{i,\mathbf{k}}(\mu_{uv,e}, T_{uv,e}) \quad (3.26)$$

If the electron-electron and electron-phonon scattering between levels u and v occur on similar time-scales, electrons of these two energy levels driven by electron-electron scattering still relax to share a Fermi-Dirac distribution with lattice temperature T_l and chemical potential $\mu_{uv,l}$ [88]. Because the electron-electron and electron-phonon scattering between the upper laser level and lower laser level occur on similar time-scales for THz QCLs due to thermally activated phonon scattering in our chosen temperatures, electrons of these two energy levels are assumed

to share a Fermi-Dirac distribution at lattice temperature, as shown in Eqs. (3.13) and (3.14). In addition, the electron-phonon interaction dominates the scattering processes between level 2 and level 1 in THz QCLs, we neglect the electron-electron scattering between level 2 and 1, as shown in Eqs. (3.15) and (3.16).

3.2.2.2 Coupling of Bloch equation and Maxwell's equations and optical gain

A laser field $E(z, t)$ will induce a microscopic electric-dipole moment. Summation of these dipoles yields a macroscopic polarization P_{ij} . This polarization then drives the laser field $E'(z, t)$ following the Maxwell's equations. Self-consistency requires $E'(z, t) = E(z, t)$. Therefore, we should know how the polarization affects the slowly varying electric field amplitude in order to obtain the optical gain.

According to the Maxwell's equations, after some algebra, we have the following wave equation [87]

$$-\nabla^2 \mathbf{E} + \left(\frac{n}{c}\right)^2 \frac{\partial^2 \mathbf{E}}{\partial t^2} = -\mu_0 \frac{\partial^2 \mathbf{P}}{\partial t^2} \quad (3.27)$$

where n is the refractive index, c is the light speed in vacuum and μ_0 is the permeability of vacuum.

Assuming single mode operation in THz QCL, the laser field can be written as

$$\mathbf{E}(z, t) = \frac{1}{2} \hat{i} \xi(z, t) e^{i[kz - \omega_\lambda t - \phi(z)]} + c.c. \quad (3.28)$$

where $\hat{i}$ is the unit vector. ξ is the slowly varying complex electric field amplitude, ω_λ is the laser frequency, k is wave vector and ϕ is the phase. The induced polarization is

$$\mathbf{P}(z, t) = \frac{1}{2} \hat{i} P(z, t) e^{i[kz - \omega_\lambda t - \phi(z)]} + c.c. \quad (3.29)$$

The slowly varying polarization amplitude is coupled with complex electric field amplitude as

$$P(z, t) = \varepsilon_0 n^2 \chi(z) \xi(z, t) \quad (3.30)$$

where ε_0 is the vacuum permittivity, and $\chi(z)$ is the complex susceptibility of the gain medium.

By inserting Eq. (3.28), Eq. (3.29) and Eq. (3.30) into Eq. (3.27) and neglecting the terms with double derivatives and multiplication of two derivatives (slowly varying envelope approximation), we can get the following equations

$$\frac{d\xi(z,t)}{dz} = -\frac{\omega_\lambda n}{2c} \chi''(z) \xi(z,t) \quad (3.31)$$

$$\frac{d\phi(z,t)}{dz} = -\frac{\omega_\lambda n}{2c} \chi'(z) \quad (3.32)$$

where $\chi(z) = \chi'(z) + i\chi''(z)$. With the definition of the amplitude gain g

$$\frac{d\xi(z,t)}{dz} = g\xi(z,t) \quad (3.33)$$

we can find the intensity gain $G=2g$ as

$$G = \frac{\omega_\lambda n}{c} \chi''(z) \quad (3.34)$$

Since the macroscopic polarization can be linked with microscopic through

$$P(z,t) = \frac{1}{V_m} \sum_{\mathbf{k}} \mu_{34} p_{34,\mathbf{k}} \quad (3.35)$$

where V_m is the volume of one period of active region, we can derive the intensity gain as

$$G = -\frac{2\omega_\lambda}{\varepsilon_0 n c V_m \xi} \text{Im} \left(\sum_{\mathbf{k}} \mu_{34} \tilde{p}_{34,\mathbf{k}} \right) \quad (3.36)$$

3.2.2.3 Results and discussions

3.2.2.2.1 Effects of many-body interaction and non-parabolicity on optical gain

Fig. 3 - 3 Simulation results for gain spectra at resonance and 100 K. (a) The gain spectra calculated from microscopic model “many-body + non-parabolicity” (solid line), microscopic model “many-body + parabolicity” (dotted lines), microscopic model “free carriers” (dot-dashed lines) and simplified matrix density (SDM) model (dashed lines). The following default parameters are used: $z_{43}=40.7 \text{ \AA}$, $2\hbar\Omega_{41'} = 2.16 \text{ meV}$, $2\hbar\Omega_{23} = 4.94 \text{ meV}$, γ_{34p}

$= 2.4 \times 10^{12} \text{ s}^{-1}$, $\gamma_{41p} = 4.0 \times 10^{12} \text{ s}^{-1}$, $\gamma_{23p} = 4.5 \times 10^{12} \text{ s}^{-1}$, $\gamma_{31p} = 3.5 \times 10^{12} \text{ s}^{-1}$, $\gamma_{24p} = 3.5 \times 10^{12} \text{ s}^{-1}$, $\gamma_{21p} = 3.5 \text{ s}^{-1}$, $\gamma_{43} = 1.3 \times 10^{11} \text{ s}^{-1}$, $\gamma_{21} = 3 \times 10^{12} \text{ s}^{-1}$, $\gamma_1 = 1.3 \times 10^{12} \text{ s}^{-1}$, $\gamma_4 = 1.3 \times 10^{12} \text{ s}^{-1}$, $\gamma_3 = 1.3 \times 10^{12} \text{ s}^{-1}$, $\gamma_2 = 1.3 \times 10^{12} \text{ s}^{-1}$. The typical values of scattering and dephasing rate are used [36, 89]. (b) The interplay of different Coulomb interactions. Solid lines: full many-body effects with non-parabolicity; dashed lines: free carriers; dotted lines: depolarization; dot-dashed lines: excitonic enhancement; dot-dot-dashed lines: exchange self-energy.

The results presented here are obtained by numerically solving the equations of motion (Eqns.(3.7)-(3.19)) for a small laser field (linear absorption). In our calculation, 250 k -points within the each subband are taken. Figure 3 - 3 shows the computed gain spectra at resonance based on the design shown in Fig. 3 - 1. In order to evaluate the importance of Coulomb interaction and non-parabolicity on optical properties for THz QCLs, we firstly compare the gain spectra calculated from the microscopic model with “many-body + non-parabolicity” (considering both many-body and non-parabolicity effects), microscopic model with “many-body + parabolicity” (considering both many-body and parabolicity effects), microscopic model with “free-carrier” (considering free carriers and non-parabolicity but neglecting the bandstructure and Rabi frequency renormalization) and the simplified density-matrix (SDM) model (Ref. [68]), as shown in Fig. 3 - 3 (a). Comparing the two gain spectra calculated by the microscopic models with “many-body + non-parabolicity” and “many-body + parabolicity”, we found that subband dispersion with different subband effective masses (non-parabolicity) causes a slight shift of peak position of gain spectrum to the lower frequency side and reduces the peak gain due to the redistribution of subband electrons in k space caused by the non-parabolicity effect. Not only the non-parabolicity, but also the many-body Coulomb interactions make the red-shift of gain spectrum and cause the decrease of peak value of optical gain of THz QCLs by comparing microscopic models with “many-body + nonparabolicity” and “free-carrier”. The red-shift of the gain spectrum is mainly caused by the depolarization terms with the consideration of the interplay of various many-body interactions, as shown in Fig. 3 - 3(b). It is shown that, in our population inverted laser system, the depolarization terms cause a red-shift of gain spectrum relative to the “free-carrier” model [100] but it induces the blue-shift in the usual non-inverted absorption system. In addition, exchange self-energy terms renormalize the subband energy level and induce non-parabolicity to slightly red-shift the gain spectrum, and excitonic enhancement terms give a peak near the higher frequency edge of the spectrum and cause the slight blue-shift of the gain spectrum in THz QCLs. Therefore, the interplay of various many-body effects leads to the red-shift of gain spectrum.

In addition, since the SDM model does not consider the many-body interactions and non-parabolicity, the obtained gain peak are overestimated when comparing the results obtained from the microscopic “many-body + non-parabolicity” model with the SDM model. The gain peak frequency calculated by the microscopic model is lower (~ 0.3 THz) than that by the SDM model. Because, as demonstrated in Ref. [29], the macroscopic density matrix model overestimates (~ 0.6 THz) the gain peak frequency compared to the experimentally measured lasing frequency, therefore the microscopic many-body model discussed in this paper predicts a better result closer to the experimental lasing frequency. However, the gain peak frequency calculated by the microscopic model is still ~ 0.3 THz higher than the experimental value. This discrepancy is probably caused by the neglected intermodule electron-light scattering in this particular density matrix model [29]. Due to the choice of basis states from the two isolated modules (the active and the injector modules) in the “tight-binding” scheme (see Fig. 3 - 1), this model considers only intramodule scatterings and hence only one intramodule dipole moment z_{43} but neglects other direct dipole moment contributions e.g. z_{42} . This limit is not inherent to the density matrix model, but is due to the choice of the basis states. Further work could be carried out to include this intermodule electron-light scattering.

3.2.2.2.2 Optical gain spectrum as a function of energy detuning

When energy levels $1 \leftrightarrow 4$ and $3 \leftrightarrow 2$ are strongly coupled, energy splitting occurs due to their anticrossing. In this case, levels $1 \leftrightarrow 4$ and $3 \leftrightarrow 2$ form the dressed states which will induce the broadening of gain spectrum in QCLs. Figure 3 - 4 shows the gain spectra at different injection and extraction detunings calculated from the microscopic model (in below discussions, unless otherwise specified, “microscopic model” refers to the microscopic model “many-body + non-parabolicity”) and free-carrier model, respectively. The gain peak is greatly shifted as the injection and extraction detunings become positive value from negative one. This change results from the contributions of the polarization $p_{31,k}$ and $p_{24,k}$ to polarization $p_{34,k}$ due to the indirect $1 \rightarrow 3$ and $4 \rightarrow 2$ radiative transition formed by the coherent coupling $1 \leftrightarrow 4$, $3 \leftrightarrow 4$ and $3 \leftrightarrow 2$. The optical gain at zero injection and extraction detunings is most boarded due to the comparable radiative transitions strengths between anticrossing energy levels at resonance. In addition, by comparing Fig. 3-4 (a) with (b), we found that the peak position and lineshape of gain spectrum more strongly follows the variations of extraction detuning due to stronger coupling. Moreover,

for each injection and extraction detunings, the many-body interactions suppress the high frequency side of gain spectrum and enhance low the frequency side by the comparison of microscopic model and free-carrier one.

Fig. 3 - 4 The effects of detunings with and without many-body interactions on the optical gain at 100 K. (a) The gain spectra at different injection detunings ε_{14} , the extractor level is kept to be in resonance with the low laser level. (b) The gain spectra at different extraction detunings ε_{32} . The injection level is kept to be in resonance with the upper laser level.

3.2.2.2.3 Optical gain spectrum as a function of coupling strength

Fig. 3 - 5 Effects of injection coupling strength with and without many-body interactions on gain spectra at resonance and 100 K. (a) Gain spectra at different injection coupling strength with the extraction coupling strength of 4.94 meV. (b) Gain spectra at different injection coupling strength with the extraction coupling strength of 2 meV.

In order to further illustrate the effects of coherence of resonant tunneling, we simulate the gain spectra at different injection, extraction and parasitic coupling strengths. Figure 3-5 shows the effects of injection coupling strength on optical gain when the extraction coupling strength is set as 4.94 meV and 2 meV, respectively. Both Fig. 3 - 5(a) and (b) show that, as the injection

coupling strength gradually increases towards the value of extraction coupling strength, the gain spectrum tends to be enhanced, peak frequency follows the variation of coupling strength, and spectrum width is slightly broadened. When the injection coupling strength is small enough (smaller than extraction coupling strength), double-peaked gain spectrum is generated and obviously shown. Moreover, by the comparison of microscopic model and free-carrier one, the many-body interaction tends to suppress the high frequency side of gain spectrum and enhance the low frequency side, as shown in Fig. 3 - 5(a).

Fig. 3 - 6 Gain spectra with and without many-body interactions at different extraction coupling strength at resonance and 100 K.

In contrast, since the extraction coupling strength is larger than the injection coupling strength, as the extraction coupling strength increases, as shown in Fig. 3 - 6, the extraction electron transport tends to be more coherent and the gain spectrum is additionally broadened, and the peak value is reduced owing to the interplay of Coulomb interaction and indirectly coherent interaction p_{24} . When the extraction coupling strength is large enough (larger than injection coupling strength), double-peaked gain spectrum is generated. Similarly, owing to modifications to gain spectrum by many-body interactions, the peak at high frequency side is suppressed and low frequency side is enhanced according to the results of microscopic model, as compared with free-carrier one.

One of the limitations to the high performance of THz QCLs comes from the difficulty in selecting injection of electrons into the upper laser level. This is due to the small energy separation between transition energy levels. The current leakage is determined by the parasitic

coupling between the injection level and lower laser level. Therefore, this parasitic coupling can greatly influence the optical gain of THz QCLs. Figure 3 - 7 shows the effects of the parasitic coupling strengths on the optical gain spectra when the injection and extraction detunings are set as $\varepsilon_{14} = -6$ meV and $\varepsilon_{32} = -6$ meV to limit additional broadening due to the coherent RT of injection and extraction processes. Due to the current leakage from the injection level to lower laser level, the peak value of gain spectrum is fast decreased as the parasitic coupling strength increases, but the gain peak is just slightly red-shifted due to the large detuning between the injection level and lower laser level. It is noted that when the parasitic coupling strength is much smaller (e.g. $2\hbar\Omega_{31'}=0.7$ meV) than that of the injection coupling strength, the gain spectrum is just slightly influenced, while it is equivalent (e.g. $2\hbar\Omega_{31'}=2.1$ meV) to the injection coupling strength, the optical gain can be strongly reduced. Therefore, in order to improve the performance of THz QCLs, we should design the eigenstates of optical transition to be diagonally coupled.

Fig. 3 - 7 Gain spectra at different parasitic coupling strength with and without many-body interactions on gain spectra at 100 K with injection and extraction detunings $\varepsilon_{14} = -6$ meV and $\varepsilon_{32} = -6$ meV.

3.2.2.2.4 Optical gain spectrum as a function of doping density

Since many-body Coulomb interaction strongly depends on the doping density, we anticipate that doping will strongly affect the gain spectra, as shown in Fig. 3-8. As the doping density increases, the many-body interactions become stronger, and hence the spectra are further red-shifted. Moreover, not only the peak value with doping density is enhanced by many-body

interactions, the lineshape of spectrum is modified, as shown in Fig. 3 - 8(b). Figure 3 - 8(c) shows the interplay of various Coulomb interactions at doping density $3 \times 10^{10} \text{ cm}^{-2}$ and $5.4 \times 10^{10} \text{ cm}^{-2}$, respectively. As shown by this figure, the red-shift and spectrum lineshape modification are mainly attributed to depolarization, as demonstrated in Ref. [101]. In contrast, the spectrum lineshape calculated from free-carrier model is not changed with the rise in doping density.

Fig. 3 - 8 (a) Effects of doping density on gain spectra at resonance and 100 K. The solid lines with right triangle and dashed lines with open circles show the results of many-body and free-carrier models, respectively. From the bottom to the top for each kind of color line, the doping density is $1.8 \times 10^{10} \text{ cm}^{-2}$, $3 \times 10^{10} \text{ cm}^{-2}$, $4.2 \times 10^{10} \text{ cm}^{-2}$, and $5.4 \times 10^{10} \text{ cm}^{-2}$. The effects of doping density on lifetimes of energy levels are neglected. (b) Normalized gain spectra from microscopic model at different doping density. (c) The interplay of various Coulomb interactions at doping density $3 \times 10^{10} \text{ cm}^{-2}$ and $5.4 \times 10^{10} \text{ cm}^{-2}$, respectively. Solid lines: full many-body effects with non-parabolicity; dashed lines: depolarization; dot-dashed lines: excitonic enhancement; dot-dot-dashed lines: exchange self-energy.

3.2.3 Optical gain in mid-IR QCLs

3.2.3.1 Equations of motion

Fig. 3 - 9 Schematic conduction band diagram of the active region with three-well vertical design plus injector and the moduli squared of the wave functions involved in the laser transition designed at 60 kV/cm in the “tight-binding” scheme [102]. The coupling between the periods, achieved by resonant tunneling (Ω_{51} 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**. $\text{In}_{0.52}\text{Al}_{0.48}\text{As}$ barrier layers are in bold, $\text{In}_{0.53}\text{Ga}_{0.47}\text{As}$ well layers are in roman, and n -doped layers ($2.5 \times 10^{17} \text{ cm}^{-3}$) are underlined.

For mid-IR QCLs, we use the active region with three-well vertical design active regions which has been used in the measurements [102, 103] of linewidth enhancement factor that we will discuss in chapter 6. Figure 3 - 9 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 (Ω_{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. 3 - 9, which characterizes the electron-light coupling, the tunneling effects, free electrons and electron-electron Coulomb interactions, can be written as

$$\begin{aligned}
 H = & - \sum_{j=3,4} \sum_{\mathbf{k}} (\mu_{j5} E b_{5,\mathbf{k}}^\dagger b_{j,\mathbf{k}} + c.c) - \sum_{\mathbf{k}} [(\Delta_{51}/2) b_{1,\mathbf{k}}^\dagger b_{5,\mathbf{k}} + c.c] \\
 & + \sum_{j=1,3,4,5} \sum_{\mathbf{k}} \varepsilon_{j,\mathbf{k}} b_{j,\mathbf{k}}^\dagger b_{j,\mathbf{k}} + \frac{1}{2} \sum_{uvv'u'}^{1,3,4,5} \sum_{\mathbf{k}\mathbf{k}'\mathbf{q}} V_{\mathbf{q}}^{uvv'u'} b_{u,\mathbf{k}+\mathbf{q}}^\dagger b_{v,\mathbf{k}'-\mathbf{q}}^\dagger b_{v',\mathbf{k}} b_{u',\mathbf{k}}
 \end{aligned}$$

(3.37)

where $E(t) = (\xi(t)/2)e^{-i\omega_\lambda t} + c.c.$ is the laser field (ξ is the slowly varying complex electric field amplitude, ω_λ is the laser frequency). μ_{j5} is the electron charge times the dipole matrix element of laser transition between energy level j and 5, $\Delta_{51'}$ is the injection coupling strengths. $\varepsilon_{j,\mathbf{k}}$ is the j th subband energy, $\mathbf{k}$ is the in-plane wave vector. $V_q^{uvv'u'}$ is the two dimensional screening Coulomb matrix element [73, 87]. The parasitic coupling between levels 1 and 4 is neglected, which is a reasonably good approximation for mid-IR QCLs [36].

According to the Heisenberg equation and many-body theory, one can then get the following equations of motion for the slowly varying polarization $p_{ij,\mathbf{k}} = \langle b_{i,\mathbf{k}}^\dagger b_{j,\mathbf{k}} \rangle$ and electron occupation $n_{i,\mathbf{k}} = \langle b_{i,\mathbf{k}}^\dagger b_{i,\mathbf{k}} \rangle$ in the rotating-wave approximation

$$\frac{dp_{51',\mathbf{k}}}{dt} = -\gamma_{51'p} p_{51',\mathbf{k}} - i \frac{\tilde{\varepsilon}_{1'5,\mathbf{k}}}{\hbar} p_{51',\mathbf{k}} + i\tilde{\Omega}_{51'}(n_{5,\mathbf{k}} - n_{1',\mathbf{k}}) - i\tilde{\Omega}_{45}^\dagger p_{41',\mathbf{k}} - i\tilde{\Omega}_{35}^\dagger p_{31',\mathbf{k}} \quad (3.38)$$

$$\frac{dp_{45,\mathbf{k}}}{dt} = -\gamma_{45p} p_{45,\mathbf{k}} - i \left(\frac{\tilde{\varepsilon}_{54,\mathbf{k}}}{\hbar} - \omega_\lambda \right) p_{45,\mathbf{k}} - i\tilde{\Omega}_{45}(n_{5,\mathbf{k}} - n_{4,\mathbf{k}}) + i\tilde{\Omega}_{35} p_{34,\mathbf{k}}^\dagger + i\tilde{\Omega}_{51'}^\dagger p_{41',\mathbf{k}} \quad (3.39)$$

$$\frac{dp_{35,\mathbf{k}}}{dt} = -\gamma_{35p} p_{35,\mathbf{k}} - i \left(\frac{\tilde{\varepsilon}_{53,\mathbf{k}}}{\hbar} - \omega_\lambda \right) p_{35,\mathbf{k}} - i\tilde{\Omega}_{35}(n_{5,\mathbf{k}} - n_{3,\mathbf{k}}) + i\tilde{\Omega}_{45}^\dagger p_{34,\mathbf{k}} + i\tilde{\Omega}_{51'}^\dagger p_{31',\mathbf{k}} \quad (3.40)$$

$$\frac{dp_{41',\mathbf{k}}}{dt} = -\gamma_{41'p} p_{41',\mathbf{k}} - i \left(\frac{\tilde{\varepsilon}_{1'4,\mathbf{k}}}{\hbar} - \omega_\lambda \right) p_{41',\mathbf{k}} - i\tilde{\Omega}_{45}^\dagger p_{51',\mathbf{k}} + i\tilde{\Omega}_{51'}^\dagger p_{45,\mathbf{k}} \quad (3.41)$$

$$\frac{dp_{34,\mathbf{k}}}{dt} = -\gamma_{34p} p_{34,\mathbf{k}} - i \frac{\tilde{\varepsilon}_{43,\mathbf{k}}}{\hbar} p_{34,\mathbf{k}} + i\tilde{\Omega}_{45}^\dagger p_{35,\mathbf{k}} - i\tilde{\Omega}_{35} p_{45,\mathbf{k}}^\dagger \quad (3.42)$$

$$\frac{dp_{31',\mathbf{k}}}{dt} = -\gamma_{31'p} p_{31',\mathbf{k}} - i \left(\frac{\tilde{\varepsilon}_{1'3,\mathbf{k}}}{\hbar} - \omega_\lambda \right) p_{31',\mathbf{k}} - i\tilde{\Omega}_{35}^\dagger p_{51',\mathbf{k}} + i\tilde{\Omega}_{51'}^\dagger p_{35,\mathbf{k}} \quad (3.43)$$

$$\frac{dn_{1',\mathbf{k}}}{dt} = i(\tilde{\Omega}_{51'} p_{51',\mathbf{k}}^\dagger - \tilde{\Omega}_{51'}^\dagger p_{51',\mathbf{k}}) - \gamma_{1'} [n_{1',\mathbf{k}} - f_{1',\mathbf{k}}(\mu_{1',e}, T_{1,e})] - \gamma_{21'} [n_{1',\mathbf{k}} - f_{1',\mathbf{k}}(\mu_{21'}, T_l)] \quad (3.44)$$

$$\begin{aligned} \frac{dn_{5,\mathbf{k}}}{dt} = & i(\tilde{\Omega}_{45} p_{45,\mathbf{k}}^\dagger - \tilde{\Omega}_{45}^\dagger p_{45,\mathbf{k}}) + i(\tilde{\Omega}_{35} p_{35,\mathbf{k}}^\dagger - \tilde{\Omega}_{35}^\dagger p_{35,\mathbf{k}}) \\ & + i(\tilde{\Omega}_{51'}^\dagger p_{51',\mathbf{k}} - \tilde{\Omega}_{51'} p_{51',\mathbf{k}}^\dagger) - \gamma_5 [n_{5,\mathbf{k}} - f_{5,\mathbf{k}}(\mu_{4,e}, T_{4,e})] - \sum_{j=4,3} \gamma_{5j} [n_{5,\mathbf{k}} - f_{5j,\mathbf{k}}(\mu_{5j}, T_l)] \end{aligned} \quad (3.45)$$

$$\frac{dn_{4,\mathbf{k}}}{dt} = i(\tilde{\Omega}_{45}^\dagger p_{45,\mathbf{k}} - \tilde{\Omega}_{45} p_{45,\mathbf{k}}^\dagger) - \gamma_4 [n_{4,\mathbf{k}} - f_{4,\mathbf{k}}(\mu_{4,e}, T_{4,e})] - \sum_{j=5,3} \gamma_{4j} [n_{4,\mathbf{k}} - f_{4j,\mathbf{k}}(\mu_{4j}, T_l)] \quad (3.46)$$

$$\begin{aligned} \frac{dn_{3,\mathbf{k}}}{dt} = & i(\tilde{\Omega}_{35}^\dagger p_{35,\mathbf{k}} - \tilde{\Omega}_{35} p_{35,\mathbf{k}}^\dagger) - \gamma_3 [n_{3,\mathbf{k}} - f_{3,\mathbf{k}}(\mu_{3,e}, T_{3,e})] - \sum_{j=5,4} \gamma_{3j} [n_{3,\mathbf{k}} - f_{3,\mathbf{k}}(\mu_{3j}, T_l)] \\ & - \gamma_{31'} [n_{3,\mathbf{k}} - f_{3,\mathbf{k}}(\mu_{31'}, T_l)] \end{aligned} \quad (3.47)$$

where

$$\tilde{\varepsilon}_{uv,\mathbf{k}} = \varepsilon_{u,\mathbf{k}} - \varepsilon_{v,\mathbf{k}} - \sum_{\mathbf{k}' \neq \mathbf{k}} (V_{\mathbf{k}-\mathbf{k}'}^{uuuu} n_{u,\mathbf{k}'} - V_{\mathbf{k}-\mathbf{k}'}^{vvvv} n_{v,\mathbf{k}'}) + \sum_{\mathbf{k}' \neq \mathbf{k}} (n_{u,\mathbf{k}'} - n_{v,\mathbf{k}'}) V_{\mathbf{k}-\mathbf{k}'}^{uvuv} \quad (3.48)$$

$$\tilde{\Omega}_{uv} = \frac{\mu_{uv} \xi}{2\hbar} + \frac{1}{\hbar} \sum_{\mathbf{k}' \neq \mathbf{k}} V_{\mathbf{k}-\mathbf{k}'}^{uvvu} p_{uv,\mathbf{k}'} - \frac{2}{\hbar} V_0^{uvuv} \sum_{\mathbf{k}'} p_{uv,\mathbf{k}'} \quad (3.49)$$

$$\tilde{\Omega}_{51'} = \frac{\Delta_{51'}}{2\hbar} + \frac{1}{\hbar} \sum_{\mathbf{k}' \neq \mathbf{k}} V_{\mathbf{k}-\mathbf{k}'}^{51'1'5} p_{51',\mathbf{k}'} - \frac{2}{\hbar} V_0^{51'51'} \sum_{\mathbf{k}'} p_{51',\mathbf{k}'} \quad (3.50)$$

where γ_{ijp} is the dephasing rate associated with energy levels i and j . γ_j is the intrasubband electron-electron scattering rate at level j , γ_{ij} is the combined electron-electron and electron-phonon scattering rate between levels i and j . γ_{sp} is the spontaneous emission rate. $T_{j,e}$ is the electron temperature at level j , T_l is the lattice temperature. $f_{j,\mathbf{k}}$ is the Fermi-Dirac distribution with chemical potential $\mu_{j,e}$ at level j . The chemical potentials and temperatures are determined by electron number conservation and energy conservation, which are described in the last section. The influence of the subband dispersion, namely the nonparabolicity, is represented by using the effective mass of electrons m^* . For subband j , we have $\varepsilon_{j,\mathbf{k}} = \varepsilon_j + \hbar^2 \mathbf{k}^2 / 2m_j^*$. For our mid-IR structure, For our mid-IR structure, we estimate that $m_s^* / m_4^* \approx 1.28$ [98, 104].

Using the above semiclassical laser theory and Maxwell's equations, the gain G are given by

$$G = -\frac{2\omega_\lambda}{\varepsilon_0 n c V_m \xi} \text{Im} \left[\sum_{\mathbf{k}} (\mu_{45} p_{45,\mathbf{k}} + \mu_{35} p_{35,\mathbf{k}}) \right] \quad (3.51)$$

3.2.3.2 Results and discussions

3.2.3.2.1 Effects of many-body interaction and non-parabolicity on optical gain

Figure 3 - 10 shows the computed gain spectra at resonance. In order to evaluate the influences of Coulomb interaction and non-parabolicity on optical properties for mid-IR QCLs, we firstly compare the gain spectra calculated from the microscopic model with “many-body + non-parabolicity” (considering both many-body and non-parabolicity effects), microscopic model

with “many-body + parabolicity” (considering both many-body and parabolicity effects), microscopic model with “free-carrier” (considering free carriers and non-parabolicity but neglecting the bandstructure and Rabi frequency renormalization) and the macroscopic density-matrix model (Ref. [68]), as shown in Fig. 3-10 (a). In contrast to the cases in the THz QCLs, the many-body Coulomb interaction exhibits a neglected effects on the lineshape of gain spectrum of mid-IR QCLs by the comparison of the microscopic models with “many-body + non-parabolicity” and “free-carrier”. Nevertheless, comparing the two gain spectra calculated by the microscopic models with “many-body + non-parabolicity” and “many-body + parabolicity”, we found that the subband dispersion with different subband effective masses (non-parabolicity) causes a strong shift of gain peak to the long wavelength side due to the redistribution of subband electrons in k space caused by the non-parabolicity effect. Moreover, the effects of non-parabolicity on optical gain spectrum will be further enhanced when a mid-IR QCL operates in shorter wavelength which has a larger non-parabolicity, as shown in Fig. 3-10(b). In addition, since the SDM model does not consider the kinetic distribution of electrons with conduction band and non-parabolicity, the obtained peak value and peak position of the optical gain are strongly overestimated when comparing the results obtained from the microscopic “many-body + non-parabolicity” model and the SDM model. Moreover, the spectral lineshape, which is important for calculating e.g. the linewidth enhancement factor, is not accurately predicted by the SDM model. Overall, the different spectral features e.g. different peak values, peak frequency positions and spectrum lineshapes from two models demonstrate that the microscopic density-matrix model can enrich in-depth understanding of optical properties of mid-IR QCLs and enables a more accurate prediction of the gain spectrum.

Fig. 3 - 10 Simulation results for gain spectra at resonance and 100 K. (a) The gain spectra calculated from microscopic model “many-body + non-parabolicity” (solid line), microscopic model “many-body + parabolicity” (dotted lines), microscopic model “free carriers” (dot-dashed lines) and SDM (dashed lines). (b) Gain spectra at the two effective mass ratios m_5/m_4 of electrons of 1.5 and 1.3 using the microscopic model “many body + non-parabolicity”.

3.2.3.2.2 Optical gain spectrum as a function of injection coupling strength

In order to further illustrate the effects of resonant tunneling, we simulate the gain spectra at different injection coupling strength, as shown in Fig. 3 – 11. As the injection coupling strength increases, the peak value of gain spectrum decrease but the lineshape of gain profile is not changed. The decrease of peak value is attributed to the increasing broadening due to energy splitting caused by the coherence of resonant tunneling. The unchanged gain profile demonstrates that the symmetric and ant-symmetric dressed states, which are composed of coupled states between the injection energy level and upper laser level, contribute equally to the optical gain.

Fig. 3 - 11 Gain spectra at different injection coupling strength with and without many-body interactions on gain spectra at resonance and 100 K.

3.2.3.2.3 Optical gain spectrum as a function of doping density

Fig. 3 - 12 (a) Effects of doping density on gain spectra at resonance and 100 K. From the bottom to the top for each kind of color line, the doping density is $0.76 \times 10^{11} \text{ cm}^{-2}$, $2.27 \times 10^{11} \text{ cm}^{-2}$, $3.79 \times 10^{11} \text{ cm}^{-2}$, and $5.30 \times 10^{11} \text{ cm}^{-2}$. The effects of doping density on lifetimes of energy levels are neglected. (b) Normalized gain spectra from microscopic model at different doping density.

Since the effects of non-parabolicity and many-body interaction depend on the doping density, we anticipate that doping will strongly affect the gain spectra, as shown in Fig. 3- 12. As the doping density increases, the spectra are further red-shifted. By the comparison of results from microscopic model and free-carrier model, we found that the redshift doesn't mainly result from the many-body interaction but from the non-parabolicity effects.

3.3 Conclusion

We have established the Hartree-Fock semiconductor Bloch equations with dephasing and scattering contributions treated at the level of a relaxation-rate approximation, which describes the electron-electron Coulomb interaction, nonparabolicity and coherence of resonant tunneling transport. We use the developed model to investigate the optical gain of both THz and mid-IR QCL. The simulation results calculated from the microscopic model with “many-body + nonparabolicity”, the microscopic model with “many-body + parabolicity”, the microscopic model with “free-carrier” and the SDM model are compared to demonstrate the importance of those parameters in the simulation of optical properties of both THz and mid-IR QCLs. The effects of energy detuning, injection and extraction coupling strength, and doping density on optical gain are also systematically investigated. The results show that the gain peak frequency calculated by the many-body model is closer to the experimentally measured lasing frequency,

compared with the SDM model. Specifically, in THz QCLs, both the many-body Coulomb interaction and nonparabolicity cause the red-shift of gain spectrum and reduce the peak gain. The interplay of various many-body interactions reveals that the optical spectral red-shift is mainly caused by the depolarization terms in THz QCLs, and this red-shift in our population inverted system is contrary to that in usual non-inverted absorption case. Furthermore, in THz QCLs, the gain spectrum is enhanced and slightly broadened as the injection coupling strength increases, while an increasing extraction coupling strength reduces the peak value and broadens the gain spectrum. When the extraction and injection coupling strengths have different value as to favor a side, the double-peaked gain spectrum is generated. In addition, as the doping density increases, the gain spectrum is red-shifted and modified. In contrast, for mid-IR QCLs, the most important modification to optical gain among the three factors is the non-parabolicity. The increasing injection coupling strength can only make the drop of optical gain peak but has few influences on the lineshape due to strong injection coupling strength in mid-IR QCLs. In addition, as the doping increases, the gain spectrum is slightly red-shifted.

4 Intrinsic linewidth in quantum cascade lasers

4.1 Introduction

The intrinsic noise of a laser is an important property that deserves careful study. Like all other lasers, QCLs can easily suffer from various noises, which play an important role in their laser performance especially in their spectral linewidth. An accurate analysis of the linewidth caused by these noises is necessary in order to ensure that QCLs can meet certain design specifications. Several research groups have measured the linewidth of QCLs ranging from a hundreds megahertz down to a few kilohertz or less, depending on the measurement techniques and the stabilization processes used [49, 105-113]. External noise factors such as mechanical vibrations, external environmental temperature variations and bias-current fluctuations [112, 114-117] can be potentially totally removed by various frequency-stabilization techniques, such as phase locking techniques. However, the intrinsic (fundamental) noises caused by *e.g.* spontaneous emission, thermal photon, and thermodynamical fluctuation cannot be overcome due to the fundamental quantum limitations [118]. These noises can induce the linewidth broadening of QCLs and reduce the performance. A thorough understanding of linewidth caused by these factors is increasingly important, as it is related to many practical applications *e.g.* trace-gas absorption spectroscopy and optical free-space data communication [41, 108, 109, 119-121]. In this chapter, we will mainly investigate the intrinsic linewidth source *i.e.* spontaneous emission and thermal photon. The effects of the critical transport mechanism *i.e.* resonant tunneling on intrinsic linewidth is included. The influences from coherent interaction of resonant tunneling transport and laser transition are also discussed. The linewidth broadening induced by thermodynamical fluctuation will be discussed in chapter 5.

4.2 Spontaneous emission induced linewidth broadening

4.2.1 Narrow intrinsic linewidth: rate-equation model

Recently, an ultra-narrow intrinsic linewidth of ~ 510 for mid-IR QCLs [106] and ~ 107 Hz [105] for THz QCLs has been experimentally observed. The results show that the intrinsic linewidth of QCLs is very narrow. They are two to three orders of magnitude narrower than those of conventional diode laser [11]. In addition to the small linewidth enhancement factor in QCLs [10], the spontaneous emission induced linewidth in a QCL is much narrower than that of diode laser. The rate-equation model can well explain the reason behind the narrow intrinsic

linewidth of QCLs [122]. In this section, we will derive the Schawlow-Townes linewidth formula by the rate-equation model followed the standard derivations [53, 122].

The linewidth of lasers can be attributed to the fluctuations of the phase of optical field due to the spontaneous emission events. We represent the complex amplitude E of laser field as

$$A = \sqrt{I}e^{i\phi} \quad (4.1)$$

where I and ϕ are the scalar intensity, which has been normalized to photon number, and phase.

The spontaneous emission is assumed to instantaneously alter the complex amplitude of laser field. The unit-magnitude change ΔA due to the p th spontaneous emission event is written as

$$\Delta A = e^{i(\phi + \theta_p)} \quad (4.2)$$

where θ_p is random. If only considering the spontaneous emission contribution, the event occurs at the rate of R_{sp} i.e. the rate of spontaneous emission of electrons in the upper laser level coupled into the lasing mode [53].

Fig. 4 - 1 The schematic of change of complex amplitude A induced by the spontaneous emission. The complex field amplitude A varies by ΔA having an amplitude of unity and phase of $\phi + \theta_p$.

As shown in Fig. 4 - 1, there are two contributions to the phase fluctuations i.e. phase change $\Delta\phi_p'$ due to the out-phase component of A and intensity variation coupled into phase change $\Delta\phi_p''$. The linewidth broadening induced by intensity and phase coupling can be represented by the α -parameter (namely linewidth enhancement factor) [53], it will be discussed in chapter 5. Here we only consider the phase change $\Delta\phi_p'$, and it can be written as

$$\Delta\phi_p' = (1/\sqrt{I})\sin(\theta_p) \quad (4.3)$$

Then the total phase change for $p=R_{sp}t$ spontaneous emission events is

$$\Delta\phi_p = \sum_{p=1}^{p=R_{sp}} \left(1/\sqrt{I}\right) \sin(\theta_p) \quad (4.4)$$

So we can obtain the average square phase change

$$\langle (\Delta\phi_p)^2 \rangle = \frac{R_{sp}}{2I} \quad (4.5)$$

The autocorrelation function of the field $A(t)$ can be expressed as [123]

$$\langle A^*(t)A(0) \rangle = |A(0)|^2 \exp\left(-\langle (\Delta\phi_p)^2 \rangle / 2\right) \quad (4.6)$$

Then we can obtain the linewidth caused by the spontaneous emission event by the Fourier transform of Eq. (4.6).

$$\delta f = \frac{R_{sp}}{4\pi I} \quad (4.7)$$

In order to investigate the linewidth of a QCL system, we should know the expression of photon number I and the rate of spontaneous emission of electrons in the upper laser level coupled into the lasing mode R_{sp} . We can achieve this by solving the rate equations of a QCL.

Fig. 4 - 2 Schematic of conduction band and wavefunction of a THz QCL. We treat the active region as an equivalent three-level-model system (inset). An effective lifetime of energy level 3 is used to characterize the relaxation of electrons to energy level 1.

Let us consider a three-level system, as shown in Fig. 4 - 2. Levels 1, 4 and 3 are the injector level, upper laser level and lower laser level. An effective lifetime of energy level 3 will be used

to characterize the relaxation of electrons to energy level 1. The rate equation of electron and photon number is written as [124]

$$\frac{dN_4}{dt} = \frac{I}{e} - \frac{N_4}{\tau_{43}} - \frac{N_4}{\tau_{41}} - \Gamma g(N_4 - N_3)N_{ph} \quad (4.8)$$

$$\frac{dN_3}{dt} = \frac{N_4}{\tau_{43}} - \frac{N_3}{\tau_{31}} + \Gamma g(N_4 - N_3)N_{ph} \quad (4.9)$$

$$\frac{dN_{ph}}{dt} = M\Gamma g(N_4 - N_3)N_{ph} - \frac{N_{ph}}{\tau_{ph}} \quad (4.10)$$

where g the differential gain coefficient, Γ the confinement factor of optical field, M is the number of gain stages in active region, τ_{ph} is the field decay time, γ_{3eff} is the effective relaxation rate from level 3 to level 1, $1/\tau_{43}$, $1/\tau_{41}$ and $1/\tau_{31}$ are the relaxation rate from level 4 to 3, level 4 to 1 and level 3 to 1, respectively.

By solving the Eqs.(4.8) - Eq.(4.10), we can get the electron and photon number

$$N_3 = \left(\frac{I}{e} \tau_{31} + \frac{1}{M\Gamma g \tau_{ph}} \right) \frac{\tau_{41}}{\tau_{31} + \tau_{41}} \quad (4.11)$$

$$N_{ph} = \left(1 - \frac{\tau_{31}}{\tau_{43}} \right) \frac{M \tau_{ph} \tau_{41}}{\tau_{31} + \tau_{41}} \frac{(I - I_{th})}{e} \quad (4.12)$$

where

$$I_{th} = \left(\frac{e}{M\Gamma g \tau_{ph}} \right) \frac{\tau_{41} + \tau_{43}}{\tau_{41} \tau_{43} (1 - \tau_{31}/\tau_{43})} \quad (4.13)$$

So we can obtain the expression of linewidth of a QCL

$$\begin{aligned} \delta f &= \frac{R_{sp}}{4\pi I} = \frac{M\Gamma g N_3}{4\pi N_{photon}} \\ &= \frac{\tau_{43} \Gamma g / \tau_{ph}}{4\pi (I/I_{th} - 1)} \left[\frac{I}{I_{th}} \left(\frac{\tau_{31}}{\tau_{41}} + \frac{\tau_{31}}{\tau_{43}} \right) \left(1 - \frac{\tau_{31}}{\tau_{43}} \right)^{-1} + 1 \right] \end{aligned} \quad (4.14)$$

Typically, the differential gain coefficient of QCLs is $\Gamma g \approx 10^{-3}$, which is much larger than that of interband semiconductor laser ($\Gamma g = 10^{-5} \sim 10^{-4}$) [122]. But the nonradiative emission

lifetime ($\tau_{43}=1\sim 10$ ps) of the upper laser level in a QCL is two to three orders of magnitude smaller than that of interband semiconductor laser ($\tau_4 = 1\sim 10$ ns) [125]. While their photon lifetimes are in the same order of magnitude. Therefore, according to Eq.(4.14), QCLs have a much smaller linewidth than interband semiconductor lasers.

4.2.2 Effects of resonant tunneling and coherent interaction: quantum Langevin approach

Although the rate-equation model can well explain the reason behind the narrow intrinsic linewidth of QCLs [122], it cannot adequately describe the electron transport characteristics of QCLs, and therefore cannot accurately demonstrate the actual linewidth feature. In the rate-equation model, localization of wavefunctions due to the dephasing scattering is overlooked which can lead to unrealistic results and limit the usefulness of the model. As reported in Refs. [36, 67, 68, 72, 73], the effects of resonant tunneling and dynamics of coherent interaction (influenced by dephasing) are important when describing the transport of QCLs. The coherent (or incoherent) injection coupling plays an important role in the electrical injection process to populate the upper radiative level, which affects the electron populations associated with stimulated emission processes and hence the ratio of spontaneous emission coupled into the lasing mode to the net stimulated emission. As a result, the resonant tunneling and the dynamics of coherent interaction may finally affect the intrinsic linewidth of QCLs. In addition, the rate equation model uses the operating pumping current as an input parameter, thus knowledge on linewidth reduction through changing the active regions of QCLs, e.g. the coupling strength and the doping level, is restricted before obtaining experimental data.

The rate equation model also assumes that the polarization follows the other dynamic variables adiabatically, and therefore the dynamics of coherent interaction associated with laser transitions are neglected. But, it is correct only if the polarization relaxation constant is much larger than other decay constant. When the polarization relaxation constant becomes comparable to the field decay rate, we need to consider the effects of polarization dynamics on the laser linewidth. Since some THz QCLs can have a narrow width of gain spectrum (*e.g.* 0.22 THz [126]) and hence relatively long polarization relaxation constant compared to bipolar semiconductor diode lasers, which is comparable to the field decay rate. Hence, the investigation on the intrinsic linewidth of QCLs should consider the effect of dynamics of polarization.

Since the resonant-tunneling transport and dynamics of coherent interaction cannot be properly included in the classical rate equation model, one has to refer to the more fundamental quantum mechanical model. The three principal methods used for this purpose are the quantum Langevin equations for the field operator and electron operator, the master equation for the density operator and *Fokker-Planck* equation for the *Glauber-Sudarshan P*-representation of density operator [127-129]. The quantum Langevin approach is relatively more intuitive and technically simpler [130]. We will refer to this model. In this section, we report a new model for calculation of intrinsic linewidth of QCLs based on the quantum Langevin equations. It differs from the results derived from the classical rate equation model in that the resonant-tunneling effects and dynamics of coherent interaction are considered. The results show that the coupling strength and the dephasing rate associated with resonant tunneling strongly affect the linewidth of QCLs in the incoherent resonant-tunneling transport regime but only induce little influence in the coherent regime. Finally, the intrinsic linewidth reduction through optimization of the doping density and lifetime of different energy levels is presented.

4.2.2.1 Quantum Langevin equations

Fig. 4 - 3 Conduction band diagram of a resonant-phonon THz QCL with a diagonal design in the “tight-binding” scheme. $\Omega_{41'}$ is the injection coupling strength (unit: eV). $\hbar\gamma$ is the electron-light interaction. For the sake of simplification, we only consider the three energy levels i.e. levels 1', 3 and 4, which are the injector level, the lower laser level and the upper laser level, respectively. An effective lifetime of energy level 3 is used to characterize the relaxation of electrons into level 1. Because the coupling between lower laser level and extraction level are usually designed to be large and the electron transport through the extraction barrier is in the coherent transport regime, this

simplification will not cause a big error. The inset showing the simplified energy levels with resonant tunneling and optical response.

We consider the same resonant-phonon THz QCL system that we had taken into account in chapter 3. But here, for the sake of simplification, we neglect the resonant-tunneling coupling between the lower laser level and extraction level, as shown in Fig. 4 - 3, while only considering the injection coupling between the injector level and upper laser level. Because the coupling between lower laser level and extraction level are designed to be large and the electrons coherently go through the extractor barrier, we think that this simplification will not cause a big error. We treat the active region as three-energy-level system i.e. levels 1', 3 and 4, which are the injector level, the lower laser level and the upper laser level, respectively. $\Omega_{41'}$ denotes the interaction of the resonant tunneling, $\hbar g$ is the electron-light interaction. The electrons in level 1' are injected into the upper laser level 4 by tunneling transport. Then, they relax from level 4 to level 3 by emitting photons. An effective lifetime of energy level 3 is used to characterize the relaxation of electrons into level 1. The Hamiltonian of this three-level QCLs system in the *rotating-wave approximation* can be expressed as [131]

$$H = \hbar\omega_\lambda c^\dagger c + \sum_{j=1',4,3} \varepsilon_j b_j^\dagger b_j + \hbar \left(g^* c^\dagger b_3^\dagger b_4 + g b_4^\dagger b_3 c \right) - (\Delta_{41'}/2) (b_4^\dagger b_{1'} + b_{1'}^\dagger b_4) \quad (4.15)$$

where ω_λ is the single-mode field lasing angular frequency, ε_j is the energy of level j , $c^\dagger (c)$ and $b_j^\dagger (b_j)$ denote the creation (annihilation) operator of the laser field and electrons in the subband j , respectively. The coupling constant g is given by

$$g = \frac{e\mu}{\hbar} \sqrt{\frac{\hbar\omega_\lambda\Gamma}{2\varepsilon_0 n^2 V_m}} \quad (4.16)$$

where μ is the radiative dipole matrix element, Γ is the confinement factor of the optical mode that overlaps with the entire active region, ε_0 is the vacuum permittivity, n is the refractive index, V_m is the volume of one single gain stage in the active region.

The operators corresponding to the observables of our interest are the single-mode intracavity slowly varying laser field operator $a = c \exp(i\omega_\lambda t)$, polarization operator $\sigma_{ij} = b_i^\dagger b_j \exp(i\omega_\lambda t)$ and electron population operator $\rho_j = b_j^\dagger b_j$ in level j . By including the noise operators f_{ij} and f_j the

quantum Langevin equations which rule the dynamics of the laser are derived according to the Hershberger's equation

$$\frac{da}{dt} = -\frac{\kappa}{2}a - ig^* \sigma_{34} + f_a(t) \quad (4.17)$$

$$\frac{d\sigma_{34}}{dt} = -\gamma_{34}\sigma_{34} + ig(\rho_4 - \rho_3)a + f_{34}(t) \quad (4.18)$$

$$\frac{d\sigma_{41'}}{dt} = -\gamma_{41'}\sigma_{41'} - i\frac{\Delta_{41'}}{\hbar}\sigma_{41'} - \frac{i\Omega_{41'}}{2\hbar}(\rho_{1'} - \rho_4) + f_{41'}(t) \quad (4.19)$$

$$\frac{d\rho_4}{dt} = -\gamma_{sp}\rho_4 - \gamma_4\rho_4 + i(g^*a^\dagger\sigma_{34} - g\sigma_{34}^\dagger a) - \frac{i\Omega_{41'}}{2\hbar}(\sigma_{41'}^\dagger - \sigma_{41'}) + f_4(t) \quad (4.20)$$

$$\frac{d\rho_3}{dt} = \gamma_{sp}\rho_4 + (1/\tau_{43})\rho_4 - \gamma_{3eff}\rho_3 + i(g\sigma_{34}^\dagger a - g^*a^\dagger\sigma_{34}) + f_3(t) \quad (4.21)$$

Where $\Delta_{41'}$ is the energy detuning between level 4 and $1'$. γ_{sp} is the rate of spontaneous emission coupling into all the field modes except for the laser mode, κ is the field damping rate, γ_4 is the total relaxation rate of level 4, γ_{3eff} is the effective relaxation rate from level 3 to level 1, $1/\tau_{43}$ is the relaxation rate from level 4 to 3, $\gamma_{ij} = \gamma_i/2 + \gamma_j/2 + T_2^*$ is the polarization decay rate (dephasing rate) with a phenomenological pure dephasing rate T_2^* due to various scattering e.g. interface roughness and impurity scattering.

The coupling efficiency β of the spontaneous emission into the lasing mode is given by the total spontaneous emission rate τ_{sp} as [132-134]

$$\beta = 2gg^* \tau_{sp} / \gamma_{23} \quad (4.22)$$

Then the γ_{sp} is given as

$$\gamma_{sp} = (1 - \beta) / \tau_{sp} \quad (4.23)$$

In the above equations, we neglect the polarization $\sigma_{31'}$ driven by higher quantum coherence effects. This assumption is, in general, valid for mid-IR designs due to a large energy separation Δ_{43} and also reasonably valid for THz QCLs due to thicker injector barriers [36].

The Langevin noise operators are fully defined by their first- and second-order moments. For the field Langevin forces $f_a(t)$ one gets [128, 131]

$$\langle f_a(t) \rangle = 0 \quad (4.24)$$

$$\langle f_a^\dagger(t) f_a(t') \rangle = \kappa n_{th} \delta(t - t') \quad (4.25)$$

$$\langle f_a(t) f_a^\dagger(t') \rangle = \kappa (n_{th} + 1) \delta(t - t') \quad (4.26)$$

$$\langle f_a^\dagger(t) f_a^\dagger(t') \rangle = \langle f_a(t) f_a(t') \rangle = 0 \quad (4.27)$$

where n_{th} is the average number of thermal photons in the laser cavity at temperature T_e (approximate electron temperature at the quasi-thermal equilibrium) and can be written as

$$n_{th} = [\exp(\hbar \omega_\lambda / k_B T_e) - 1]^{-1} \quad (4.28)$$

For the electron population and polarization noise operators, the correlation functions can be derived according to the *generalized Einstein relations* [128, 131], as explained in the Appendix D.

4.2.2.2 Laser intrinsic linewidths

4.2.2.2.1 Equivalent c -number Langevin equations

To solve the present problem, we have to convert the above operator equations into c -number equations [131]. For this we have to choose certain particular ordering for field and electron density operators, because the c -number variables commute with each other while the operators do not. Here we choose the normal ordering of field and electron density operators to be $a^\dagger, \sigma_{34}^\dagger, \sigma_{41'}^\dagger, \rho_4, \rho_3, \sigma_{41'}, \sigma_{34}, a$, that is, the stochastic c -number variables corresponding to the operators $a, \sigma_{34}, \sigma_{41'}, \rho_4, \rho_3$ are replaced with their classical counterparts $A, p_{34}, p_{41'}, n_4, n_3$. Then the c -number Langevin equations of Eqs. (4.17)-(4.21) can be written as

$$\frac{dA}{dt} = -\frac{\kappa}{2} A - ig^* p_{34} + F_A(t) \quad (4.29)$$

$$\frac{dp_{34}}{dt} = -\gamma_{34} p_{34} + ig (n_4 - n_3) A + F_{34}(t) \quad (4.30)$$

$$\frac{dp_{41'}}{dt} = -\gamma_{41'} p_{41'} - i \frac{\Delta_{41'}}{\hbar} p_{41'} - \frac{i\Omega_{41'}}{2\hbar} (n_{1'} - n_4) + F_{41'}(t) \quad (4.31)$$

$$\frac{dn_4}{dt} = -\gamma_{sp}n_4 - \gamma_4n_4 + i\left(g^*A^\dagger p_{34} - gp_{34}^\dagger A\right) - \frac{i\Omega_{41'}}{2\hbar}(p_{41'}^\dagger - p_{41'}) + F_4(t) \quad (4.32)$$

$$\frac{dn_3}{dt} = \gamma_{sp}n_4 + (1/\tau_{43})n_4 - \gamma_{3eff}n_3 + i\left(gp_{34}^\dagger A - g^*A^\dagger p_{34}\right) + F_3(t) \quad (4.33)$$

Here, the Langevin forces F_μ have the following properties,

$$\langle F_\mu(t) \rangle = 0 \quad (4.34)$$

$$\langle F_u^\dagger(t)F_v(t') \rangle = 2D_{u^\dagger v} \delta(t-t') \quad (4.35)$$

The diffusion coefficients $D_{u^\dagger v}$ of c -number Langevin forces may be different from the corresponding diffusion coefficients $d_{u^\dagger v}$ of the operator Langevin forces in Appendix D. The diffusion coefficients $D_{u^\dagger v}$ are determined in such a way that the second moments calculated from the c -number equations agree with those calculated from the operator equations as explained in Appendix E.

4.2.2.2.2 Steady-state solution for above-threshold operation

The steady-state solutions for the mean values of the field and electron number variables above threshold are obtained by setting the time derivatives to be zero and dropping the noise terms in Eqs.(4.29)-(4.33). Neglecting the correlations between the electron populations and the photons, one then finds for the steady-state mean photon numbers, population inversion and population of the upper and lower laser levels

$$I_0 = A_0^2 = \frac{\gamma_{3eff}n_{3,0} - \gamma_{sp}n_{4,0} - (1/\tau_{43})n_{4,0}}{\kappa} \quad (4.36)$$

$$n_{4,0} - n_{3,0} = \frac{\gamma_{23}\kappa}{2gg^*} \quad (4.37)$$

$$n_{4,0} = \left[-\frac{\gamma_{3eff}\gamma_{34}\kappa}{2gg^*} - \left(\frac{\Omega_{41'}}{2\hbar}\right)^2 \left(N_0 + \frac{\gamma_{34}\kappa}{2gg^*} \right) \frac{2\gamma_{41'}}{(\gamma_{41'})^2 + (\Delta_{41'}/\hbar)^2} \right] \bigg/ \left[1/\tau_{43} - \gamma_4 - \gamma_{3eff} - \left(\frac{\Omega_{41'}}{2\hbar}\right)^2 \frac{6\gamma_{41'}}{\gamma_{41'}^2 + (\Delta_{41'}/\hbar)^2} \right] \quad (4.38)$$

$$n_{3,0} = \left[-\frac{\gamma_{34}(1/\tau_{43} - \gamma_4)\kappa}{2gg^*} + \left(\frac{\Omega_{41'}}{2\hbar}\right)^2 \left(\frac{\gamma_{34}\kappa}{gg^*} - N_0 \right) \frac{2\gamma_{41'}}{\gamma_{41'}^2 + (\Delta_{41'}/\hbar)^2} \right] \bigg/ \left[1/\tau_{43} - \gamma_4 - \gamma_{3eff} - \left(\frac{\Omega_{41'}}{2\hbar}\right)^2 \frac{6\gamma_{41'}}{\gamma_{41'}^2 + (\Delta_{41'}/\hbar)^2} \right] \quad (4.39)$$

where N_0 is the electron number of one module in the cavity.

On the other hand, the steady-state polarization can be expressed in terms of the mean value of the field as

$$p_{34,0} = \frac{\kappa}{i2g} A_0 \quad (4.40)$$

$$p_{41',0} = -\frac{i\Omega_{41'}}{2\hbar} (N_0 - n_{3,0} - 2n_{4,0}) \left/ \left(\gamma_{41'} + i \frac{\Delta_{41'}}{\hbar} \right) \right. \quad (4.41)$$

From Eq. (4.40), we find that if A_0 is real, $p_{34,0}$ is purely imaginary. Therefore, we can choose the mean value of the phase of the laser field to be zero for simplification.

The current at and above threshold can be derived as, respectively

$$I_{c,th} = |e| \left(\frac{n_{4,th}}{\tau_{41}} + \frac{n_{3,th}}{\tau_{31}} \right) = |e| \gamma_{34} \kappa \frac{\gamma_{3eff} / 2\tau_{41} + (\gamma_{sp} + 1/\tau_{43}) / 2\tau_{31}}{(\gamma_3 - \gamma_{sp} - 1/\tau_{43}) g g^*} \quad (4.42)$$

$$I_c = |e| \left(\frac{n_{4,0}}{\tau_{41}} + \frac{n_{3,0}}{\tau_{31}} \right) = |e| \frac{\left(\frac{\Omega_{41'}}{2\hbar} \right)^2 \left[-\frac{(\tau_{41} + \tau_{31})N_0}{\tau_{41}\tau_{31}} + \frac{(2\tau_{41} - \tau_{31})\gamma_{34}\kappa}{2\tau_{41}\tau_{31}g g^*} \right] \frac{2\gamma_{41'}}{(\gamma_{41'})^2 + (\Delta_{41'}/\hbar)^2}}{1/\tau_{43} - \gamma_4 - \gamma_{3eff} - \left(\frac{\Omega_{41'}}{2\hbar} \right)^2 \frac{6\gamma_{41'}}{\gamma_{41'}^2 + (\Delta_{41'}/\hbar)^2}} \quad (4.43)$$

where τ_{41} and τ_{31} is the lifetime from the upper and lower laser level to injector level, respectively. It is noted that current contributions are made by electrons from the second phonon energy level to injector level in addition to the transition from the upper level to injector level for two-phonon resonance mid-IR QCLs.

4.2.2.2.3 Dynamics of fluctuations around steady state

To determine the frequency noise spectrum and linewidth we need first of all to investigate the small fluctuations of the field and electron number variables around the steady state. Neglecting terms of the second and higher order in the fluctuations, we set

$$A = A_0 + \delta A \quad (4.44)$$

$$p_{34} = p_{34,0} + \delta p_{34} \quad (4.45)$$

$$p_{41'} = p_{41',0} + \delta p_{41'} \quad (4.46)$$

$$n_4 = n_{4,0} + \delta n_4 \quad (4.47)$$

$$n_3 = n_{3,0} + \delta n_3 \quad (4.48)$$

Thus, we get the following equations for fluctuations

$$\frac{d\delta A}{dt} = -\frac{\kappa}{2}\delta A - ig^* \delta p_{34} + F_A(t) \quad (4.49)$$

$$\frac{d\delta p_{34}}{dt} = -\gamma_{34}\delta p_{34} + ig(\delta n_4 - \delta n_3)A_0 + ig(n_{4,0} - n_{3,0})\delta A + F_{34}(t) \quad (4.50)$$

$$\frac{d\delta p_{41'}}{dt} = -\gamma_{41'}\delta p_{41'} - i\frac{\Delta_{41'}}{\hbar}\delta p_{41'} - \frac{i\Omega_{41'}}{2\hbar}(\delta n_{1'} - \delta n_4) + F_{41'}(t) \quad (4.51)$$

$$\begin{aligned} \frac{d\delta n_4}{dt} = & -\gamma_{sp}\delta n_4 - \gamma_4\delta n_4 + i(g^*\delta A^\dagger p_{34,0} - gp_{34,0}\delta A) + i(g^*A_0\delta p_{34} - g\delta p_{34}^\dagger A_0) \\ & - \frac{i\Omega_{41'}}{2\hbar}(\delta p_{41'}^\dagger - \delta p_{41'}) + F_4(t) \end{aligned} \quad (4.52)$$

$$\frac{d\delta n_3}{dt} = \gamma_{sp}\delta n_4 + (1/\tau_{43})\delta n_4 - \gamma_{3eff}\delta n_3 + i(gp_{34,0}\delta A - g^*\delta A^\dagger p_{34,0}) + i(g\delta p_{34}^\dagger A_0 - g^*A_0\delta p_{34}) + F_3(t) \quad (4.53)$$

These equations can now be solved exactly by taking the Fourier transform of $\delta n_4(\omega)$,

$$\delta n_4(\omega) = \int_{-\infty}^{+\infty} e^{-i\omega t} \delta n_4(t) dt \quad (4.54)$$

as well as all the other variables. Then, one gets

$$i\omega\delta A(\omega) = -(\kappa/2)\delta A(\omega) - ig^*\delta p_{34}(\omega) + F_A(\omega) \quad (4.55)$$

$$i\omega\delta p_{34}(\omega) = -\gamma_{34}\delta p_{34}(\omega) + ig[\delta n_4(\omega) - \delta n_3(\omega)]A_0 + ig(n_{4,0} - n_{3,0})\delta A(\omega) + F_{34}(\omega) \quad (4.56)$$

$$i\omega\delta p_{41'}(\omega) = -\gamma_{41'}\delta p_{41'}(\omega) - i\frac{\Delta_{41'}}{\hbar}\delta p_{41'}(\omega) - \frac{i\Omega_{41'}}{2\hbar}[\delta n_{1'}(\omega) - \delta n_4(\omega)] + F_{41'}(\omega) \quad (4.57)$$

$$\begin{aligned} i\omega\delta n_4(\omega) = & -\gamma_{sp}\delta n_4(\omega) - \gamma_4\delta n_4(\omega) + i[g^*\delta A^\dagger(-\omega)p_{34,0} - gp_{34,0}\delta A(\omega)] \\ & + i[g^*A_0\delta p_{34}(\omega) - g\delta p_{34}^\dagger(-\omega)A_0] - \frac{i\Omega_{41'}}{2\hbar}[\delta p_{41'}^\dagger(-\omega) - \delta p_{41'}(\omega)] + F_4(\omega) \end{aligned} \quad (4.58)$$

$$\begin{aligned} i\omega\delta n_3(\omega) = & \gamma_{sp}\delta n_4(\omega) + (1/\tau_{43})\delta n_4(\omega) - \gamma_{3eff}\delta n_3(\omega) \\ & + i[gp_{34,0}\delta A(\omega) - g^*\delta A^\dagger(-\omega)p_{34,0}] + i[g\delta p_{34}^\dagger(-\omega)A_0 - g^*A_0\delta p_{34}(\omega)] + F_3(\omega) \end{aligned}$$

(4.59)

where the frequency-dependent fluctuation forces satisfy

$$\langle F_u(\omega) F_v(\omega') \rangle = 4\pi D_{uv} \delta(\omega + \omega') \quad (4.60)$$

The solution of this linear system can then be easily obtained.

4.2.2.2.4 Noise spectra

To calculate noise spectra, we use the semiclassical expression of the laser field

$$A = \sqrt{I} \exp(i\varphi) \quad (4.61)$$

Then the dynamical equations of phase fluctuations are obtained as

$$\delta\varphi(t) = \frac{i}{2\sqrt{I_0}} [\delta A^\dagger(t) - \delta A(t)] \quad (4.62)$$

By taking Fourier transform of Eq. (4.62) on both sides, one gets

$$\delta\varphi(\omega) = \frac{i}{2\sqrt{I_0}} [\delta A^\dagger(-\omega) - \delta A(\omega)] \quad (4.63)$$

According to Eq. (4.55), the Fourier amplitudes of the field fluctuations can be expressed as

$$\delta A(\omega) = \frac{ig\delta p_{34}(\omega) + F_A(\omega)}{i\omega + \kappa/2} \quad (4.64)$$

where

$$\delta p_{34}(\omega) = \frac{ig[\delta n_4(\omega) - \delta n_3(\omega)]A_0 + ig(n_{4,0} - n_{3,0})\frac{F_A(\omega)}{i\omega + \kappa/2} + F_{34}(\omega)}{i\omega + \gamma_{34} - \frac{gg^*(n_{4,0} - n_{3,0})}{i\omega + \kappa/2}} \quad (4.65)$$

Therefore, with applying Eq.(4.64) and Eq.(4.65) into Eq.(4.63), the phase fluctuations can be explicitly written as

$$\delta\varphi(\omega) = \frac{i}{2\sqrt{I_0}} \frac{(i\omega + \gamma_{34})[F_A^\dagger(-\omega) - F_A(\omega)] + ig[F_{34}^\dagger(-\omega) - F_{34}(\omega)]}{(i\omega + \gamma_{34})(i\omega + \kappa/2) - gg^*(n_{4,0} - n_{3,0})} \quad (4.66)$$

The autocorrelation function of the phase fluctuations can be expressed as

$$\langle \delta\varphi(\omega) \delta\varphi^*(\omega') \rangle = (\delta\varphi)_\omega^2 \delta(\omega - \omega') \quad (4.67)$$

According to the Wiener-Khinchine theorem [123], the relations between autocorrelation function and power spectral density satisfy

$$\langle \delta\varphi(\omega) \delta\varphi^*(\omega') \rangle = 2\pi S_\varphi \delta(\omega - \omega') \quad (4.68)$$

where the power spectral density is defined as the following Fourier transform

$$S_\varphi = \int_{-\infty}^{+\infty} \langle \delta\varphi(t) \delta\varphi(t - \tau) \rangle \exp(-i\omega\tau) d\tau \quad (4.69)$$

The frequency noise spectral density S_f , therefore, can be derived as

$$S_f = \omega^2 S_\varphi = \frac{\kappa(\omega^2 + \gamma_{34}^2)(2n_h + 1) + 2gg^* \gamma_{34} n_{4,0}}{4I_0 [\omega^2 + (\gamma_{34} + \kappa/2)^2]} \quad (4.70)$$

Once we know the power spectrum density of the frequency noise, the linewidth can then be obtained by the following derivation. The autocorrelation function of the lasing field $A(t)$ can be explicitly derived for single-mode QCLs when the phase fluctuations $\delta\varphi(t)$ are considered as a Gaussian distribution [123].

$$\langle A(t) A^*(t - \tau) \rangle = I_0 \exp(i\omega_\lambda t) \exp(-\langle \delta\varphi^2 \rangle / 2) \quad (4.71)$$

where we have neglected the amplitude fluctuations. The phase change induced by the amplitude change can be characterized by the α -parameter (linewidth enhancement factor) [53]. The overall linewidth should be multiplied by $1 + \alpha^2$ if this factor is considered. This parameter is not included in our model in this section, and we will discuss it in the chapter 6.

In general, if the spectral density of the frequency fluctuation is known, the mean squared value of the phase change can be obtained as [57]

$$\langle \delta\varphi^2 \rangle = \frac{2}{\pi} \int_0^\infty \frac{S_f(\omega)}{\omega^2} [1 - \cos(\omega\tau)] d\omega \quad (4.72)$$

namely

$$\begin{aligned} \langle \delta\varphi^2 \rangle = & \frac{2\kappa n_{th} + \kappa + 2(gg^*/\gamma_{34})n_{4,0}}{4(1 + \kappa/2\gamma_{34})^2 I_0} t \\ & + \frac{-2\kappa^2 n_{th}/\gamma_{34} - \kappa^2/\gamma_{34} + 2(gg^*/\gamma_{34})n_{4,0}}{4(1 + \kappa/2\gamma_{34})^2 I_0} \left[\frac{e^{-(\gamma_{34} + \kappa/2)|t|} - 1}{\gamma_{34} + \kappa/2} \right] \end{aligned} \quad (4.73)$$

The linewidth is then obtained from the following spectral density by taking the Fourier transform of Eq. (4.73)

$$S_A(\omega) = I_0 \int_{-\infty}^{+\infty} \exp[-i(\omega - \omega_\lambda)t] \exp(-\langle \delta\varphi^2 \rangle/2) dt \quad (4.74)$$

Then the intrinsic linewidth of the laser can then be expressed as

$$\begin{aligned} \delta f = & \frac{\kappa(2n_{th} + 1) + 2(gg^*/\gamma_{34})n_{4,0}}{8\pi(1 + \kappa/2\gamma_{34})^2 I_0} \\ \simeq & \frac{\kappa n_{th} + 2(gg^*/\gamma_{34})n_{4,0}}{4\pi(1 + \kappa/2\gamma_{34})^2 I_0} \end{aligned} \quad (4.75)$$

4.2.2.3 Results and discussions

In Eq. (4.75), n_{th} is due to the contribution of the incoming fluctuation of thermal photon, and it will be discussed in the next section of this chapter. If we only consider the contribution of spontaneous emission, equation (4.75) becomes

$$\delta f = \frac{(2gg^*/\gamma_{34})n_{4,0}}{4\pi(1 + \kappa/2\gamma_{34})^2 I_0} \quad (4.76)$$

By comparing the first line of Eq. (4.14) and Eq.(4.76), and with the following connections,

$$n_{4,0} \sim N_4 \quad (4.77)$$

$$I_0 \sim N_{photon} \quad (4.78)$$

$$\kappa \sim \gamma \quad (4.79)$$

$$2(gg^*/\gamma_{23}) \sim M\Gamma g \quad (4.80)$$

we can deduce that the linewidth deduced from the quantum mechanical Langevin model is different from the expression based on the rate-equation model. The explicit difference between these two formulas is caused by the coefficient $(1 + \kappa/2\gamma_{34})^2$. In the rate equation model, $\gamma_{34} \gg \kappa$ is assumed, and the polarization of the active medium is adiabatically eliminated. Hence, the information on dynamics of coherent interaction associated the laser transition is missed. The inclusion of the coherent interaction can induce a smaller linewidth by a factor of $(1 + \kappa/2\gamma_{34})^2$ if the coherent time is comparable to the cavity loss according to Eq.(4.76). The intrinsic difference between these two models is from the resonant-tunneling effects. Electrons in the ground injector level are injected into the upper laser level in QCLs through resonant tunneling. Since coherence plays an important role in the resonant-tunneling mechanism, which can significantly influence the electron transport [36, 67, 68, 72, 73], it is necessary to include this factor in laser dynamics simulations of QCLs.

Fig. 4 - 4 The comparisons of linewidths derived from our model and the classical rate equation model at different operation currents (a) in THz QCLs. The default parameters are chosen from the typical values for resonant-tunneling injection THz QCLs designs [36]: $\gamma_{34} = 4.5 \times 10^{12} \text{ s}^{-1}$, $\gamma_{41'} = 2.5 \times 10^{12} \text{ s}^{-1}$, $\kappa = 2 \times 10^{11} \text{ s}^{-1}$, $1/\tau_{43} = 3 \times 10^{11} \text{ s}^{-1}$, $\gamma_4 = 5 \times 10^{11} \text{ s}^{-1}$, $\gamma_{3eff} = 2 \times 10^{12} \text{ s}^{-1}$, $\epsilon_{41'} = 0$, $\Omega_{41'} = 1.5 \text{ meV}$, $\mu = 3.7 \text{ nm}$, $\Gamma \approx 1$, $n = 3.6$, $T = 20 \text{ K}$, the doping sheet density is $3 \times 10^{10} \text{ cm}^{-2}$. (b) in Mid-IR QCLs. The default parameters are chosen from two phonon resonance mid-IR QCLs designs [135]: $\gamma_{34} = 20 \times 10^{12} \text{ s}^{-1}$, $\kappa = 2 \times 10^{11} \text{ s}^{-1}$, $1/\tau_{43} = 3 \times 10^{11} \text{ s}^{-1}$, $\gamma_4 = 8 \times 10^{11} \text{ s}^{-1}$, $\gamma_{3eff} = 4 \times 10^{12} \text{ s}^{-1}$, $\gamma_{41'} = 15 \times 10^{12} \text{ s}^{-1}$, $\epsilon_{31'} = 0$, $\Omega_{41'} = 8 \text{ meV}$, $\mu = 2.1 \text{ nm}$, $\Gamma \approx 0.5$, $n = 3.2$, $T = 300 \text{ K}$, the doping sheet density is $2 \times 10^{11} \text{ cm}^{-2}$.

Figure 4 - 4 shows the comparisons of the laser linewidths derived from the quantum mechanical Langevin model and the classical rate-equation model at different operation currents

in both THz (Fig. 4 - 4(a)) and mid-IR (Fig. 4 - 4(b)) QCLs. With the comparison of these two models, the apparent difference for THz QCLs and relatively smaller difference for mid-IR QCLs are caused by including the resonant-tunneling transport effect and coherent interaction in the quantum mechanical Langevin model. These differences increase as the operation current increases. Because the effects of resonant tunneling increases when the injector level and upper laser level tend to be aligned as the increasing operation current.

The role of coherence in resonant-tunneling transport in QCLs has been investigated [36, 67, 68, 72, 73]. It has been suggested that the inclusion of coherent transport and dephasing in calculations is essential especially when transport is dominated by transitions between weakly coupled states. Here we are mainly concerned about how coupling strength and dephasing rate associated with resonant tunneling influence the intrinsic linewidth. Figure 4 - 5 shows the effects of the injection coupling strength on the linewidth at resonance. As shown in Fig. 4 - 5(a), the linewidth decreases as the injection coupling strength increases. When the injection coupling strength is below 3 meV, the linewidth diminishes rapidly. But once the coupling strength exceeds 3 meV, there only exists slight changes. This can be attributed to the coherence of electrons transport across the injector barrier, and this coherence can be determined by the factor $(\Omega_{41'}/\hbar)^2 (\gamma_{31'}\gamma_3)^{-1}$ ($\gg 1$ coherent) and ($\ll 1$ incoherent) [36]. The small injection coupling strength corresponds to the regime of incoherent resonant-tunneling transport. But when the coupling becomes strong, the resonant-tunneling transport tends to be coherent. As a result, more excited electrons are injected into upper laser level, and then stimulated to the lower laser level by emitting photons, current density and photon number increases, as shown in Fig. 4 - 5(b). Hence, the noise associated with the spontaneous emission is strongly suppressed, leading to a major reduction in the linewidth. Therefore, THz QCLs will have a much larger linewidth for structures with largely incoherent resonant-tunneling transport, but a smaller linewidth with coherent resonant-tunneling transport. Since the injector coupling strength of THz QCLs based on resonant-tunneling injection is typically 1~2 meV [65], the resonant tunneling can strongly influence the linewidth of THz QCLs.

Fig. 4 - 5 The effects of coupling strength on the linewidth of THz QCLs at resonance. (a) Linewidth as a function of coupling strength. The linewidth decreases as the coupling strength between the injector level and the upper laser level increases. The factor $(\Omega_{41'}/\hbar)^2 (\gamma_{41'}\gamma_4)^{-1}$ is used to determine the transition of resonant tunneling from coherent ($\gg 1$) to incoherent ($\ll 1$) (Ref.[36]). Coupling strengths more than 3 meV lay in the coherent regime. (b) Current density and photon number as a function of coupling strength.

Since the resonant-tunneling transport can be strongly influenced by the scattering, the linewidth has a strong dependence on the damping of the coherent interaction. Fig. 4 - 6(a) shows the effects of dephasing rate associated with resonant-tunneling transport on the linewidth. When the dephasing rate $\gamma_{41'}$ increases, only a small fraction of the electrons tunnels through the injector barrier into the upper laser level. As a result, the current density and photon number decreases (Fig. 4 - 6(b)) and the noise associated with spontaneous emission becomes strong. Therefore, the linewidth increases as the dephasing rate increases.

Fig. 4 - 6 The effects of dephasing rate associated with resonant tunneling on the linewidth of THz QCLs at resonance. (a) Linewidth as a function of dephasing rate. The linewidth increases as the dephasing rate increases. (b) Current density and photon number as a function of dephasing rate.

As demonstrated in the above analysis, the characteristics of resonant-tunneling transport can strongly influence the linewidth of THz QCLs in the incoherent resonant-tunneling transport regime, but only have small effect on the linewidth in the coherent resonant-tunneling transport regime. For the mid-IR QCLs, the injector barrier is usually designed to be thinner, and the electrons are injected into the upper laser level by coherent resonant-tunneling processes. We can deduce that the resonant-tunneling transport can exhibit negligible effect on the linewidth of mid-IR QCLs. Figures 4 - 7 and 4 – 8 show the effects of coupling strength and dephasing rate associated with resonant tunneling on the linewidth. Owing to the much larger coupling strength of mid-IR QCLs, the electron can be injected into the upper laser level by coherent resonant tunneling. As a result, further increasing the coupling strength and dephasing rate $\gamma_{41'}$ can only induce few changes of photon numbers, and hence only introduce little influence on the linewidth of mid-IR QCLs.

Fig. 4 - 7 The effects of coupling strength on the linewidth of mid-IR QCLs at resonance. (a) Linewidth as a function of coupling strength. The linewidth slowly decreases as the coupling strength of the injector level and the upper laser level increases. (b) Current density and photon number as a function of coupling strength.

Fig. 4 - 8 The effects of dephasing rate associated with resonant tunneling on the linewidth of mid-IR QCLs at resonance. (a) Linewidth as a function of dephasing rate. The linewidth slowly increases as the dephasing rate increases. (b) Current density and photon number as a function of dephasing rate.

In the rate equation model, the operation current is taken as an input parameter. However, many other parameters e.g. coupling strength, dephasing rate and doping level can influence the current. Therefore quantum design with the optimization of the active region of a QCL to reduce the intrinsic linewidth is limited by using rate equation model, but it can be easily made according to our quantum mechanical Langevin model. To take the effect of doping on the linewidth as an example, we can optimize the doping density to achieve a reduced linewidth. Increasing doping density results in an increase in free-carrier absorption and waveguide loss. According to Ref. [115], the loss can be assumed to be roughly proportional to the doping density. Doping not only affects the cavity loss (thus the intrinsic linewidth) but also the electron population distribution according to our model (we neglect the effects of doping density on relaxation rates in our chosen ranges of doping). For the rate equation model, doping affects the linewidth only by the cavity loss. Figure 4 - 9 (a) shows the self-induced linewidth variation by doping, the linewidth variation due to the change of doping induced cavity loss, and the overall effects from these two factors. The linewidth is enhanced as the doping density increases. Doping density does not cause the change of photon number but induce the increasing of current density in this calculation, as shown in Fig. 4 - 9(b). This shows that more electrons populate the upper laser level and take part in the spontaneous emission, then induce the linewidth broadening. The simulation tells us that we can optimize the doping density to reduce the linewidth of QCLs. It is noted that the optimization of the injector barrier is complex, since the thickness of barrier not only influences the coupling strength, but also it affects the lifetimes of the upper and lower laser level. Figure 4 - 10 shows the effects of lifetimes of the upper and laser levels on the linewidth. The linewidth increases and decreases as increasing the relaxation rate of upper laser and lower laser level, respectively. This is due to the variation of ratio of current to threshold current and photon number caused by the change of their relaxation rates, as shown in Fig. 4 - 11. Thus, a careful design of active region should be made to reduce the intrinsic linewidth of QCLs.

Fig. 4 - 9 Effect of doping density on the intrinsic linewidth of THz QCLs at resonance. (a) The self-induced linewidth variation by doping, the linewidth variation due to the change of doping induced cavity loss, and the overall effects from these two factors. (b) Current density and photon number as a function of doping density.

Fig. 4 - 10 (a) Linewidth as a function of relaxation rate of the upper laser level. (b) Linewidth as a function of the relaxation rate of the lower laser level

Fig. 4 - 11 (a) Ratio of current to threshold current and photon number as a function of relaxation rate of the upper laser level. (b) Ratio of current to threshold current and photon number as a function of relaxation rate of the lower laser level.

4.3 Thermal photon induced linewidth broadening

In addition to the spontaneous emission events, thermal photon may induce the linewidth broadening. The contribution of thermal photon to linewidth depends on the lasing wavelength and temperature. According to the study of Yamanishi [122, 136], the linewidth re-broadening induced by thermal photons is estimated to be tens of hertz at electron temperature of ~90 K. The thermal photon contributions are not only due to incoming fluctuation of thermal photon but also the noisy stimulated emission induced by thermal photon in the cavity. The complete thermal photon contribution to linewidth broadening is complicated [136], but here we just consider the simple case, namely we assume the system is in equilibrium. Then, the linewidth formula is modified as [122]

$$\delta f = \frac{\kappa n_{th} + 2(gg^*/\gamma_{34})(n_{4,0} + n_{th})}{4\pi(1 + \kappa/2\gamma_{34})^2 I_0} \quad (4.81)$$

Figure 4 - 12 shows the linewidth with and without the consideration of thermal photon. When THz QCLs operate at high temperature, the contribution of thermal photon to linewidth cannot be neglected.

Fig. 4 - 12 Effects of thermal photon on the linewidth broadening in THz QCLs at resonance.

4.4 Conclusions

In this chapter, we theoretically investigate the intrinsic linewidth source of both THz and mid-IR QCLs i.e. the spontaneous emission and thermal photon based on quantum Langevin equations. It includes the effects of resonant-tunneling transport and dynamics of coherent interaction on the intrinsic linewidth. The results show that the coupling strength and dephasing rate have significant effects on the linewidth of THz QCLs in the incoherent resonant-tunneling regime, but small effects on that of mid-IR QCLs due to their strong coherent resonant-tunneling. The linewidth decreases with the increase of the injection coupling strength and reduction of the dephasing rate associated with resonant tunneling. According to our model, a reduced intrinsic linewidth can be easily designed through optimization of the injector barrier and doping density of the active region of QCLs. In addition, the intrinsic linewidth caused by the thermal photon cannot be neglected for THz QCLs at high-temperature operation.

5 Fundamental thermal noise and linewidth broadening in quantum cascade lasers

5.1 Introduction

In addition to the contributions of spontaneous emission and thermal photon, the temperature fluctuations in thermodynamic equilibrium can induce the linewidth broadening. This fundamental thermal noise is caused by random temperature fluctuations within the laser cavity. It cannot be overcome due to the fundamental quantum limitations. Even for a cavity in the perfect thermal equilibrium with its surroundings, this thermal noise always exists. From our analysis, the linewidth broadening caused by fundamental thermal fluctuations for both THz and mid-IR QCLs are comparable to those caused by spontaneous emission and thermal photon. Therefore, it is important to investigate the effects of the fundamental thermal noise in both THz and mid-IR QCLs.

On the other hand, for many applications, it is the intrinsic frequency noise spectrum of QCLs that is of the primary interest. The studies of the intrinsic frequency noise of QCLs have been focused on high frequency at 10 MHz and even up to GHz [41]. For most spectroscopic applications [137-139], the detected signal is in the kHz range or below. Thus it is of great interest to investigate the thermal fluctuation frequency noise, which is indeed one of low-frequency noises.

In this chapter, the Green function methods and the Λ -theorem (*i.e.* the *Van Vliet–Fassett* form) are used to derive the analytical expressions of the frequency noise caused by temperature fluctuation for single-mode THz (in the double-metal waveguide structure) and mid-IR (in the buried heterostructure) QCLs. The effects of the temperature, heat conductivity and active region thickness on the fundamental frequency noise and linewidth broadening are theoretically investigated.

5.2 Analysis of THz QCLs

The quantum cascade lasers can be designed for light emissions covering the mid-IR and THz regimes. The waveguide structures of mid-IR and THz QCLs are different because they are operated in different wavelength regimes. Mid-IR QCLs are typically based on a dielectric waveguide structure in which the active region is embedded between semiconductor cladding layers with a smaller refractive index [5]. For THz QCLs, there are two types of waveguides

used, namely, the surface-plasmon waveguide [15, 42] and the double-metal waveguide [62]. The surface-plasmon waveguide sandwiches the active region with metal and a thin heavily doped semiconductor layer followed by a semi-insulating GaAs substrate. It has the advantages of high output powers and good beam patterns in terms of the beam divergence. The double-metal waveguide uses metal layers, which are placed just above and below the epitaxial active region. It has a perfect optical confinement with a confinement factor close to 1 and an efficient heat removal [37, 140-142], hence demonstrating the best temperature performance of THz QCLs. In this paper we focus on double-metal waveguides due to their high temperature performance, which is one of the most important research subjects in the field. We will firstly investigate the frequency noise and linewidth broadening in the THz QCLs. The results for mid-IR QCLs are shown in the next section. As for this section, the stochastic heat source is first evaluated, and then the thermal fluctuation spectrum and the resultant frequency noise and linewidth broadening are analytically derived.

5.2.1 Stochastic heat source

The statistical property of the random internal heat source driving the temperature fluctuation can be evaluated by fluctuations-dissipation theorem [143] from a Langevin diffusion equation, which has been used to investigate the thermal noise in solid-state devices [144-148]. This stochastic equation is written as

$$\frac{\partial \Delta T}{\partial t} - \frac{\eta}{\rho c} \nabla^2 \Delta T = \vec{\nabla} \cdot \mathbf{F}(\vec{r}, t), \quad (5.1)$$

where $\Delta T(\vec{r}, t) = T(\vec{r}, t) - \langle T(\vec{r}, t) \rangle$, $\langle \dots \rangle$ means time or ensemble averaging, $\mathbf{F}(\vec{r}, t)$ is the stochastic thermal-flux density of the internal heat sources of temperature fluctuation, η is thermal conductivity ($W\ cm^{-1}\ K^{-1}$), ρ is the density ($g\ cm^{-3}$), and c is the specific heat capacity ($J\ g^{-1}\ K^{-1}$).

According to the theory of thermodynamics [149, 150], the total variance of temperature fluctuation is described by the following well-known formula

$$\langle \Delta T^2 \rangle = \frac{k_B T_0^2}{C}, \quad (5.2)$$

where k_B is Boltzmann constant, $C = \rho c V$ is heat capacity ($J\ K^{-1}$), $V = Al$ is the volume, and T_0 is the equilibrium temperature.

The stochastic heat source function $F(\vec{r}, t)$ can be characterized by the following correlation function [144]

$$\langle F(\vec{r}, t) F(\vec{r}', t') \rangle = F_0^2 \delta^3(\vec{r} - \vec{r}') \delta(t - t'), \quad (5.3)$$

where F_0 is the constant and should be normalized in the following way to satisfy Eq. (5.2).

$$\langle \Delta T^2 \rangle = \frac{1}{V^2} \left\langle \int_V \Delta T(\vec{r}, t) d\vec{r} \right\rangle. \quad (5.4)$$

$\Delta T(\vec{r}, t)$ can be obtained by taking the Fourier transformation of Eq. (5.1)

$$\Delta T(\vec{r}, t) = \iint \frac{i\vec{k} \cdot \vec{F}(\vec{k}, \omega)}{i\omega + Dk^2} \exp(i\omega t + i\vec{k} \cdot \vec{r}) \frac{d\vec{k} d\omega}{(2\pi)^4}, \quad (5.5)$$

where $k^2 = \vec{k} \cdot \vec{k}$, $D = \eta / \rho c$, and ω is the angular frequency.

Hence, one can calculate $\langle \Delta T^2 \rangle$ according to Eq. (5.4)

$$\begin{aligned} \langle \Delta T^2 \rangle &= \frac{1}{V} \int_V d\vec{r} \frac{1}{V} \int_V d\vec{r}' \iiint \int_{-\infty}^{\infty} \frac{d\vec{k} d\vec{k}' d\omega d\omega'}{(2\pi)^8} \frac{k^2 \langle F(\vec{k}, \omega) F^*(\vec{k}', \omega') \rangle}{[Dk^2 + i\omega][Dk'^2 - i\omega']} e^{i\vec{k} \cdot \vec{r} - i\vec{k}' \cdot \vec{r}' + i(\omega - \omega')t} \\ &= \frac{1}{V} \int_V d\vec{r} \frac{1}{V} \int_V d\vec{r}' \iiint \int_{-\infty}^{\infty} \frac{d\vec{k} d\omega}{(2\pi)^4} \frac{k^2 F_0^2}{D^2 k^4 + \omega^2} e^{i\vec{k} \cdot (\vec{r} - \vec{r}')} = \frac{F_0^2}{2DV}, \end{aligned} \quad (5.6)$$

Comparing Eq. (5.2) with Eq. (5.6), we have

$$F_0^2 = 2DV k_B T_0^2 / C. \quad (5.7)$$

5.2.2 Equilibrium thermal fluctuation spectrum in THz QCLs

Figure 5 - 1 shows a schematic structure of the cross section of THz QCLs. Since THz QCLs are typically measured at a cryogenic temperature in a cryostat (less than 200 K, which is the maximum operating temperature reported) under vacuum operation, a constant-temperature boundary condition is applied to the bottom of the substrate, and neglected heat conduction is applied to the surfaces exposed to the vacuum [151]. One dimensional heat transport model is established due to the fast heat dissipation along the z -direction by the heat sink [152]. The top metal contact is not considered in the below discussion due to its very high heat conductivity compared to the active region in our pure thermal model. The stochastic heat-conduction equation has the following expression for

Fig. 5 - 1 The schematic diagram of the cross section of THz QCLs with a double-metal waveguide structure. A constant temperature boundary condition is applied to the bottom of the substrate, and neglected heat conduction is applied to the surfaces exposed to vacuum.

$$\frac{\partial \Delta T}{\partial t} - D(z) \frac{\partial^2 \Delta T}{\partial z^2} = \vec{\nabla}_z \cdot \vec{F}(\vec{z}, t), \quad (5.8)$$

where

$$\begin{aligned} D(z) &= D_1 = \eta_{\perp} / \rho c & 0 \leq z < w_1, \\ D(z) &= D_2 = \eta_m / \rho_m c_m & w_1 \leq z \leq w_2, \\ D(z) &= D_3 = \eta_s / \rho_s c_s & w_2 < z \leq w_3. \end{aligned}$$

The boundary conditions of the above equation are

$$\begin{aligned} \eta_{\perp} \frac{\partial \Delta T}{\partial z} \Big|_{z=0} &= 0, \quad \Delta T \Big|_{z=w_3} = 0, \\ \Delta T \Big|_{z=w_1-0} &= \Delta T \Big|_{z=w_1+0}, \quad \eta_{\perp} \frac{\partial \Delta T}{\partial z} \Big|_{z=w_1-0} = \eta_m \frac{\partial \Delta T}{\partial z} \Big|_{z=w_1+0}, \\ \Delta T \Big|_{z=w_2-0} &= \Delta T \Big|_{z=w_2+0}, \quad \eta_m \frac{\partial \Delta T}{\partial z} \Big|_{z=w_2-0} = \eta_s \frac{\partial \Delta T}{\partial z} \Big|_{z=w_2+0}, \end{aligned}$$

where η_m and η_s are the heat conductivity of the bottom metal and the GaAs substrate, respectively. As shown in Refs. [152] and [153], the heat conduction of the active region exhibits strong anisotropy in terms of in-plane and cross-plane heat flow. Here $\eta_{\perp}$ is the cross-plane heat conductivity of active region.

According to the derivation in section 5.2.1, the correlation function of the Langevin source satisfies

$$\langle F_i(\vec{z}, t) F_j(\vec{z}', t') \rangle = 2D\Delta w_j k_B T_0^2 / C_j \delta_{ij} \delta^3(z - z') \delta(t - t') \quad i, j = 1, 2, 3, \quad (5.9)$$

where

$$\Delta w_j = w_j - w_{j-1}.$$

The Fourier transformation of this Langevin equation gives the spectra information

$$i\omega \Delta T(z, \omega) - D(z) \frac{\partial^2}{\partial z^2} \Delta T(z, \omega) = \vec{\nabla}_z \cdot \vec{F}(\vec{z}, \omega). \quad (5.10)$$

Let G be the Green's function of the above equation, we have

$$i\omega G(z, z', i\omega) - D(z) \frac{\partial^2}{\partial z^2} G(z, z', i\omega) = \delta(z - z'), \quad (5.11)$$

with homogeneous boundary conditions

$$\begin{aligned} \eta_\perp \frac{\partial G}{\partial z} \Big|_{z=0} &= 0, \quad G \Big|_{z=w_3} = 0, \\ G \Big|_{z=w_1-0} &= G \Big|_{z=w_1+0}, \quad \eta_\perp \frac{\partial G}{\partial z} \Big|_{z=w_1-0} = \eta_m \frac{\partial G}{\partial z} \Big|_{z=w_1+0}, \\ G \Big|_{z=w_2-0} &= G \Big|_{z=w_2+0}, \quad \eta_m \frac{\partial G}{\partial z} \Big|_{z=w_2-0} = \eta_s \frac{\partial G}{\partial z} \Big|_{z=w_2+0}, \end{aligned}$$

For the self-adjoint operator, the relation of reciprocity satisfies [154]

$$G^*(z, z', -i\omega) = G(z', z, i\omega). \quad (5.12)$$

The following equation is obtained by multiplying Eq. (5.10) by G and Eq. (5.11) by ΔT and subtracting of the two

$$\begin{aligned} & \Delta T(z, \omega) - \int_w G(z', z, i\omega) \vec{\nabla}_{z'} \cdot \vec{F}(\vec{z}', \omega) dz' \\ &= \int_w [G(z', z, i\omega) D(z') \nabla_{z'}^2 \Delta T(z', \omega) - \Delta T(z', \omega) D(z') \nabla_{z'}^2 G(z', z, i\omega)] dz'. \end{aligned} \quad (5.13)$$

According to Green's theorem [155] and the homogeneous boundary conditions, it yields

$$\Delta T(z, \omega) = \int_w G(z, z', i\omega) \vec{\nabla}_{z'} \cdot \vec{F}(\vec{z}', \omega) dz'. \quad (5.14)$$

Then the correlation function can be deduced by

$$\langle \Delta T(z, \omega) \Delta T^*(z', \omega') \rangle = \iint G(z, z_1, i\omega) G^*(z', z_2, i\omega') S_F(z_1, z_2, \omega) dz_1 dz_2, \quad (5.15)$$

where

$$S_F(z_1, z_2, \omega) = \vec{\nabla}_{z_1} \cdot \vec{\nabla}_{z_2} \langle F(z_1, \omega) F^*(z_2, \omega') \rangle = 4\pi D \Delta w_j k_B T_0^2 / C_j \vec{\nabla}_{z_1} \cdot \vec{\nabla}_{z_2} \delta(z_1 - z_2) \delta(\omega - \omega').$$

Equation (5.15) is a quadratic Green's function. It can be transformed to be linear using the Λ -theorem, *i.e.* the *Van Vliet–Fassett* form [156, 157]. With the Λ -theorem, we have the following relation

$$\Lambda_z \Gamma(z, z') + \Lambda_{z'} \Gamma(z, z') = S_F(z, z', \omega) / 2, \quad (5.16)$$

where $\Lambda = -D(z) \nabla_z^2$.

This yields

$$\Gamma(z, z') = \pi \Delta w_j k_B T_0^2 \delta(z - z') \delta(\omega - \omega') / C_j. \quad (5.17)$$

According to Eq. (5.16), equation (5.15) can be written as

$$\langle \Delta T(z, \omega) \Delta T^*(z', \omega') \rangle = 2 \iint G(z, z_1, i\omega) G^*(z', z_2, i\omega') [\Lambda_{z_1} + \Lambda_{z_2}] \Gamma(z_1, z_2) dz_1 dz_2. \quad (5.18)$$

The right-hand side of Eq. (5.18) can be expressed as follows

$$\begin{aligned} & 2 \iint G(z, z_1, i\omega) G^*(z', z_2, i\omega') [(\Lambda_{z_1} + i\omega) + (\Lambda_{z_2} - i\omega)] \Gamma(z_1, z_2) dz_1 dz_2 \\ &= 2 \int dz_2 G^*(z', z_2, i\omega') \int G(z, z_1, i\omega) \ell_{z_1} \Gamma(z_1, z_2) dz_1 + \\ & \quad 2 \int dz_1 G(z, z_1, i\omega) \int G^*(z', z_2, i\omega') \ell_{z_2}^* \Gamma(z_1, z_2) dz_2 \\ &= 2 \int dz_2 G(z', z_2, -i\omega') \int \Gamma(z_1, z_2) \ell_{z_1} G(z, z_1, i\omega) dz_1 + \\ & \quad 2 \int dz_1 G(z, z_1, i\omega) \int \Gamma(z_1, z_2) \ell_{z_2}^* G(z', z_2, -i\omega') dz_2. \end{aligned} \quad (5.19)$$

where $\ell = \Lambda + i\omega$ and the following deduction is used

$$\begin{aligned} & \int_w dz [G(z, z', i\omega) \ell_z \Gamma(z, z') - \Gamma(z, z') \ell_z G(z, z', i\omega)] \\ &= \pi D \Delta w_j k_B T_0^2 \delta(\omega - \omega') / C_j \int_w dz [G(z, z', i\omega) \partial^2 \delta(z - z') / \partial z^2 - \delta(z - z') \partial^2 G(z, z', i\omega) / \partial z^2] \\ &= \pi D \Delta w_j k_B T_0^2 \delta(\omega - \omega') / C_j [\partial^2 G(z, z', i\omega) / \partial z^2 \Big|_{z=z'} - \partial^2 G(z, z', i\omega) / \partial z^2 \Big|_{z=z'} \Big|_{w_i}^{w_j}] = 0. \end{aligned} \quad (5.20)$$

Using the definition of Green's function, equation (5.18) can be simplified as

$$\begin{aligned} \langle \Delta T(z, \omega) \Delta T^*(z', \omega') \rangle &= 2 \int [G(z', z_1, -i\omega') \Gamma(z, z_1) + G(z', z_1, i\omega) \Gamma(z_1, z')] dz_1 \\ &= 4\pi \Delta w_j k_B T_0^2 \text{Re}[G(z, z', i\omega)] \delta(\omega - \omega') / C_j, \end{aligned}$$

(5.21)

where the notation $Re[\cdot]$ denotes real part.

It is noted that equation (5.16) is independent of the choice of the particular solution of the $\mathcal{A}$ -theorem [157].

The temperature fluctuations need to be averaged over the mode volume

$$\Delta\bar{T}(\vec{r}, t) = \int \Delta T(\vec{r}, t) |E(\vec{r})|^2 d\vec{r}, \quad (5.22)$$

where $|E(\vec{r})|^2$ is the normalized optical mode intensity distribution.

Therefore, the correlation function of $\Delta T(\vec{r}, t)$ averaged over the mode volume in the frequency domain can be written as

$$\langle \Delta\bar{T}(\omega) \Delta\bar{T}^*(\omega') \rangle = \iint_w \langle \Delta T(z, \omega) \Delta T^*(z', \omega') \rangle |E(z)|^2 |E(z')|^2 dz dz'. \quad (5.23)$$

From the Wiener-Khintchine relations, it is readily verified that the thermal noise spectrum (power spectral density) is merely the coefficient of the delta function divided by 2π . Hence the one-sided spectral density $S_{\Delta T}(\omega)$ of fluctuations of temperature reads

$$S_{\Delta T}(\omega) = 4\Delta w_j k_B T_0^2 / C_j \iint_w \text{Re}[G(z, z', i\omega)] |E(z)|^2 |E(z')|^2 dz dz'. \quad (5.24)$$

The following two additional conditions are required to determine Green's function at $z = z'$ by integrating Eq. (5.11) from $z = z' - 0$ to $z = z' + 0$

$$\left. \frac{\partial G(z, z', i\omega)}{\partial z} \right|_{z=z'-0}^{z=z'+0} = -\frac{1}{D(z)}, \quad (5.25)$$

$$G(z = z' - 0, z', i\omega) = G(z = z' + 0, z', i\omega). \quad (5.26)$$

The Green's function G can be achieved with standard methods [158, 159], we thus obtain $G_1^<$ ($0 \leq z < z'$), $G_1^>$ ($z' < z \leq w_1$), G_2 ($w_1 \leq z \leq w_2$), G_3 ($w_2 \leq z \leq w_3$) for the source point z' lay in the active region.

$$\begin{aligned} G_1^< &= c_1(z') \sinh(\alpha_1 z) + c_2(z') \cosh(\alpha_1 z), \\ G_1^> &= c_1(z') \sinh(\alpha_1 z) + c_2(z') \cosh(\alpha_1 z) - \sinh[\alpha_1(z - z')]/D_1 \alpha_1, \\ G_2 &= c_3(z') \sinh[\alpha_2(z - w_2)] + c_4(z') \cosh[\alpha_2(z - w_2)], \\ G_3 &= c_5(z') \sinh[\alpha_3(z - w_3)] + c_6(z') \cosh[\alpha_3(z - w_3)], \end{aligned} \quad (5.27)$$

where

$$\alpha_1 = \sqrt{i\omega/D_1}, \quad \alpha_2 = \sqrt{i\omega/D_2}, \quad \alpha_3 = \sqrt{i\omega/D_3}.$$

Coefficients $c_1(z'), \dots, c_6(z')$ can be determined from the boundary conditions. We show here the expressions of coefficients $c_1(z')$ and $c_2(z')$, which are of interests to us.

$$c_1(z') = 0, \quad (5.28)$$

$$c_2(z') = \frac{E_2 \eta_m \alpha_2 \sinh[\alpha_1(w_1 - z')] + E_1 \eta_{\perp} \alpha_1 \cosh[\alpha_1(w_1 - z')]}{[E_2 \eta_m \alpha_2 \cosh(\alpha_1 w_1) + E_1 \eta_{\perp} \alpha_1 \sinh(\alpha_1 w_1)] D_1 \alpha_1}, \quad (5.29)$$

where

$$E_1 = \eta_s \alpha_3 \cosh[\alpha_3(w_2 - w_3)] \sinh[\alpha_2(w_2 - w_1)] - \eta_m \alpha_2 \sinh[\alpha_3(w_2 - w_3)] \cosh[\alpha_2(w_2 - w_1)],$$

$$E_2 = \eta_s \alpha_3 \cosh[\alpha_3(w_2 - w_3)] \cosh[\alpha_2(w_2 - w_1)] - \eta_m \alpha_2 \sinh[\alpha_3(w_2 - w_3)] \sinh[\alpha_2(w_2 - w_1)].$$

In THz QCLs, the metal-metal waveguide has a perfect optical confinement [14] (confinement factor ~ 1) in the active region and its fundamental mode intensity is assumed to be approximately uniformly distributed across the active region. Thus, the spectrum of the averaged temperature fluctuations in the active region can be written as

$$\begin{aligned} S_{\Delta T}(\omega) &= 4w_1 k_B T_0^2 / (Cw_1^2) \text{Re}[\int_0^{w_1} dz' \int_0^{z'} dz G_1^<(z, z', i\omega) + \int_0^{w_1} dz' \int_{z'}^{w_1} dz G_1^>(z, z', i\omega)] \\ &= 4k_B T_0^2 / (\rho c A w_1^2) \text{Re}[\int_0^{w_1} \cosh(\alpha_1 z) dz \int_0^{w_1} c_2(z') dz' - \int_0^{w_1} dz' \int_{z'}^{w_1} \frac{\sinh[\alpha_1(z - z')]}{D_1 \alpha_1} dz] \\ &= 4k_B T_0^2 / (\rho c A w_1^2) \text{Re}[\frac{E_2 \eta_m \alpha_2 \sinh(\alpha_1 w_1) [\cosh(\alpha_1 w_1) - 1] + E_1 \eta_{\perp} \alpha_1 \sinh^2(\alpha_1 w_1)}{[E_2 \eta_m \alpha_2 \cosh(\alpha_1 w_1) + E_1 \eta_{\perp} \alpha_1 \sinh(\alpha_1 w_1)] D_1 \alpha_1^3} \\ &\quad - \frac{\sinh(\alpha_1 w_1) - \alpha_1 w_1}{D_1 \alpha_1^3}]. \end{aligned} \quad (5.30)$$

5.2.3 Frequency noise spectrum and linewidth broadening

If we only focus on the temperature dependence of the refractive index fluctuation, and ignore the thermal expansion and energy level broadening caused by the self heating, the instantaneous frequency shift caused by local thermal fluctuation is given by [145]

$$f(t) = -\nu_0 \int \frac{1}{n} \frac{dn}{dT} \Delta T(\vec{r}, t) |E(\vec{r})|^2 d\vec{r}, \quad (5.31)$$

where ν_0 is laser frequency, n is refractive index, dn/dT is the thermo-optic coefficient.

The correlation function of the frequency noise reads

$$\langle f(t)f(t') \rangle = (\nu_0 \frac{1}{n} \frac{dn}{dT})^2 \iint \langle \Delta T(\vec{r}, t) \Delta T(\vec{r}', t') \rangle |E(\vec{r})|^2 |E(\vec{r}')|^2 d\vec{r} d\vec{r}'. \quad (5.32)$$

With the Fourier transform of Eq. (5.32), the one-sided frequency noise spectrum caused by the temperature fluctuation yields

$$S_f = (\nu_0 \frac{1}{n} \frac{dn}{dT})^2 S_{\Delta T}(2\pi\nu). \quad (5.33)$$

Once we know the power spectrum density of the frequency noise, the linewidth then can be specified by the following derivation. The autocorrelation function of the lasing field $O(t)$ can be expressed for single-mode QCLs in the case where the phase change $\Delta\phi$ is considered as a Gaussian distribution [123].

$$\langle O(t)O^*(t-\tau) \rangle \sim \exp(i2\pi\nu_0\tau) \exp(-\langle \Delta\phi^2 \rangle / 2). \quad (5.34)$$

In general, if the spectral density of the frequency fluctuation is known, the mean square value (variance) of the phase change can be obtained as [57]

$$\langle \Delta\phi^2 \rangle = 8\pi \int_0^{+\infty} S_f(\omega) \frac{\sin^2(\omega\tau/2)}{\omega^2} d\omega, \quad (5.35)$$

where $\omega = 2\pi\nu$. The linewidth is then obtained from the one-sided power spectral density S_o of the autocorrelation function of $O(t)$, here S_o is given at $\Delta f = \nu_0 - \nu$ by taking the Fourier transform.

$$S_o(\Delta f) \sim \text{Re}[2 \int_{-\infty}^{+\infty} \exp(i2\pi\Delta f \tau) \exp(-\langle \Delta\phi^2 \rangle / 2) d\tau]. \quad (5.36)$$

5.2.4 Results and discussions

Throughout this section, the parameters and their values used for frequency noise calculations in THz QCLs are listed in Table 5 - 1. The active region is made of GaAs/Al_{0.15}Ga_{0.85}As semiconductor materials, and copper is used for metal waveguide. To simplify, we neglected the effects on heat conductivity changes caused by the active region thickness, the distribution and transport of hot injected electrons and their interactions with lattice. The refractive index change here is only related to current-induced device self-heating. The refractive index variations associated with the intersubband gain transitions are not included in the above derivations due to the lack of experimental and theoretical data. Estimations on the linewidth broadening caused by

the refractive index variations owing to the intersubband transitions in QCLs are given in section 5.3 of this chapter.

Table 5 - 1. Device parameters of THz QCLs used in numerical simulations (From Ref. [65, 111, 153, 160, 161]).

Symbol	Value	Symbol	Value
ρ	5.32 g cm^3	w_1	$10 \text{ }\mu\text{m}$
c	$0.18 \text{ J g}^{-1} \text{ K}^{-1}$	w_2	$11 \text{ }\mu\text{m}$
$\eta_{\perp}$	$9.6T^{-0.14} \text{ W m}^{-1} \text{ K}^{-1}$	w_3	$181 \text{ }\mu\text{m}$
ρ_m	8.96 g cm^{-3}	$1/n(dn/dT)$	$6.17 \times 10^{-5} \text{ K}^{-1}$
c_m	$0.385 \text{ J g}^{-1} \text{ K}^{-1}$	ν_0	3.9 THz
η_m	$398 \text{ W m}^{-1} \text{ K}^{-1}$	A	$2.51 \text{ mm} \times 170 \text{ }\mu\text{m}$

Figure 5 - 2 shows the temperature dependent frequency noise in the range of kHz and high frequency regime (1 MHz to 10 MHz), respectively. Throughout this chapter, the temperature refers to the lattice temperature of QCLs, where it is noted that the lattice temperature is normally 50-100 K higher than the heat-sink temperature [162]. The temperature ranging from 100 K to 250 K is of great interest, as most of the high performance THz QCLs are working in this range. It is noted that the highest operation temperature (heat-sink temperature) without external magnetic field control for THz QCLs is about 200 K [29]. As shown in Fig. 5 - 2, the frequency noise caused by the temperature fluctuation influences device performance mainly in the low frequency range (below a few kHz). The frequency noise drops fast close to zero as the frequency exceeds 1 MHz. We find that this frequency noise doesn't exhibit the $1/f$ characteristics. In terms of temperature performance, the frequency noise due to thermal fluctuations shows strong temperature dependence, which increases with the increase of the temperature.

Fig. 5 - 2 Frequency noise of THz QCLs caused by temperature fluctuation at different lattice temperatures from 100K to 250K. The frequency noise is more prominent in the low frequency range than that in the high frequency range. (a) In kHz range of less than 10 kHz, (b) In MHz range from 1 MHz to 10 MHz.

In order to investigate the linewidth broadening caused by fundamental thermal noise, we have evaluated $S_o(\Delta f)$ using a fast Fourier transform program, as shown in Fig. 5 - 3.

Fig. 5 - 3 Simulation of the linewidth broadening of THz QCLs caused by fundamental thermal noise from 100 K to 250 K. (a) The mean square value of the phase change vs. time at different temperatures. (b) The linewidth broadening as a function of temperature.

Figure 5 - 3(a) shows the temperature dependent mean square phase change $\langle \Delta \phi^2 \rangle$. If the sampling interval is long enough, the periodical oscillation of the mean square phase change can be observed. Figure 5 - 3(b) shows the linewidth broadening at different temperatures. The linewidth caused by thermal fluctuation can nearly be neglected in THz QCLs. However, this

linewidth broadening is comparable to the value of ~ 3 Hz caused by spontaneous emission and thermal photon predicted by Jirasek [121] in the high power regime in the THz QCLs.

Fig. 5 - 4 (a) Frequency noise as a function of the thickness of the active region at 200 K. A critical frequency of 5 kHz is found. When the frequency is less than 5 kHz, the frequency noise decreases as the thickness of the active region increases, while if the frequency is above 5 kHz, the thicker the active region, the lower the frequency noise. (b) Frequency noise as a function of the thickness of the substrate at 200 K. A critical frequency of ~ 1 kHz is found. The insets of the figures are partial enlarged views, respectively.

The thickness of the active region and the substrate can also influence the laser frequency noise. The frequency-noise responses to the thicknesses of various layers are different in different frequency ranges. It needs to be mentioned that the effects of the active region thickness on the heat conductivity are neglected. As shown in Fig. 5 - 4, when the frequency is less than 5 kHz, the frequency noise increases as the thickness of the active region increases. While if the frequency exceeds 5 kHz, the thicker active region, the lower frequency the noise. These different noise characteristics below and above the critical frequency 5 kHz can be ascribed to the noise correlation length [144] which is frequency dependent. As shown in Ref. [144], the correlation length of thermal fluctuation is inverse proportional to the frequency. At the low frequencies, the fluctuations become correlated across the entire active region. Therefore, a thicker active region can induce a larger fluctuation amplitude at the low-frequency region. For the substrate, this critical frequency is about 1 kHz. The frequency noise response below and above this critical frequency in the substrate is similar to those in the active region. However, when frequency is more than 4 kHz, the thickness of the substrate nearly cannot influence the temperature fluctuation. Hence, the frequency noise of THz QCL is mainly affected by the thickness of the active region in the high frequency range (> 5 kHz).

Fig. 5 - 5 Comparison of the frequency noise caused by thermal fluctuations when gold and copper are used as metal cladding layers at 200 K, respectively. Both gold and copper give similar performance to the frequency noise.

Although copper has a better waveguide performance than gold in terms of thermal conduction, the noise properties for these two waveguides are nearly the same, as shown in Fig. 5 - 5. For gold, the following parameters are used $\rho_m=19.3 \text{ g cm}^{-3}$, $c_m=0.128 \text{ J g}^{-1} \text{ K}^{-1}$, $\eta_m=317 \text{ W m}^{-1} \text{ K}^{-1}$ (Ref. [161]).

5.3 Analysis of mid-IR QCLs

5.3.1 Fundamental thermal noise

Fig. 5 - 6 Buried heterostructure of a mid-IRQCL mounted epilayer down on a diamond heat sink.

Various device geometries have been used to improve the heat dissipation from the active region of mid-IR QCLs [151, 163]. In this paper, we use the ridge waveguide structure with a buried heterostructure mounted epilayer down on a diamond heat sink [16, 137] (see Fig. 5 - 6),

which demonstrated a very good heat dissipation effect. In this structure, most of the heat is transferred effectively via the diamond heat sink. Thus a constant temperature condition is applied to the heat sink considering the high heat conductivity of diamond (about $2600 \text{ W m}^{-1} \text{ K}^{-1}$ at 300 K [163]). On the top surface of the device, the heat is partially transferred to the air by convection. For this buried heterostructure, the heat dissipation can also be partially removed by the lateral heat flow. Since the heat conduction of the active region exhibits a strong anisotropy in the in-plane and cross-plane directions, the in-plane thermal noise is much smaller compared with the cross-plane thermal noise if the thermal fluctuations are independently considered (without considering the correlation/coupling of their thermal fluctuations) owing to much larger in-plane heat conductivity. This conclusion is based on the fact that the thermal frequency noise caused by temperature fluctuations is inversely proportional to the heat conductivity, as shown in Fig. 5 - 9 (a similar conclusion is also shown in Ref. [148]). Hence, the thermal fluctuations in the x-, y-directions are neglected. By considering the heat flow in the z-direction, we have

$$\frac{\partial \Delta T}{\partial t} - D(z) \frac{\partial^2 \Delta T}{\partial z^2} = \vec{\nabla} \cdot \mathbf{F}(\vec{z}, t), \quad (5.37)$$

where

$$\begin{aligned} D(z) &= D_1 = \eta_{\text{inp}} / \rho_{\text{inp}} c_{\text{inp}} & 0 \leq z < w_1 \quad \text{and} \quad w_2 < z \leq w_3, \\ D(z) &= D_2 = \eta_{\perp} / \rho c & w_1 \leq z \leq w_2, \end{aligned}$$

and the boundary conditions are:

$$\begin{aligned} \eta_{\perp} \frac{\partial \Delta T}{\partial z} \Big|_{z=w_3} + h \Delta T \Big|_{z=w_3} &= 0, \quad \Delta T \Big|_{z=0} = 0, \\ \Delta T \Big|_{z=w_1-0} &= \Delta T \Big|_{z=w_1+0}, \quad \eta_{\perp} \frac{\partial \Delta T}{\partial z} \Big|_{z=w_1+0} = \eta_{\text{inp}} \frac{\partial \Delta T}{\partial z} \Big|_{z=w_1-0}, \\ \Delta T \Big|_{z=w_2-0} &= \Delta T \Big|_{z=w_2+0}, \quad \eta_{\perp} \frac{\partial \Delta T}{\partial z} \Big|_{z=w_2-0} = \eta_{\text{inp}} \frac{\partial \Delta T}{\partial z} \Big|_{z=w_2+0}, \end{aligned}$$

where η_{inp} is the heat conductivity of the InP cladding and substrate, $\eta_{\perp}$ is the cross-plan heat conductivity of active region.

The Green's function of Eq.(5.37) is

$$i\omega G(z, z', i\omega) - D(z) \frac{\partial^2}{\partial z^2} G(z, z', i\omega) = \delta(z - z'), \quad (5.38)$$

with the boundary conditions

$$\begin{aligned} \eta_{inp} \frac{\partial G}{\partial z} \Big|_{z=w_3} + h G \Big|_{z=w_3} &= 0, \quad G \Big|_{z=0} = 0, \\ G \Big|_{z=w_1-0} &= G \Big|_{z=w_1+0}, \quad \eta_{\perp} \frac{\partial G}{\partial z} \Big|_{z=w_1+0} = \eta_{inp} \frac{\partial G}{\partial z} \Big|_{z=w_1-0}, \\ G \Big|_{z=w_2-0} &= G \Big|_{z=w_2+0}, \quad \eta_{\perp} \frac{\partial G}{\partial z} \Big|_{z=w_2+0} = \eta_{inp} \frac{\partial G}{\partial z} \Big|_{z=w_2-0}, \end{aligned}$$

Furthermore, the continuity of G and the jumping condition on $\partial G/\partial z$ yield, respectively

$$G(z = z' - 0, z', i\omega) = G(z = z' + 0, z', i\omega), \quad (5.39)$$

$$\frac{\partial G(z, z', i\omega)}{\partial z} \Big|_{z=z'-0}^{z=z'+0} = -\frac{1}{D(z)}. \quad (5.40)$$

With the Green's theorem and the Λ -theorem, the correlation function of $\Delta T(\vec{r}, t)$ in the frequency domain yields:

$$\langle \Delta T(z, \omega) \Delta T^*(z', \omega') \rangle = 4\pi \Delta w_j k_B T_0^2 \text{Re}[G(z, z', i\omega)] \delta(\omega - \omega') / C_j. \quad (5.41)$$

The Green's functions $G_1 (0 \leq z \leq w_1)$, $G_2^< (w_1 \leq z < z')$, $G_2^> (z' < z \leq w_2)$, $G_3 (w_2 \leq z \leq w_3)$ for the source point z' lay in active region are obtained

$$\begin{aligned} G_1 &= c_1(z') \sinh(\alpha_1 z) + c_2(z') \cosh(\alpha_1 z), \\ G_2^< &= c_3(z') \sinh[\alpha_2(z - w_2)] + c_4(z') \cosh[\alpha_2(z - w_2)] + \sinh[\alpha_2(z - z')] / D_2 \alpha_2, \\ G_2^> &= c_3(z') \sinh[\alpha_2(z - w_2)] + c_4(z') \cosh[\alpha_2(z - w_2)], \\ G_3 &= c_5(z') \sinh[\alpha_1(z - w_3)] + c_6(z') \cosh[\alpha_1(z - w_3)], \end{aligned} \quad (5.42)$$

where

$$\alpha_1 = \sqrt{i\omega/D_1}, \quad \alpha_2 = \sqrt{i\omega/D_2}.$$

Coefficients $c_1(z'), \dots, c_6(z')$ can be determined from the boundary conditions. The expressions of coefficients $c_3(z')$ and $c_4(z')$ of interest to us read

$$c_3(z') = \frac{E_2 \eta_{inp} \alpha_1 \{ \eta_{\perp} \alpha_2 \sinh(\alpha_1 w_1) \cosh[\alpha_2(w_1 - z')] - \alpha_1 \eta_{inp} \cosh(\alpha_1 w_1) \sinh[\alpha_2(w_1 - z')] \}}{[E_3 E_2 \eta_{inp} \alpha_1 + E_4 E_1 \alpha_2 \eta_{\perp}] D_2 \alpha_2}, \quad (5.43)$$

$$c_4(z') = \frac{E_1 \eta_{\perp} \{ \eta_{\perp} \alpha_2 \sinh(\alpha_1 w_1) \cosh[\alpha_2 (w_1 - z')] - \alpha_1 \eta_{imp} \cosh(\alpha_1 w_1) \sinh[\alpha_2 (w_1 - z')] \}}{[E_3 E_2 \eta_{imp} \alpha_1 + E_4 E_1 \alpha_2 \eta_{\perp}] D_2}, \quad (5.44)$$

where

$$\begin{aligned} E_1 &= -h \sinh[\alpha_1 (w_2 - w_3)] + \eta_{imp} \alpha_1 \cosh[\alpha_1 (w_2 - w_3)], \\ E_2 &= -h \cosh[\alpha_1 (w_2 - w_3)] + \eta_{imp} \alpha_1 \sinh[\alpha_1 (w_2 - w_3)], \\ E_3 &= \alpha_1 \eta_{imp} \cosh(\alpha_1 w_1) \sinh[\alpha_2 (w_1 - w_2)] - \alpha_2 \eta_{\perp} \sinh(\alpha_1 w_1) \cosh[\alpha_2 (w_1 - w_2)], \\ E_4 &= \alpha_1 \eta_{imp} \cosh(\alpha_1 w_1) \cosh[\alpha_2 (w_1 - w_2)] - \alpha_2 \eta_{\perp} \sinh(\alpha_1 w_1) \sinh[\alpha_2 (w_1 - w_2)]. \end{aligned}$$

The normalized fundamental mode intensity is assumed to be Gaussian [164]

$$|E(z)|^2 = \exp\{-[z - (w_1 + w_2)/2]^2 / m_0^2\} / \sqrt{\pi} m_0, \quad (5.45)$$

where m_0 is the mode field radius.

Therefore, for the spectrum of the averaged temperature fluctuations, we find

$$\begin{aligned} S_{\Delta T} &= 4k_B T_0^2 / (\rho c A) \operatorname{Re} \left[\int_{w_1}^{w_2} dz' \int_{w_1}^{z'} dz G_2^<(z, z', i\omega) |E(z)|^2 |E(z')|^2 \right. \\ &\quad \left. + \int_{w_1}^{w_2} dz' \int_{z'}^{w_2} dz G_2^>(z, z', i\omega) |E(z)|^2 |E(z')|^2 \right] \\ &= 4k_B T_0^2 / (\rho c A) \operatorname{Re} \left[\int_{w_1}^{w_2} \sinh[\alpha_2 (z - w_2)] |E(z)|^2 dz \int_{w_1}^{w_2} c_3(z') |E(z')|^2 dz' \right. \\ &\quad \left. + \int_{w_1}^{w_2} \cosh[\alpha_2 (z - w_2)] |E(z)|^2 dz \int_{w_1}^{w_2} c_4(z') |E(z')|^2 dz' \right. \\ &\quad \left. + \int_{w_1}^{w_2} dz' \int_{w_1}^{z'} dz \sinh[\alpha_2 (z - z')] / D_2 \alpha_2 |E(z)|^2 |E(z')|^2 \right]. \end{aligned} \quad (5.46)$$

For mid-IR QCLs, the expressions of frequency noise and linewidth broadening caused by the temperature fluctuation are the same as Eq. (5.33) and Eq. (5.36), respectively. The main difference of the thermal noise between mid-IR and THz QCLs is in $S_{\Delta T}$.

5.3.2 Results and discussions

For mid-IR QCLs, material system of $\text{In}_{0.29}\text{Al}_{0.71}\text{As}/\text{In}_{0.73}\text{Ga}_{0.27}\text{As}/\text{InP}$ is considered. The parameters used for frequency noise calculations are listed in Table 5 - 2. As indicated in Ref. [163], we also assume that the cross-plane heat conductivity of the active region is independent of temperature. Similar to the analysis in THz QCLs, the effects of active region thickness, the distribution and transport of hot injected electrons and their interactions with lattice on the heat conductivity are neglected. The refractive index change discussed here is related to current-

induced device self-heating; the refractive index variations associated with the intersubband gain transition are neglected due to the lack of experimental and theoretical data in the literature.

Table 5 - 2 Device parameters of mid-IR QCLs used in numerical simulations. (From Ref. [163, 165, 166])

Symbol	Value	Symbol	Value
ρ	5.55 g cm^{-3}	w_1	$2 \text{ }\mu\text{m}$
c	$0.30 \text{ J g}^{-1} \text{ K}^{-1}$	w_2	$3.57 \text{ }\mu\text{m}$
$\eta_{\perp}$	$0.76 \text{ W m}^{-1} \text{ K}^{-1}$	w_3	$5.57 \text{ }\mu\text{m}$
ρ_{inp}	4.81 g cm^{-3}	λ	$3.8 \text{ }\mu\text{m}$
c_{inp}	$0.18 \text{ J g}^{-1} \text{ K}^{-1}$	A	$4 \text{ mm} \times 11.5 \text{ }\mu\text{m}$
η_{inp}	$282000T^{-1.45} \text{ W m}^{-1} \text{ K}^{-1}$	h	$20 \text{ W m}^{-2} \text{ K}^{-1}$
dn/dT	$0.45 \times 10^{-4} \text{ K}^{-1}$	m_0	$(w_2 - w_1)/2$
n	3.27

Fig. 5 - 7 Frequency noise of mid-IR QCLs as a function of temperature from 200K to 400K. (a) In the frequency range less than 100 kHz, (b) In the frequency range from 100 kHz to 1MHz

As shown in Fig. 5 - 7, the same characteristics of temperature dependent laser frequency noise for mid-IR QCLs are observed as for THz QCLs. The frequency noises can be almost neglected when the frequency exceeds 400 kHz. It is noted that the frequency noise does not show a $1/f$ trend in the whole frequency region. This characteristic is not reflected in the

experimental measurements using an ultralow-technical-noise laser current source, where a $1/f$ frequency noise exists in mid-IRQCLs at the low frequency region from 10 Hz to 100 kHz [106, 108, 109, 120]. The origin of $1/f$ noise in low frequency range is not clear in QCLs at the moment. In Ref. [106], due to the uses of an ultralow-technical-noise laser current source, the parasitic noise from the characterization equipments is minimized. In addition, the spontaneous emission noise and carrier shot noise have a white noise spectrum. Thus, the origin of $1/f$ noise characteristic could probably come from the thermal noise owing to the non-Langevin heat source associated with the emissive and absorptive processes of photons and carriers, *e.g.* random spontaneous emission events as shown in fiber laser [146]. It could also come from the noise due to the optical feedback in cold cavities or from the non-Markovian processes or due to the contacts, lattice scattering [167], carriers trapped by material defects [168] or from fluctuations of electron tunneling through the multi-barriers QCL structure [120]. Further investigations are required to understand this phenomenon.

Fig. 5 - 8 (a) The mean square value of the phase change vs. time at different temperatures. (b) Normalized power spectral density $S_o(\Delta f)$ vs. frequency at different temperatures. (c) The linewidth broadening as a function of temperature.

Figure 5 - 8(a) shows the temperature dependent mean square phase change $\langle \Delta \phi^2 \rangle$. Through curve fitting, we find the variance $\langle \Delta \phi^2 \rangle \propto a + b\tau^{1.11}$ (a, b are constants). Hence, the resultant lineshape of mid-IR QCLs is neither Lorentzian nor Gaussian shape (see Fig. 5 - 8(b)) in contrast to that of diode lasers [169]. The main reason is due to the geometric structure studied in this paper, where a three-layer structure is investigated for mid-IR QCLs. Figure 5 - 8(c) shows the linewidth broadening at different temperatures considering only the temperature dependence of the refractive index fluctuations caused by current-induced device self-heating. The linewidth increases from 14.74 Hz to 62.02 Hz as the temperature increases from 200 K to 400 K.

In the above analysis, we only consider the macroscopic physics of the linewidth broadening. The microscopic physics of the gain medium of QCLs is ignored. We note that the refractive index can be affected by not only the current-induced device self-heating but also the laser transitions near the lasing wavelength, which can have a strong temperature dependence associated with electron populations and scattering rates, as discussed in Ref. [116]. The thermal expansion and energy level broadening caused by current-induced device self-heating can also cause a significant linewidth broadening. If all of these factors are considered, the frequency noise spectrum caused by the temperature fluctuation becomes

$$S_f = [\nu_0 (\frac{1}{n} \frac{dn}{dT} + \frac{1}{n} \frac{dn_e}{dT} + \frac{1}{l} \frac{dl}{dT} + \frac{2\pi}{\hbar \nu_0} \frac{d\Delta \epsilon}{dT})]^2 S_{\Delta T} (2\pi \nu). \quad (5.47)$$

The second term (on the right side of the equation above) stands for the refractive index change due to the laser transition near the lasing wavelength; the third term is due to the thermal expansion at various temperatures; and the fourth is owing to the temperature-dependent energy level fluctuation of the lasing emission. From the measurement results in Ref. [116], we can deduce that the third term has the same order of amplitude with the first term, and the second term is negligible. The fourth term is also believed to have an essential effect on the linewidth broadening of QCLs as is the case in quantum dots lasers [170], while the value of this parameter was not reported in QCLs in the literature, to be best of our knowledge. Therefore, even we assume both the second and the fourth items are negligible, we can conclude that at least the first

and the third items are comparable. In this case, the linewidth broadening is at least four times larger than previous calculations, which achieves from 64.3 Hz to 266.4 Hz as the temperature of 400 K. However, it needs to be mentioned that the exact values of these three parameters are not available in the literature. Exact calculation of the linewidth broadening needs further theoretical and experimental investigations.

Fig. 5 - 9 (a) Frequency noise as a function of the cross-plan conductivity of active region at 300 K. (b) Linewidth broadening at different the cross-plan conductivity of active region at 300 K.

The fundamental thermal frequency noise is strongly dependent on the heat conductivity of active region, as shown in Fig. 5 - 9(a). A higher heat conductivity of the active region induces a smaller thermal fluctuation, hence reducing the linewidth broadening (see Fig. 5 - 9(b)).

Figure 5 - 10 shows the influence of the thicknesses of the active region and the metal cladding on the laser frequency noise caused by temperature fluctuations. Unlike the case in THz QCLs, the active region can greatly influence the temperature fluctuation in mid-IR QCLs. The frequency noise first increases with the increase of the thickness of the active region, then decreases. The effects of the thicknesses of the InP cladding layers are more complicated. Decreasing the thickness of the upper cladding layer can reduce the frequency noise, while decreasing the thickness of the lower cladding layer can increase the temperature fluctuations. Therefore the overall effect is that reducing simultaneously the thicknesses of both cladding layers can reduce the frequency noise at the beginning, but increase it at around 5 kHz, as shown in Fig. 5 - 10(d). With the optimized cladding layers, the temperature fluctuation can be effectively minimized.

Fig. 5 - 10 Frequency noise as a function of the thickness of mid-IR QCL structure at 300 K. (a) Active region. (b) Upper-layer InP cladding. (c) Lower-layer InP cladding. (d) The thickness of the two-layer InP cladding changes simultaneously.

The effects of the lower- and upper- cladding layers on the linewidth broadening can be directly predicted from Fig. 5 - 10(b) and (c). The frequency noise, as well as the linewidth broadening, decreases with the decrease of the thickness of the upper cladding layer and the increase of the thickness of the lower cladding layer. In addition, influence of the thickness variation of the active region on the linewidth broadening is complicated, and should be numerically calculated. As shown in Fig. 5 - 11, the thinner the active region the smaller the linewidth broadening.

Fig. 5 - 11 The linewidth broadening as a function of thickness of active region at 300 K.

We also study the effects of optical mode intensity distribution on the frequency noise, as shown in Fig. 5 - 12. The calculations indicate that the frequency noise strongly increases with the decrease of the mode field radius, hence the increasing linewidth broadening. Therefore, by optimizing the geometry of the waveguide structure of mid-IR QCLs, one can change the frequency noise effectively.

Fig. 5 - 12 Frequency noise of mid-IR QCLs as a function of the optical mode field radius at 300 K.

5.4 Experimental approach

In this section, we provide a simple demonstration of the experimental approach to measure the intrinsic linewidth of QCLs. Because QCLs have a very narrow intrinsic linewidth, the accuracy of intrinsic linewidth measurement is limited by many factors e.g. the driver current noise and intensity noise contribution. The first accurate measurement of intrinsic linewidth in QCLs are undertaken by Bartalini *et al* [106], where a driver with ultra-low current noise is used.

The intrinsic linewidth can be derived from the frequency noise spectrum. In the experiment, the frequency noise of the QCL cannot be directly measured. It is extracted from the measurement of the intensity fluctuation converted by a frequency-to-amplitude converter [171]. Figure 5 - 13 shows the schematic of the experimental setup for the linewidth measurement in THz QCLs [105]. A single-mode distributed feedback QCL is used. The laser beam is first collimated and then sent to the absorption cell for spectroscopy experiments. This cell is served as frequency-to-amplitude converter and based on the side of a Doppler-broadened methanol molecular transition [105]. The converter function of the absorption molecular line is shown in the bottom plot. After the absorption cell, the beam is split by a polarizer. The reflected beam is chopped and sent to the detector. The lineshape is acquired and sent to feedback loop for frequency stabilization. During the frequency noise measurement, the QCL frequency should be stabilized at the centre of the discriminator slope. This can make sure the maximum conversion factor constant. Finally, the transmitted beam is acquired for frequency noise measurements. In combination with fast detection and low-noise fast-Fourier transformation acquisition, the wide frequency-spectrum measurements from 10 Hz to 100 MHz can be achieved. After we get a wide-band frequency noise spectrum, the intrinsic linewidth can be derived from the white component of the frequency spectrum. The laser intrinsic linewidth can be also read from the emission spectrum of the laser, which is directly measured by the beat-note experiments. The measurement setup is complicated, and more details can be found in Ref. [172]

Fig. 5 - 13 Schematic of the experimental setup of frequency noise and linewidth for THz QCLs. The figure is reproduced from Ref. [105].

5.5 Conclusions

A theoretical model based on Green function analysis and *Van Vliet-Fassett* theory has been presented for the analysis of the fundamental frequency noise and linewidth broadening of QCLs caused by intrinsic thermal fluctuations. It is found that the intrinsic frequency noise of QCLs due to thermal fluctuation is a low frequency noise, which shows strong temperature dependence in both THz and mid-infrared QCLs. For both THz and mid-infrared QCLs, frequency noise does not exhibit a $1/f$ trend in the whole frequency region. This characteristic is not reflected in the experimental measurements, which shows a $1/f$ frequency noise in mid-infrared QCLs at low frequency region of 10 Hz-100 kHz. The origin of $1/f$ noise characteristic could probably come from the thermal noise owing to the non-Langevin heat source associated with the emissive and absorptive processes of photons and carriers. It could also come from the noise due to the optical feedback in cold cavities or from the non-Markovian processes or due to the contacts, lattice

scattering, carriers trapped by material defects or from fluctuations of electron tunneling through the multi-barriers QCL structure. Further investigations are required to understand this phenomenon.

The linewidth broadening due to the temperature dependent refractive index variations caused by current-induced device self-heating can nearly be neglected in THz QCLs. For mid-infrared QCLs, the linewidth increases from 14.74 Hz to 62.02 Hz as the temperature changes from 200 K to 400 K. The resulting lineshape is neither Lorentzian nor Gaussian shape. We also find that if the refractive index change associated with the intersubband gain transition, the thermal expansion and the energy level broadening caused by self-heating are considered, the linewidth broadening should increase at least from 64.3 Hz to 266.4 Hz at 400 K. These microscopic effects should be further determined theoretically and experimentally. The effects of heat and structure parameters, such as heat conductivity of the active region, the thickness of the active region/substrate and the optical mode intensity distribution, on the frequency noise and the linewidth broadening of mid-infrared QCLs have also been investigated. The results show that the fundamental frequency noise and the intrinsic linewidth broadening decrease when the heat conductivity, the thickness of the lower cladding layer and the mode field radius increase. They also increase with the decrease of the thicknesses of the active region and the upper cladding layer. Through the optimization of these heat and structure parameters, the effects on the temperature fluctuations can be effectively reduced.

6 Linewidth enhancement factor

6.1 Introduction

Linewidth enhancement factor (LEF, α) 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 [53]. Owing to this coupling, a variation of the intensity of optical field induces an excess perturbation of the phase of a laser mode, which influences the performance of lasers e.g. the frequency responses of a laser [173]. For many practical applications of a laser system, a small LEF is highly desirable. A large LEF will cause the laser linewidth increases well beyond the Schawlow-Townes limit, making the devices more sensitive to the external optical feedback and induce frequency chirping effect. In addition, LEF reflects the properties of the gain spectrum. Thus, systematic studies of dependence of LEF on device parameters are needed.

Typical value of LEF in interband semiconductor diode lasers is about 2-7 [55], 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* [10]. However, the currently reported experiments [103, 107, 116, 174-176] 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 $8.22 \mu\text{m}$ [103] and $\sim 116 \mu\text{m}$ ($\sim 2.55 \text{ THz}$) [174], respectively. In addition, a strong dependence of the LEF on frequency detuning is observed, and a large value of up to -2 is reported for distributed feedback (DFB) mid-IR QCLs [176].

Although several experimental works have been carried out in studying the LEF of QCLs, few theoretical investigations on LEF has been reported [116] 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 [116]. 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 characteristics 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.

In chapter 3, we have developed a microscopic density matrix model to examine the optical gain of THz QCLs. It is demonstrated that the many body effects, which lead to renormalization of band structure and Rabi frequency, have significant modifications to the gain profile. In this chapter, 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 α -spectrum is blue-shifted as the injection current increases, which may explain the recent experimental result of α -value increases as 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.

6.2 Amplitude-phase coupling and linewidth broadening

Spontaneous emission can not only induce the phase change of optical field but also cause the instantaneous variation of field intensity. Due to the deviation of field intensity from the steady-state value, the laser will undergo oscillations. During this period, there will be a net gain change $\Delta G = (-\omega/c)\Delta n''$, where $\Delta n''$ is the change of the imaginary part of the refractive index around its steady-state value caused by the variation of carrier density. The change in imaginary part of the refractive index will alter its real part $\Delta n'$ [53]. The ratio of changes in real and imaginary parts of the refractive index is called the linewidth enhancement factor (or amplitude-phase coupling factor)

$$\alpha = \frac{\Delta n'}{\Delta n''} \quad (6.1)$$

As a result, $\Delta n'$ will induce an additional phase shift $\Delta\phi_p''$ of the laser field. According to the laser theory, we have

$$\begin{cases} \frac{d\phi}{dt} = \frac{\alpha}{2}(G - \kappa) \\ \frac{dI}{dt} = (G - \kappa)I \end{cases} \quad (6.2)$$

Then we can obtain the phase change $\Delta\phi_p''$

$$\Delta\phi_p'' = -\frac{\alpha}{2I}\Delta I \quad (6.3)$$

According to Fig. 4-1, the intensity change can be expressed as

$$\Delta I = 1 + 2\sqrt{I} \cos(\theta_p) \quad (6.4)$$

Then the total phase change due to the amplitude-phase coupling is

$$\Delta\phi_p'' = -\sum_{p=1}^{p=R_{sp}} \frac{\alpha}{2I} \left[1 + 2\sqrt{I} \cos(\theta_p) \right] \quad (6.5)$$

The first term is small and is neglected. We can obtain the average square phase change

$$\left\langle (\Delta\phi_p'')^2 \right\rangle = \frac{\alpha^2 R_{sp}}{2I} \quad (6.6)$$

So the linewidth broadening due to the amplitude-phase coupling is

$$\delta f'' = \frac{\alpha^2 R_{sp}}{4\pi I} \quad (6.7)$$

Therefore, when the amplitude-phase coupling is considered, the linewidth will be broadened by $1 + \alpha^2$.

6.3 Linewidth enhancement factor in THz QCLs

For THz QCLs, we consider the resonant-phonon (RP) design, as shown in Fig. 3-1. The Hamiltonian of this system is rewritten as

$$\begin{aligned} H = & -\sum_{\mathbf{k}} (\mu E b_{4,\mathbf{k}}^\dagger b_{3,\mathbf{k}} + c.c.) - \sum_{\mathbf{k}} \left[(\Delta_{41'}/2) b_{1',\mathbf{k}}^\dagger b_{4,\mathbf{k}} + c.c. \right] - \sum_{\mathbf{k}} \left[(\Delta_{23}/2) b_{3,\mathbf{k}}^\dagger b_{2,\mathbf{k}} + c.c. \right] \\ & - \sum_{\mathbf{k}} \left[(\Delta_{31'}/2) b_{1',\mathbf{k}}^\dagger b_{3,\mathbf{k}} + c.c. \right] + \sum_{j=1,2,3,4} \sum_{\mathbf{k}} \varepsilon_{j,\mathbf{k}} b_{j,\mathbf{k}}^\dagger b_{j,\mathbf{k}} + \frac{1}{2} \sum_{uvv'u'} \sum_{\mathbf{k}\mathbf{k}'\mathbf{q}} V_{\mathbf{q}}^{uvv'u'} b_{u,\mathbf{k}+\mathbf{q}}^\dagger b_{v,\mathbf{k}'-\mathbf{q}}^\dagger b_{v',\mathbf{k}} b_{u',\mathbf{k}} \end{aligned} \quad (6.8)$$

where we consider the many-body Coulomb interaction, nonparabolicity and resonant tunneling effects.

According to chapter 3, the optical gain and the refractive index change are written as

$$G = -\frac{2\omega_\lambda}{\varepsilon_0 n c V_m \xi} \text{Im} \left(\sum_{\mathbf{k}} \mu_{34} \tilde{P}_{34,\mathbf{k}} \right) \quad (6.9)$$

$$\delta n = \frac{\mu}{\varepsilon_0 n V_m \xi} \text{Re} \left(\sum_{\mathbf{k}} \mu_{34} \tilde{P}_{34,\mathbf{k}} \right) \quad (6.10)$$

The linewidth enhancement factor α can be obtained by the ratio of the change in the real part of the refractive index change δn to the change in the gain G with respect to the carrier density N_0 [87]

$$\alpha = \frac{\Delta n'}{\Delta n''} = -2 \frac{\omega_\lambda}{c} \frac{d(\delta n)}{dN_0} \bigg/ \frac{dG}{dN_0} \quad (6.11)$$

Fig. 6 - 1 LEF including many body Coulomb interactions, coherence of resonant-tunneling transport and non-parabolicity for different biases at 100 K. The inset shows the results in a short scale. The points indicate the values of LEF at the peak position of gain spectra.

Figure 6 - 1 shows the α -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 α -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 achieves around -0.7 when electric pumping does not exceed the designed bias. The sign of α -value at gain peak reflects the asymmetry of gain spectrum relative to the lasing central frequency ω_λ . 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 lower laser level and extraction level (see Fig. 3 - 4 in chapter 3). 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 6 - 2 shows the details of α -parameter at frequencies of gain peak, 0.2 THz red-shift and 0.2 THz blue-shift relative to the peak position for different biases computed from the microscopic model with “many body + non-parabolicity” (considering both many-body and non-parabolicity effects), microscopic free-carrier model (considering both free carriers and non-parabolicity but neglecting the renormalization of band structure and Rabi frequency), the SDM models with and without RT, respectively. According to the SDM models with and without RT, 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) simultaneously tend to be resonance, the absolute α value rises with the increasing bias in the regime of negative injection and extraction detunings according to SDM model with RT. 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 ω_λ . Furthermore, the non-parabolicity can only induce the slight influence on the LEF according to the comparisons between free-carrier model and macroscopic one with RT, 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 demonstrated in chapter 3. 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.

Fig. 6 - 2 LEF as a function of the applied bias at the gain peak, 0.2 THz redshift and 0.2 THz blueshift points, respectively, at 100 K. From the left to the right, the curves correspond to microscopic model “many body + non-parabolicity”, microscopic free-carrier model, simplified density matrix (SDM) model with and without RT, respectively. The lines are meant to guide the eye.

6.4 Linewidth enhancement factor in mid-IR QCLs

For mid-IR QCLs, we performed the microscopic density matrix modeling of the device based on the three-well vertical design, as shown in Fig. 3 - 9 in chapter 3. This structure has been used in the LEF experiment [102, 103]. It is also very similar with the one used for another LEF experiment in [175]. The Hamiltonian of the system of this mid-IR QCL which characterizes the electron-light coupling, the tunneling effects, free electrons and electron-electron Coulomb interactions, can be rewritten as

$$\begin{aligned}
 H = & - \sum_{j=3,4} \sum_{\mathbf{k}} (\mu_{j5} E b_{5,\mathbf{k}}^\dagger b_{j,\mathbf{k}} + c.c) - \sum_{\mathbf{k}} [(\Delta_{51'}/2) b_{1',\mathbf{k}}^\dagger b_{5,\mathbf{k}} + c.c] \\
 & + \sum_{j=1,3,4,5} \sum_{\mathbf{k}} \varepsilon_{j,\mathbf{k}} b_{j,\mathbf{k}}^\dagger b_{j,\mathbf{k}} + \frac{1}{2} \sum_{uvv'u'} \sum_{\mathbf{k}\mathbf{k}'\mathbf{q}} V_{\mathbf{q}}^{uvv'u'} b_{u,\mathbf{k}+\mathbf{q}}^\dagger b_{v,\mathbf{k}'-\mathbf{q}}^\dagger b_{v',\mathbf{k}'} b_{u',\mathbf{k}}
 \end{aligned} \tag{6.12}$$

The gain G and carrier-induced refractive index change δn are given by

$$G = - \frac{2\omega_\lambda}{\varepsilon_0 n c V_m \xi} \text{Im} \left[\sum_{\mathbf{k}} (\mu_{45} P_{45,\mathbf{k}} + \mu_{35} P_{35,\mathbf{k}}) \right] \tag{6.13}$$

$$\delta n = \frac{\mu}{\varepsilon_0 n V_m \xi} \text{Re} \left(\sum_{\mathbf{k}} (\mu_{45} P_{45,\mathbf{k}} + \mu_{35} P_{35,\mathbf{k}}) \right) \tag{6.14}$$

Then the linewidth enhancement factor α can be obtained by Eq. (6.11).

Figure 6-3 shows the α -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 α -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. The sign of the α -value at gain peak reflects the asymmetry of gain spectrum relative to the lasing central frequency ω_λ . The experimental α -value of this active region design at gain peak is around -0.5 [103]. This experimental value is -0.28 larger than our simulation result. It can be attributed to the refractive index change due to device self-heating effect demonstrated in [116], which needs a more complicated calculation 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 relative large LEF at lasing frequency. In addition, for DFB-QCLs, the emission wavelengths will be shifted to longer wavelengths as the injection current increases [177], the value of LEF at the operation wavelength will rise as the external bias increases. Because the α -spectrum is blue-shifted as the external bias increases deduced from Fig. 6-3, the LEF will increase as the injection current rises. This could explain the experimental observations in a QCL, where the active region is very similar with the one used in this calculation (see Fig. 3 - 9 in chapter 3), of the increases of LEF with injection current in [175]. It is noted that the measured LEF does not include the contribution from the device self-heating effect due to self-mixing technique used in this experiment.

Fig. 6 - 3 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 α at the peak position of gain spectrum.

Figure 6-4 shows the details of α -parameter at the gain peak position, the 0.18 μm 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), 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. 6-4, 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. 3-10 in chapter 3, where the non-parabolicity can more greatly modify the gain spectrum as compared with Coulomb interaction.

Fig. 6 - 4. LEF as a function of the applied bias at the gain peak, 0.18 μm redshift and blueshift points, respectively, at 100 K. From the left to the right, the curves correspond to microscopic model “many body + non-parabolicity”, microscopic free-carrier model, SDM model with and without resonant tunneling, respectively. The lines are meant to guide the eye.

The non-parabolicity in $\text{In}_{0.53}\text{Ga}_{0.47}\text{As}$ 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. 6 - 5. The increase is attributed to the influence of non-parabolicity on symmetry of gain spectrum, as shown in Fig. 3 - 10 in chapter 3. 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 α -value.

Fig. 6 - 5. LEF at the two effective mass ratios m_5/m_4 of electrons of 1.5 and ~ 1.3 using the microscopic model “many body + non-parabolicity”.

6.5 Experimental approach

Some methods have been developed to measure the linewidth enhancement factors e.g. self-mixing method [176] and Hakki-Paoli method [116]. For the Hakki-Paoli method, the LEF is extracted from the refractive index change by measuring the frequency shift of longitudinal mode resonance and the differential gain as the carrier density by slightly changing the current. However, the Hakki-Paoli method is only applied below threshold. The self-mixing method has been used to measure the LEF of both mid-IR [176] and THz QCLs [174], and can be applied above threshold. In this section, the basic principle of self-mixing method is presented.

The self-mixing method employs the laser internal cavity and an external cavity with weak optical feedback as the interferometer, as shown in Fig. 6-11. Due to optical feedback with sinusoidally changing phase, the mixing optical field shows a time resolved intensity variation in the intracavity, and depends on the phase difference of the laser field and the back-reflected light. The phase difference results from the propagation distance difference between the laser light inside the cavity and the light back-reflected. Therefore, the threshold condition and carrier density is changed. This results in the variation of the intensity of the laser light reflecting the LEF, and intensity variation can be adjusted by driving the external mirror. According to the Lang-Kobayashi theory [178, 179], because the threshold gain and the lasing frequency shows a dependence on the phase of the back-reflected light, the power of the emitted light can be written as the function of phase difference ϕ as

$$P(\phi) = P_0 [1 + mF(\phi)] \quad (6.15)$$

where P_0 is the power emitted without feedback, m is the modulation index and $F(\phi)$ is a periodic function and its shape relies on the LEF through the following feedback parameter [180]

$$C = \beta \frac{L\sqrt{1+\alpha^2}}{nl} \sqrt{R_{ext}} \frac{1-R_2}{\sqrt{R_2}} \quad (6.16)$$

where β is unknown coefficient, α is the LEF. L and l are the length of internal and external cavity. n is the intracavity refractive index. R_2 and R_{ext} are reflectivity of end facet of intracavity and external mirror. For very weak feedback ($C \ll 1$), $F(\phi)$ is cosine function, for weak feedback ($C < 1$), $F(\phi)$ is distorted cosine function and moderate feedback ($C > 1$), $F(\phi)$ is sawtooth-like function.

By defining the following parameter [178, 179],

$$X_M = \frac{\phi_{\max} - \phi_{\min}}{2\pi} \quad (6.17)$$

$$X_Z = \frac{\phi_{Z2} - \phi_{Z1}}{2\pi} \quad (6.18)$$

where X_M is the relative distance between the maximum and minimum of the self-mixing waveform, and X_Z is the relative distance between two zero-crossings. Then the feedback parameter and LEF can be derived as [178, 179]

$$C = \pi(X_Z - 0.5) \sqrt{1 + \frac{(X_M - 0.5)^2}{(X_Z - 0.5)^2}} \quad (6.19)$$

$$\alpha = \frac{X_M - 0.5}{X_Z - 0.5} \quad (6.20)$$

Therefore the LEF can be calculated from the measured self-mixing waveforms.

Fig. 6 - 6 The schematic of the experimental setup for the measurement of LEF by using self-mixing mehod.

6.6 Conclusions

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 measured value, our microscopic model can well explain some experimental observations *e.g.* the increases 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.

7 Conclusions and Future work

7.1 Conclusions

This thesis presents the theoretical investigations on the linewidth of mid-IR and THz QCLs. The main focus is on the intrinsic linewidth caused by spontaneous emission, thermal photon, fundamental thermal fluctuation and amplitude-phase coupling (linewidth enhancement factor), which cannot be reduced due to the fundamental quantum limit unless the parameters of the laser system are altered.

Many factors can influence the intrinsic linewidth of QCLs e.g. many-body coulomb interaction, nonparabolicity and coherence of resonant tunneling. Their effects are mainly exhibited by the optical gain and therefore the amplitude-phasing coupling. In order to have a better understanding of the operational principles of QCLs and their effects on linewidth, we firstly study the optical gain by considering these factors. Starting from the microscopic density-matrix model, we have established the Hartree-Fock semiconductor Bloch equations with dephasing and scattering contributions treated at the level of the relaxation-rate approximation, which describes the electron-electron Coulomb interaction, non-parabolicity and coherence of resonant tunneling transport. We use the developed model to investigate the optical gain of a resonant-phonon THz QCL and two-phonon resonance mid-IR QCLs. The simulation results calculated from the microscopic model with “many-body + nonparabolicity”, the microscopic model with “many-body + parabolicity”, the microscopic model with “free-carrier” and the SDM model are compared to demonstrate the importance of those parameters in the simulation of optical properties of QCLs. The results show that the lasing frequency at gain peak calculated by the proposed microscopic density matrix model is closer to the experimentally measured lasing frequency as compared to that calculated by the existing macroscopic density matrix model. The effects of energy detuning, injection and extraction coupling strength, and doping density on optical gain are also systematically investigated. This model provides a more comprehensive picture of the optical properties of QCLs from a microscopic point of view, and potentially enables a more accurate and faster prediction and calculation of the device performance e.g. gain spectra and nonlinear amplitude-phase coupling.

Experimental studies showed both mid-IR and THz QCLs exhibit a very narrow intrinsic linewidth as compared to conventional diode laser. The rate-equation model demonstrated the

narrow linewidth is due to the fast nonradiative process in QCLs. Although the rate-equation model can well explain the reason behind the narrow intrinsic linewidth of QCLs, it cannot adequately describe the electron transport characteristics of QCLs, and therefore cannot accurately describe the linewidth characteristic. We have developed the rate-equation model and built up a new model for the calculation of intrinsic linewidth of QCLs based on quantum Langevin equations. It includes the effects of resonant-tunneling transport and dynamics of coherent interaction associated with the tunneling and laser transition processes. The results show that the coherent interaction of laser transition reduces the linewidth of THz QCLs to some extent due to narrow gain spectrum width. The coupling strength and dephasing rate associated with tunneling have significant effects on the linewidth of THz QCLs in the incoherent resonant-tunneling regime, but small effects on that of mid-IR QCLs due to their strong coherent resonant-tunneling. The linewidth decreases with the increase of the injection coupling strength and reduction of the dephasing rate associated with tunneling. According to our model, a reduced intrinsic linewidth can be easily designed through optimization of the injector barrier and doping density of the active region of QCLs. In addition, the results also demonstrated that, not only the spontaneous emission but also the thermal photon can contribute greatly to the intrinsic linewidth for THz QCLs at high temperature.

In addition to the contributions of spontaneous emission and thermal photon, the temperature fluctuations in thermodynamic equilibrium can induce the linewidth broadening. Even for a cavity in the perfect thermal equilibrium with its surroundings, this thermal noise always exists. A theoretical model based on Green function analysis and *Van Vliet-Fassett* theory has been presented for the analysis of the fundamental frequency noise and linewidth broadening of QCLs caused by intrinsic thermal fluctuations. It is found that the intrinsic frequency noise of QCLs due to thermal fluctuation is a low frequency noise, which shows strong temperature dependence in both THz and mid-IR QCLs. For both THz and mid-IR QCLs, frequency noise does not exhibit a $1/f$ trend in the whole frequency region. This characteristic is not reflected in the experimental measurements, which shows a $1/f$ frequency noise in QCLs at low frequency region of 10 Hz-100 kHz. We attribute this discrepancy to other contributions to frequency noise *e.g.* thermal noise owing to the non-Langevin heat source and carriers trapped by material defects. Further in-depth investigations are required, but the presented study indeed provides groundwork for such derivations. Furthermore, the linewidth broadening due to the temperature dependent

refractive index variations caused by current-induced device self-heating can nearly be neglected in THz QCLs. It is only around 1 Hz, but is comparable to the predicted value of 3 Hz caused by spontaneous emission and thermal photon in certain high power THz QCLs. For mid-IR QCLs, the linewidth increases from 14.74 Hz to 62.02 Hz as the temperature changes from 200 K to 400 K. We also find that if the refractive index change associated with the intersubband gain transition, the thermal expansion and the energy level broadening caused by self-heating are considered, the linewidth broadening should increase at least from 64.3 Hz to 266.4 Hz at 400 K. These microscopic effects should be further determined theoretically and experimentally. The effects of heat and structure parameters, such as heat conductivity of the active region, the thickness of the active region/substrate and the optical mode intensity distribution, on the frequency noise and the linewidth broadening of mid-infrared QCLs have also been investigated. The results show that the fundamental frequency noise and the intrinsic linewidth broadening decrease when the heat conductivity, the thickness of the lower cladding layer and the mode field radius increase. They also increase with the decrease of the thicknesses of the active region and the upper cladding layer. Through the optimization of these heat and structure parameters, the effects on the temperature fluctuations can be effectively reduced.

Finally, we investigate the linewidth enhancement factor of QCLs. Due to the intersubband transition characteristics, QCLs were expected to have a narrow and symmetric gain spectrum and therefore a zero LEF at the gain peak. However, the currently reported experiments 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. We attribute the non-zero LEF at the gain peak to the interplay of the many body interactions, coherence of resonant-tunneling effects and non-parabolicity based on our study on the optical gain of QCLs. We extend the microscopic density-matrix analysis 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. A non-zero LEF at the gain peak is obtained due to these combined effects. Our study demonstrates a strong dependence of the LEF on the lasing frequency detuning is observed. The results show that, for mid-IR QCLs, the coherence of resonant-tunneling effects play a minor role in LEF while the many body Coulomb interaction and non-parabolicity effect plays a key role. In contrast, the coherence of resonant tunneling and many body Coulomb interactions 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 measured value, our microscopic model can well explain some experimental observations *e.g.* the increases 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. By considering all these parameters, one can optimize the active region design to reduce the LEF of QCLs.

7.2 Future work

There are still some aspects that should be further investigated. Here we list some of the perspectives that might be needed for the future works.

- (1) The theoretical investigation on the $1/f$ noise in low frequency range. The flicker $1/f$ -type frequency noise, occurring at lower frequencies, was experimentally observed in QCLs. However, the physical mechanism is not clear and a complex subject in itself. The origin of $1/f$ noise characteristic could probably come from the thermal noise owing to the non-Langevin heat source associated with the emissive and absorptive processes of photons and carriers. It could also come from the noise due to the optical feedback in cold cavities or from the non-Markovian processes or due to the contacts, lattice scattering, carriers trapped by material defects or from fluctuations of electron tunneling through the multi-barriers QCL structure. Further investigations are required to understand this phenomenon.
- (2) The effects of spatial hole burning on intrinsic linewidth. In conventional semiconductor diode lasers, carrier diffusion eliminates spatial hole burning. But, in QCLs, the gain recovery process is faster than carrier diffusion, and spatial hole burning is dominant. It has been well investigated that the spatial hole burning can influence the linewidth enhancement factor and also induce an excess noise due to the non-orthogonality of longitudinal mode. The spatial hole burning may have an effect on linewidth of QCLs, which is worthy to be investigated.
- (3) The many-body model discussed in this thesis provides a relatively comprehensive picture of optical properties of QCLs and enables a more accurate and faster prediction and calculation of the device performance. It also provides an essential design tool for the optimization of the quantum wells/barriers thicknesses to improve the performance of THz QCLs, such as the maximum operation temperature. There are still certain aspects that the model can be further improved. First, this model takes into account dephasing mechanisms in laser transition and transport coherence, but only considers them under the relaxation-rate approximation using

phenomenological values, where the nondiagonal scattering contributions are neglected. Nondiagonal scattering contribution is expected to yield not only a smaller linewidth but also a reshaping of the gain spectrum. A more stringent treatment of the dephasing parameters is required for future theoretical studies. Another limitation is that the present implementation of many-body model considers only intramodule electron-light scatterings but neglects intermodule dipole moment contributions due to the choice of the basis states from the isolated modules in the “tight-binding” scheme. Further study is required to include intermodule electron-light scattering.

- (4) Experimental verification on the theoretical calculations presented in this thesis. The effects of resonant tunneling and coherent interaction on intrinsic linewidth, the fundamental thermal noise and $1/f$ frequency noise, and the microscopic contributions, in addition to the contribution of self-heating induced refractive index change, to the LEF should be all experimentally verified. Because QCLs have a very narrow intrinsic linewidth, the accuracy of intrinsic linewidth measurement is limited by many factors e.g. the driver current noise and intensity noise contribution. The very low-noise source, signal detector and frequency stabilization techniques should be used. Further experimental study is required.

Appendix A: Tunnel coupling

Let us consider the following double quantum wells, which are subjected to an external electric field F .

Fig. A1 Conduction band and wavefunction for the (a) uncoupled and (b) coupled double quantum wells under an electric field. E_1 and E_2 are the energy level of uncoupled quantum wells. E' and E'' are eigenenergies of coupled quantum wells.

The Hamiltonian of the double quantum wells are

$$H = -\frac{\hbar^2}{2m^*} \frac{d^2}{dz^2} + V_1(z - z_1) + V_2(z - z_2) + eFz \quad (\text{A.1})$$

with

$$V_i(z - z_i) = \begin{cases} 0 & |z - z_i| > \frac{L_i}{2} \\ -V_0 & |z - z_i| \leq \frac{L_i}{2} \end{cases} \quad i = 1, 2 \quad (\text{A.2})$$

where V_0 is the potential energy. z_1 and z_2 is the central position of quantum wells, L_1 and L_2 are the width of quantum wells.

If φ_1 and φ_2 are the wavefunctions of the isolated well, then the wavefunction of coupled quantum wells are the linear combination of them [92]

$$\varphi(z) = a\varphi_1(z) + b\varphi_2(z) \quad (\text{A.3})$$

The coefficients a and b can be obtained from

$$\begin{pmatrix} E_1 + s_{11} - E & (E_2 - E)r + t_{12} \\ (E_1 - E)r + t_{21} & E_2 + s_{22} - E \end{pmatrix} \begin{pmatrix} a \\ b \end{pmatrix} = 0 \quad (\text{A.4})$$

where r , s and t are called overlap, shift and transfer integral and are defined as [92, 181]

$$\begin{aligned} s_{11} &= \langle \varphi_1 | V_2 | \varphi_1 \rangle, \quad s_{22} = \langle \varphi_2 | V_1 | \varphi_2 \rangle \\ t_{12} &= \langle \varphi_1 | V_1 | \varphi_2 \rangle, \quad s_{21} = \langle \varphi_2 | V_2 | \varphi_1 \rangle \\ r &= \langle \varphi_1 | \varphi_2 \rangle \end{aligned} \quad (\text{A.5})$$

Then the energy levels of each coupled quantum wells can be calculated by solving Eq.(A.4) and Eq.(A.5). If we use the following Hamiltonian to approximation the coupled quantum wells

$$H = \begin{pmatrix} E_1 & \hbar\Omega_{12} \\ \hbar\Omega_{12} & E_2 - \varepsilon_{12} \end{pmatrix} \quad (\text{A.6})$$

Then we can obtain the coupling strength as

$$\Omega_{12} = \frac{1}{2} \sqrt{(E_1 - E_2)^2 - \varepsilon_{12}^2} \quad (\text{A.7})$$

where

$$\varepsilon_{12} = (E_1 + s_{11}) - (E_2 + s_{22}) \quad (\text{A.8})$$

Appendix B: Coulomb matrix elements

The screening Coulomb matrix element $V_q^{uvv'u'}$ is

$$V_q^{uvv'u'} = \frac{e^2}{2A\varepsilon_0\varepsilon_r\varepsilon(q)q} F_{uu'vv'} \quad (\text{B.1})$$

where the form factor F is

$$F_{uu'vv'} = \int dz \int \varphi_u(z) \varphi_{u'}(z) e^{-q|z-z'|} \varphi_v(z') \varphi_{v'}(z') dz' \quad (\text{B.2})$$

The integral of the form factor F needs a heavy computation. We need to ease the numerical burden by using the approximation but without loss of accuracy. This can be achieved by the use of Fourier decomposition proposed by Bonno et al [96]. The following provides the details and the treatment follows the procedures of Ref. [96, 182].

A fast Fourier transform of wavefunction can be expressed as

$$\varphi_u(z) \varphi_{u'}(z) = \sum_{\sigma=-N_H}^{N_H} \gamma_{\sigma}^{uu'} e^{ik_0\sigma z} \quad (\text{B.3})$$

where $k_0 = 2\pi/L$ and L is the integral interval of the wavefunction in z direction. $\gamma_{\sigma}^{uu'}$ is the σ th Fourier coefficient.

Rewriting Eq. (B.2) using the Fourier series, we obtain

$$F_{uu'vv'} = \sum_{\sigma_1} \sum_{\sigma_2} \gamma_{\sigma_1}^{uu'} \gamma_{\sigma_2}^{vv'} G_{\sigma_1\sigma_2} \quad (\text{B.4})$$

where

$$G_{\sigma_1\sigma_2} = \int_0^L dz \int_0^L dz' e^{ik_0\sigma_1 z} e^{-q|z-z'|} e^{ik_0\sigma_2 z'} \quad (\text{B.5})$$

With the variable change

$$\begin{cases} Z = (z + z')/2 \\ Z' = (z - z')/2 \end{cases} \quad (\text{B.6})$$

we have

$$\begin{aligned} G_{\sigma_1\sigma_2} &= 2 \int_0^L dZ e^{ik_0(\sigma_1+\sigma_2)Z} \int_{-\min\{Z, L-Z\}}^{\min\{Z, L-Z\}} dZ' e^{-q|Z'|} e^{ik_0(\sigma_1-\sigma_2)Z'} \\ &= \frac{2L\delta_{\sigma_1, -\sigma_2}}{q^2 + \sigma_2^2 k_0^2} + \frac{2(e^{-qL} - 1)(q^2 + \sigma_1\sigma_2 k_0^2)}{(q^2 + \sigma_1^2 k_0^2)(q^2 + \sigma_2^2 k_0^2)} \end{aligned}$$

(B.7)

Obviously, equation (B.7) can help us speed up the computation dramatically. Pereira et al [95] have proposed an alternative method to efficiently evaluate the Coulomb matrix elements. The idea is to use two limits, $q \rightarrow 0$ and $q \rightarrow \infty$, to calculate the main matrix elements, and then interpolate for finite wave vector q .

Appendix C: Hartree-Fock approximation

In the derivation in this thesis, some following relations are used:

$$[b_1^\dagger b_2, b_3^\dagger b_4] = b_1^\dagger b_4 \delta_{2,3} - b_3^\dagger b_2 \delta_{1,4} \quad (C.1)$$

$$[b_1^\dagger b_2^\dagger b_3 b_4, b_c^\dagger b_d] = b_1^\dagger b_2^\dagger b_3 b_d \delta_{c,4} - b_c^\dagger b_2^\dagger b_3 b_4 \delta_{d,1} - b_1^\dagger b_2^\dagger b_4 b_d \delta_{c,3} + b_c^\dagger b_1^\dagger b_3 b_4 \delta_{d,2} \quad (C.2)$$

For the simplicity, we drop the script of wave vector $\mathbf{k}$ and assume that any Coulomb interactions Hamiltonian can be expressed as

$$H_{cc} = \frac{1}{2} \sum_{a,b,c,d} V_{abcd} b_a^\dagger b_b^\dagger b_c b_d \quad (C.3)$$

Then we can write out the following relation

$$\begin{aligned} [b_p^\dagger b_q, H_{cc}] &= \frac{1}{2} \sum_{a,b,c,d} V_{abcd} [b_p^\dagger b_q, b_a^\dagger b_b^\dagger b_c b_d] \\ &= \frac{1}{2} \sum_{a,b,c,d} V_{abcd} (b_p^\dagger b_b^\dagger b_c b_d \delta_{q,a} - b_p^\dagger b_a^\dagger b_c b_d \delta_{q,b} - b_a^\dagger b_b^\dagger b_c b_q \delta_{p,d} + b_a^\dagger b_b^\dagger b_d b_q \delta_{p,c}) \\ &= \frac{1}{2} \left(\sum_{b,c,d} V_{qbcd} b_p^\dagger b_b^\dagger b_c b_d - \sum_{a,c,d} V_{aqcd} b_p^\dagger b_a^\dagger b_c b_d - \sum_{a,b,c} V_{abcp} b_a^\dagger b_b^\dagger b_c b_q + \sum_{a,b,d} V_{abpd} b_a^\dagger b_b^\dagger b_d b_q \right) \\ &= \sum_{a,b,d} (V_{qabd} b_p^\dagger b_a^\dagger b_b b_d + V_{abpd} b_a^\dagger b_b^\dagger b_d b_q) \end{aligned} \quad (C.4)$$

Therefore, according to Eq. (C.4) and using the Heisenberg equation, we have

$$\frac{d}{dt} \langle b_p^\dagger b_q \rangle \Big|_{cc} = \frac{i}{\hbar} \sum_{a,b,d} (V_{qabd} \langle b_p^\dagger b_a^\dagger b_d b_b \rangle - V_{abpd} \langle b_a^\dagger b_b^\dagger b_d b_q \rangle) \quad (C.5)$$

Obviously, in order to solve the above two-operator term, we must determine the four-operator term. The four-operator term can be again obtained using the Heisenberg equation. However, due to the electron-electron interaction, the four-operator term will be coupled with six-operator term, and so on. Hence, we should solve an infinite hierarchy of differential equations involving higher-order density matrix elements. But, in order to obtain a solution, we only consider the finite order terms without loss of accuracy. We factorize the four-operator term $\langle b_1^\dagger b_2^\dagger b_3 b_4 \rangle$ into products of two-operator term $\langle b_1^\dagger b_2^\dagger b_3 b_4 \rangle_f$ and a correlation term $\langle b_1^\dagger b_2^\dagger b_3 b_4 \rangle_c$, namely

$$\langle b_1^\dagger b_2^\dagger b_3 b_4 \rangle = \langle b_1^\dagger b_2^\dagger b_3 b_4 \rangle_f + \langle b_1^\dagger b_2^\dagger b_3 b_4 \rangle_c \quad (C.6)$$

where

$$\langle b_1^\dagger b_2^\dagger b_3 b_4 \rangle_f = \langle b_1^\dagger b_4 \rangle \langle b_2^\dagger b_3 \rangle - \langle b_1^\dagger b_3 \rangle \langle b_2^\dagger b_4 \rangle \quad (\text{C.7})$$

By inserting Eq. (C.5) into Eq. (C.6), the dynamical equation can be decomposed into Hartree-Fock contributions and collision (correlation) contributions [86].

$$\frac{d}{dt} \langle b_p^\dagger b_q \rangle \Big|_{cc} = \frac{d}{dt} \langle b_p^\dagger b_q \rangle \Big|_{HF,cc} + \frac{d}{dt} \langle b_p^\dagger b_q \rangle \Big|_{col,cc} \quad (\text{C.8})$$

where

$$\frac{d}{dt} \langle b_p^\dagger b_q \rangle \Big|_{HF,cc} = \frac{i}{\hbar} \sum_{a,b,d} \left(V_{qabd} \langle b_p^\dagger b_a^\dagger b_d b_b \rangle_f - V_{abpd} \langle b_a^\dagger b_b^\dagger b_d b_q \rangle_f \right) \quad (\text{C.9})$$

$$\frac{d}{dt} \langle b_p^\dagger b_q \rangle \Big|_{col,cc} = \frac{i}{\hbar} \sum_{a,b,d} \left(V_{qabd} \langle b_p^\dagger b_a^\dagger b_d b_b \rangle_c - V_{abpd} \langle b_a^\dagger b_b^\dagger b_d b_q \rangle_c \right) \quad (\text{C.10})$$

Hartree-Fock contributions can be obtained by inserting Eq. (C.7) into Eq. (C.9)

$$\begin{aligned} \frac{d}{dt} \langle b_p^\dagger b_q \rangle \Big|_{HF,cc} &= \frac{i}{\hbar} \sum_{a,b,d} V_{qabd} \left(\langle b_p^\dagger b_b \rangle \langle b_a^\dagger b_d \rangle - \langle b_p^\dagger b_d \rangle \langle b_a^\dagger b_b \rangle \right) \\ &\quad - \frac{i}{\hbar} \sum_{a,b,d} V_{abpd} \left(\langle b_a^\dagger b_q \rangle \langle b_b^\dagger b_d \rangle - \langle b_a^\dagger b_d \rangle \langle b_b^\dagger b_q \rangle \right) \end{aligned} \quad (\text{C.11})$$

According to Eq.(C.3) and Eq. (C.11), in the Hartree-Fock approximation, the decomposition of the four particle correlations leads to the same result as approximation H_{cc} by the one-particle Hamiltonian H_{cc}^{MF} [97]

$$H_{cc}^{MF} = \sum_{ab} \left[\sum_{cd} (V_{cabd} - V_{acbd}) \langle b_c^\dagger b_d \rangle \right] b_a^\dagger b_b \quad (\text{C.12})$$

Therefore, for the Hartree-Fock approximation, the many-particle system is assumed as a set of independent particles without the possibility of collisions, and each particle motions through the average field induced by all particles. The collision contributions are described in the frame of the second order correlation [86].

Appendix D: Operator diffusion coefficients

For the dynamical variation o_u , we have the following quantum Langevin equations

$$\frac{do_u}{dt} = d_u + f_u(t) \quad (D.1)$$

where d_u is the drift coefficients and F_u is assumed to have zero average and to be delta-correlated Langevin forces, i.e.

$$\langle f_u(t) \rangle = 0 \quad (D.2)$$

$$\langle f_{u^\dagger}(t) f_v(t') \rangle = 2 \langle d_{u^\dagger v} \rangle \delta(t-t') \quad (D.3)$$

The associated diffusion coefficients d_{uv} can be calculated using the *generalized Einstein relations* [128, 131]

$$2 \langle d_{u^\dagger v} \rangle = \frac{d}{dt} \langle o_u^\dagger o_v \rangle - \langle d_u^\dagger o_v \rangle - \langle o_u^\dagger d_v \rangle \quad (D.4)$$

For example, the diffusion coefficient corresponding to the fluctuation force $f_4(t)$,

$$\langle f_4(t) f_4(t') \rangle = 2 d_{44} \delta(t-t') \quad (D.5)$$

can be obtained from Eq. (D.4)

$$\begin{aligned} \langle f_4(t) f_4(t') \rangle &= \left\langle \frac{d}{dt} (\rho_4)^2 - \left(\frac{d}{dt} \rho_4 \right) \rho_4 - \rho_4 \left(\frac{d}{dt} \rho_4 \right) \right\rangle \delta(t-t') \\ &= \left\langle -\gamma_{sp} \rho_4 - \gamma_4 \rho_4 + i(g^* a^\dagger \sigma_{34} - g \sigma_{34}^\dagger a) - \frac{i\Omega_{41'}}{2\hbar} (\sigma_{41'}^\dagger - \sigma_{41'}) \right\rangle \\ &\quad - \left\langle -\gamma_{sp} \rho_4 \rho_4 - \gamma_4 \rho_4 \rho_4 + i(g^* a^\dagger \sigma_{34} - g \sigma_{34}^\dagger a) \rho_4 - \frac{i\Omega_{41'}}{2\hbar} (\sigma_{41'}^\dagger - \sigma_{41',k}) \rho_4 \right\rangle \\ &\quad - \left\langle -\gamma_{sp} \rho_4 \rho_4 - \gamma_{4f} \rho_4 \rho_4 + i\rho_4 (g^* a^\dagger \sigma_{34} - g \sigma_{34}^\dagger a) - \frac{i\Omega_{41'}}{2\hbar} \rho_4 (\sigma_{41'}^\dagger - \sigma_{41'}) \right\rangle \\ &= \langle (\gamma_{sp} + \gamma_4) \rho_4 \rangle \delta(t-t') \end{aligned} \quad (D.6)$$

Applying the same procedure to other Langevin forces, one can obtain

$$\langle f_3(t) f_3(t') \rangle = \langle (\gamma_{sp} + 1/\tau_{43}) \rho_4 + \gamma_{3eff} \rho_3 \rangle \delta(t-t') \quad (D.7)$$

$$\langle f_{34}^\dagger(t) f_{34}(t') \rangle = \langle (2\gamma_{34} - \gamma_{sp} - 1/\tau_{43}) \rho_4 + \gamma_{3eff} \rho_3 \rangle \delta(t-t') \quad (D.8)$$

$$\langle f_{34}(t) f_{34}^\dagger(t') \rangle = \langle (\gamma_{sp} + 1/\tau_{43}) \rho_4 + (2\gamma_{34} - \gamma_{3eff}) \rho_3 \rangle \delta(t-t') \quad (D.9)$$

$$\langle f_{34}(t) f_{34}(t') \rangle = 0 \quad (D.10)$$

$$\langle f_{41'}^\dagger(t) f_{41'}(t') \rangle = \langle \gamma_{3eff} n_3 + (\gamma_4 - 1/\tau_{43}) \rho_4 + 2\gamma_{41'} \rho_{1'} \rangle \delta(t-t') \quad (D.11)$$

$$\langle f_{41'}(t) f_{41'}^\dagger(t') \rangle = \langle (1/\tau_{43} - \gamma_4 + 2\gamma_{41'}) \rho_4 - \gamma_{3eff} \rho_3 \rangle \delta(t-t') \quad (D.12)$$

Appendix E: c -number diffusion coefficients

For c -number O_u corresponding to the operator o_u , we have the following c -number quantum Langevin equations

$$\frac{dO_u}{dt} = D_u + F_u(t) \quad (\text{E.1})$$

The diffusion coefficients D_{uv} can be calculated using the *generalized Einstein relations*

$$2\langle D_{u^\dagger v} \rangle = \frac{d}{dt} \langle O_u^\dagger O_v \rangle - \langle D_u^\dagger O_v \rangle - \langle O_u^\dagger D_v \rangle \quad (\text{E.2})$$

If $o_u^\dagger o_v$ is normally ordered, its expectation value is equal to the expectation value of the corresponding c -number product [128, 131]. Therefore, we have

$$\frac{d}{dt} \langle o_u^\dagger o_v \rangle = \frac{d}{dt} \langle O_u^\dagger O_v \rangle \quad (\text{E.3})$$

According to Eq. (E.3) and (D.3), we find that

$$\langle F_{u^\dagger} F_v \rangle = \langle f_{u^\dagger} f_v \rangle + \langle d_{u^\dagger} o_v \rangle + \langle o_{u^\dagger} d_v \rangle - \langle D_{u^\dagger} O_v \rangle - \langle O_{u^\dagger} D_v \rangle \quad (\text{E.4})$$

If $o_u^\dagger o_v$ is not normally ordered, we can use the commutator relationship of operator $o_u^\dagger o_v = o_v o_u^\dagger - [o_v, o_u^\dagger]$ to bring it into the chosen order.

Then the c -number second moments have

$$\langle F_A(t) F_A(t') \rangle = \langle f_A(t) f_A(t') \rangle \quad (\text{E.5})$$

$$\langle F_{34}^\dagger(t) F_{34}(t') \rangle = \langle f_{34}^\dagger(t) f_{34}(t') \rangle \quad (\text{E.6})$$

$$\langle F_{34}(t) F_{34}(t') \rangle = \langle f_{34}(t) f_{34}(t') \rangle + 2ig \langle p_{34} A \rangle \delta(t - t') \quad (\text{E.7})$$

Publication

Journal

1. Z. Yan, Y. Tang, X. Li, T. Liu, P. Shum, X. Yu, Y. Zhang, and Q. J. Wang, "Numerical study of mid-infrared supercontinuum generation in 2- noise-like pulses," *submitted*.
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1. T. Liu and Q. J. Wang "Linewidth in quantum cascade lasers" ACP 2013, Beijing, Nov. 2013, invited talk.
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