MONTE CARLO SIMULATION OF ELECTRON DYNAMICS IN QUANTUM CASCADE LASERS

by

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<td>Modal gain vs current density at the lattice temperature of 80 K, without the X-valley transport, for the improved 7.9 µm QCL. Electronic states used in the simulation were obtained from the self-consistent Schrödinger-Poisson solver, as described in Chapter 2. The dash-dot horizontal line denotes the total waveguide loss (the waveguide was designed by Mithun D’Souza). The intersection of the loss line with the modal-gain linear curve identifies the threshold current density.</td>
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MONTE CARLO SIMULATION OF ELECTRON DYNAMICS IN QUANTUM CASCADE LASERS

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Quantum cascade lasers (QCLs) are electrically pumped unipolar coherent semiconductor lasers, grown on InP or GaAs substrates and based on intersubband transitions and tunneling. QCLs have potential for wide applications in molecular spectroscopy, chemical sensing, and telecommunication in the mid- and far-infrared spectral regions. However, intervalley leakage and internal heating have limited the performance of GaAs-based mid-infrared (mid-IR) QCLs: their emission wavelengths are limited to above 7.4 \( \mu \text{m} \), and their wallplug efficiency is poor (\( \leq 2\% \)).

To address these issues, a detailed Monte Carlo simulator has been developed, which includes both \( \Gamma \)- and X-valley transport for the first time in the QCL community. The \( \Gamma \)-valley electron states are obtained using the \( k \cdot p \) method, and the X-valley states are obtained within the effective mass framework. All the relevant scattering mechanisms are included: electron-longitudinal optical phonon, electron-electron, and intervalley scattering.

The simulator has been applied to investigate the X-valley leakage in two published equivalent-design GaAs/AlGaAs mid-IR QCLs. The X-valley leakage current is quantified and the dominant X-valley leakage mechanism is identified for the first time. The X-valley leakage is shown to have very different effects on the performance of the two devices. Good agreement with experiment is obtained at both cryogenic and room temperatures.

The simulator has also been used to simulate and optimize the injector design of deep-active-well GaAs-based QCLs for room-temperature operation, emitting at 6.7 \( \mu \text{m} \), the shortest wavelength to date in GaAs QCLs. The optimized structure obtained shows sufficient gain for lasing at
both low and room temperatures, with its performance comparable to the state-of-the-art 9.4 μm GaAs QCL.

Spatial phonon confinement has been modeled in detail and fully incorporated in the simulator developed, and its effects on the electronic transport properties of mid-IR GaAs QCLs are thoroughly investigated. Simulation results demonstrate that the inclusion of phonon confinement induces a minor correction to the device performance, so the bulk-GaAs-phonon approximation in GaAs QCLs remains useful due to its simplicity and high accuracy. This finding resolves a long-term concern regarding the importance of phonon confinement in QCL structures.

Irena Knezevic
ABSTRACT

Quantum cascade lasers (QCLs) are electrically pumped unipolar coherent semiconductor lasers, grown on InP or GaAs substrates and based on intersubband transitions and tunneling. QCLs have potential for wide applications in molecular spectroscopy, chemical sensing, and telecommunication in the mid- and far-infrared spectral regions. However, intervalley leakage and internal heating have limited the performance of GaAs-based mid-infrared (mid-IR) QCLs: their emission wavelengths are limited to above 7.4 µm, and their wallplug efficiency is poor (≤ 2%).

To address these issues, a detailed Monte Carlo simulator has been developed, which includes both Γ- and X-valley transport for the first time in the QCL community. The Γ-valley electron states are obtained using the k·p method, and the X-valley states are obtained within the effective mass framework. All the relevant scattering mechanisms are included: electron-longitudinal optical phonon, electron-electron, and intervalley scattering.

The simulator has been applied to investigate the X-valley leakage in two published equivalent-design GaAs/AlGaAs mid-IR QCLs. The X-valley leakage current is quantified and the dominant X-valley leakage mechanism is identified for the first time. The X-valley leakage is shown to have very different effects on the performance of the two devices. Good agreement with experiment is obtained at both cryogenic and room temperatures.

The simulator has also been used to simulate and optimize the injector design of deep-active-well GaAs-based QCLs for room-temperature operation, emitting at 6.7 µm, the shortest wavelength to date in GaAs QCLs. The optimized structure obtained shows sufficient gain for lasing at both low and room temperatures, with its performance comparable to the state-of-the-art 9.4 µm GaAs QCL.
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Chapter 1

Introduction

Quantum cascade lasers (QCLs) are electrically pumped unipolar coherent light sources. They operate based on intersubband transitions in multiple-quantum-well (MQW) heterostructures, designed by means of band-structure engineering. The operating principle of these lasers is based on two fundamental phenomena in quantum mechanics: tunneling and quantum confinement. The emission wavelength in this type of laser is not limited by the band gap of constituent materials, but can be tuned in a wide range by tailoring the layers thicknesses.

The basic concept of QCLs was introduced by Kazarinov and Suris [2] in 1971. It took more than twenty years before an actual working device was first demonstrated by Faist et al. [3] at Bell Laboratories in 1994. Since then, the lasers’ performance has remarkably improved. The emission wavelength has spanned the mid- to far-infrared spectral range (from 3 to 190 $\mu$m). At present, the best device performances are obtained at wavelengths between 3.8-9.5 $\mu$m, where continuous-wave (CW) room temperature operation is achieved [4].

QCLs are rapidly acquiring new applications, due to their wide tunable emission wavelength range and the capability for above-room-temperature pulsed and CW operation. Mid-infrared (mid-IR) QCLs have been used for real-time spectroscopic detection of environmental gases, whose spectra lie in the 3-5 $\mu$m and 8-13 $\mu$m atmospheric windows [5–7]. Mid-IR QCLs can also be well adapted for chemical spectroscopy in medical applications [8, 9]. They are also candidates for free-space telecommunications: e.g., the Jet Propulsion Laboratory has chosen a QCL-based sensor for the 2009 Mars Science Laboratory mission [7]. Other applications may include
cruise control in conditions of poor visibility, collision-avoidance radar, and industrial process control [10]. Thus, QCLs appear as the semiconductor solution for lasers in the mid- and far-infrared spectral regions [4].

1.1 Quantum Cascade Lasers

Quantum cascade lasers are made of a sequence of alternating wide band-gap and narrow band-gap semiconductor layers, which form quantum wells for carriers. The thickness of each layer is typically a few nanometers. These multiple-quantum-well structures consist of repeated stages (typically 25-70), each stage containing an active region and an electron-injecting region (injector). The injector also works as the electron-collecting region (collector) as well as the Bragg-mirror region (for the electrons in the upper lasing level) for the previous stage. A certain number of layers in each injector region are $n$-type doped with silicon to serve as an electron reservoir. Figure 1.1 depicts the conduction band profile in two adjacent stages of a generic QCL under an applied bias. Due to the quantum confinement in multiple quantum wells, a miniband (green in Fig. 1.1) and a minigap (light blue) are formed in the injector region, but discrete energy levels are formed in the active region (levels 3, 2, 1 in the figure). When an electrical current flows through such a structure, electrons from the injector region tunnel through the barriers and fill the upper lasing level 3 in the active region. Electrons in level 3 make radiative transitions (the yellow wavy lines in Fig. 1.1) to the lower lasing level 2, giving off photons, provided there is population inversion between the two levels (i.e., level 3 has more electrons than level 2). Once electrons relax to level 2, this level gets depopulated fast to level 1 by the electron-longitudinal optical phonon interaction processes, which helps maintain the population inversion between levels 3 and 2. Electrons then tunnel to the next-stage injector region and trigger the photon-emitting process in the next stage. Intuitively, when an electrical current flows through a quantum cascade laser, electrons cascade down an energy staircase (which makes this type of laser earn its name) and each electron emits a photon at each step. An injected electron therefore generates $N_p$ photons, where $N_p$ is the number of stages, leading to an optical power proportional to $N_p$. This cascading process leads to the intrinsic high-power capability of QCLs, but also to high voltages and subsequently low
power-conversion efficiency (so-called wallplug efficiency) which, at the present time, prevents their practical use as CW or quasi-CW light sources.

Figure 1.1 Conduction band profile in two adjacent stages of a generic QCL under an applied bias.

1.1.1 State-of-the-art quantum cascade lasers

The first demonstration of a QCL was achieved in 1994 by Faist et al. [3] using the InGaAs/InAlAs heterostructure lattice-matched to an InP substrate. Since then, tremendous progress has been made in InP-based QCL device performance. CW room-temperature (RT) operation was first demonstrated by Beck et al. [11]. Following this breakthrough, InP-based QCLs have been reported to achieve CW RT operation at wavelengths between 3.8 and 9.5 µm [11–18], with devices in the 4.5-5.5 µm range [15–18] reaching optical output powers in the range of 300-600 mW. However, with one exception [18], all QCLs operating in CW at RT have a relatively low (< 5%) wallplug efficiency, and their electro-optical characteristics are extremely temperature sensitive, due both to carrier backfilling of the lower lasing level and carrier leakage out of
the upper lasing level. Even the exception, that is a narrow-width, buried-heterostructure device which achieved 9% wallplug efficiency at RT [18], has a rapidly decreasing slope efficiency due to carrier leakage to the continuum. The emission wavelengths from 3.5 [19] to 24 μm [20] have been obtained using the same InGaAs/InAlAs materials system grown on InP. A shorter wavelength of 3.05 μm was recently achieved by both Semtsiv et al. [21], using strain-compensated In$_{0.73}$Ga$_{0.27}$As/In$_{0.55}$Al$_{0.45}$As/AlAs on InP, and by Revin et al. [22], using In$_{0.55}$Ga$_{0.47}$As/AlAs$_{0.56}$Sb$_{0.44}$ lattice-matched to InP. Although these shorter-wavelength QCLs can currently only operate in pulsed mode up 150 K, the emission wavelength of high-performance, CW RT InP-based QCLs is expected to cover the entire 3-5 μm atmospheric window in the very near future.

The GaAs/AlGaAs heterostructure grown on a GaAs substrate is another very important materials system for the fabrication of QCLs. In fact, GaAs substrate is the only contender to InP for fabricating QCLs [23]. Compared to InP-based QCLs, GaAs QCLs have several advantages. First of all, GaAs is the most technologically mature semiconductor after silicon, so its feature parameters are accurately determined and documented. Secondly, GaAs QCLs based on the deep-active-well design [24] hold the potential to achieve wallplug efficiency as high as 15% and watt-range CW optical power [25], thus making them potentially the first CW-operating QCLs of practical use. In addition, GaAs QCLs can achieve wavelengths in a very wide range due to the design flexibility offered by the good lattice matching across the full range of Al contents. The emission wavelengths of reported GaAs QCLs have spanned from 7.4 μm (mid-infrared) [26] to 190 μm (far-infrared) [27]. However, GaAs-based QCLs have several drawbacks that limit their performance and will be discussed in Sec. 1.1.3. This work is restricted to the study of GaAs-based mid-IR QCLs.

The very first attempt at using the GaAs/AlGaAs materials system in QCL structures was back in 1997, when Strasser et al. [28] reported the electroluminescence of a GaAs QCL design at 6.9 μm using 45% Al in the barriers (no lasing was observed). Since then, mid-infrared (mid-IR) GaAs-based QCLs have utilized Al contents of 33% [29–31], 45% [1, 32–36], and 100% [37–39] in the AlGaAs barrier layers. The first lasing GaAs QCL structure [29] employed 33% Al in the barriers, and emitted at $\lambda = 9.4$ μm under pulse operation up to 140 K. With a low-loss Al-free
waveguide [30], the threshold current density \( J_{th} \) of the 33% Al QCL design was reduced to an average of 5.0 kA/cm\(^2\) at 77 K, and a maximum pulsed operation temperature of 200 K. Mid-IR GaAs QCLs with 33% Al in the barriers have so far achieved the highest operating temperature of 285 K under pulsed operation [31], and have not achieved CW operation.

On the other hand, great progress has been made in the device performance of GaAs/Al\(_{0.45}\)Ga\(_{0.55}\)As QCLs since their first realization [1]. Room temperature pulsed operation has been reported for several active region designs, i.e., three quantum-well active region design emitting at 9.4 \( \mu m \) [1,32], a superlattice active region design emitting at 12.6 \( \mu m \) [33], and a bound-to-continuum design emitting at 11.0 \( \mu m \) [34]. Continuous wave operation of 45% Al QCLs based on the original design [1] has recently been achieved [35, 36] through optimized device processing, with a maximum temperature of 150 K [36].

Several GaAs/AlAs QCLs emitting in the mid-IR spectral region have also been reported [37–39]. Though showing the lowest temperature dependence of \( J_{th} \) [37] and CW operation close to 30 K [38], these lasers are limited by the negative differential resistance effect, which strongly limits the maximum operating temperature [23]. By depositing InAs monolayers in the device active regions, Carder et al. reported room temperature pulse operation of an InAs/GaAs/AlAs QCL [40] lasing at 8.5 \( \mu m \).

1.1.2 Theoretical approaches for QCL transport simulation

Much theoretical work has been done to accompany the rapid experimental developments of mid-IR QCLs. These include Monte Carlo simulations [41–44], self-consistent rate equations [45, 46], as well as the nonequilibrium Green’s function (NEGF) formalism [47, 48]. Among these simulation approaches, there has been a long-term debate of whether the nature of charge transport in QCL structures is coherent or incoherent. Although recent work [48] using the NEGF formalism argued that the transport in QCLs should be treated as coherent, this formalism takes enormous computation time and has to be carefully customized for each particular structure. As a result, it takes a long time to obtain useful results for improvement of experimental designs.
On the other hand, theoretical studies by Iotti and Rossi [41, 49] showed that, although clear coherent oscillations of population inversion and gain overshoot were observed during the initial transient, the oscillations are damped on the sub-picosecond timescale. This time is much shorter than the average transit time across one stage. These features have been observed experimentally [50, 51] and the timescales are comparable. Hence, they concluded that the resulting steady-state transport in mid-IR QCLs is incoherent (i.e., governed by scattering). Therefore, the QCL simulation community tends to adopt the semiclassical transport concept in their models [43–45]. The self-consistent rate equations model [45] has been used extensively to study the influence of external parameters (e.g., doping density) on the QCL devices performance [46, 52] and provide insight in improving QCL designs [53–55], but this model does not account for the electron dynamics in the plane parallel to the layers, which has been shown critical in determining the actual transport properties in mid-IR QCLs [49].

Based on the broad study of existing theoretical approaches for mid-IR QCL transport simulation, this work employed the incoherent stationary transport model [41] and utilized the popular ensemble Monte Carlo method (EMC) [56] to solve the kinetic Boltzmann-like equations. The incoherent description implies that the transport in mid-IR QCLs is governed by scattering. One of the main advantages of the EMC approach is that it allows one to include, on an equal footing, a large variety of scattering mechanisms on the microscopic level, and the in-plane electron dynamics is automatically taken into account. The computational time of this approach may be longer than that of the rate equations model, but it certainly takes much less time than the NEGF formalism, so it can produce useful results promptly for a QCL design. Furthermore, this approach is very versatile and robust: once the simulator has been developed, it can easily be adapted to simulate transport properties of different QCL structures. As will be shown in the following chapters, the Monte Carlo simulator developed has been used to simulate several QCL structures and all the results are very good and reliable. Another important feature of the simulator is that its results provide a proper direction for QCL design optimization. Therefore, the Monte Carlo simulator developed has the right balance of versatility and rigor.
1.1.3 Issues in GaAs-based QCLs

Although mid-IR GaAs QCLs have achieved dramatic progress in the past few years, these devices have not achieved CW operation at room temperature. Several issues have limited their device performance:

(1) Carrier leakage

The relatively large threshold current density ($J_{th}$) and power consumption [23] of these QCLs have made it very challenging to achieve room temperature CW operation. The high value of $J_{th}$ primarily arises from the high leakage, including the leakage to the $\Gamma$-continuum [57] and to the satellite X-valley [58, 59]. To further improve the performance of GaAs/AlGaAs QCLs, a detailed knowledge of possible current leakage paths and carrier loss in a particular design is highly desirable. Although the leakage via the $\Gamma$-continuum states has been well recognized [1, 23] and studied [60], satellite-valley leakage was not systematically investigated in theory. Properly quantifying both the continuum leakage and the intervalley leakage in GaAs/AlGaAs QCL structures is an essential step towards the improvement of existing designs.

(2) Electron heating

The challenge for GaAs QCLs to operate in CW regime at RT is in part coming from the enormous internal (electron and lattice) heating at high heatsink temperatures [61]. The electron heating is caused by the high input power density and the low power conversion efficiency: the electron system is not able to dissipate (via optical-phonon emission) the relatively large amount of energy provided by the applied bias. The excessive input power heats up the electron system first, and ultimately the whole device through multiple-phonon emission processes. It is important to characterize the degree of electron heating in a given QCL.

(3) Short wavelength limit

The emission wavelength of mid-IR GaAs/AlGaAs QCLs incorporating a single injection barrier has been limited to above 8 $\mu$m due to the intervalley $\Gamma$-X electron transfer [39]. By utilizing a double injection barrier [39] or by depositing InAs monolayers in the wide active wells [26], the emission wavelength is reduced from 8 $\mu$m to 7.3 $\mu$m for the former case, and from 9.4 $\mu$m to 7.4 $\mu$m for the latter case, but the lasing was observed only at cryogenic temperatures in pulsed mode.
for both cases. A novel GaAs-based QCL design is needed to achieve room-temperature emission wavelength below 7 µm, and a rigorous simulation is necessary to characterize the transport properties of the novel design and optimize it for high-performance operation.

(4) Phonon confinement

QCLs are multiple-quantum-well heterostructures, spatially confined along the growth direction. Transport simulation of QCLs generally neglects the influence of spatial confinement on the phonon spectra, and assumes that phonons behave the same way as in bulk semiconductors. It has long been unclear whether the spatial confinement of phonons has a strong effect on the electron transport properties of cascaded structures. There has so far been no systematic account of the phonon confinement on the transport properties of GaAs-based QCLs, such as the current-field characteristics or population inversion.

There are other issues related to GaAs-based mid-IR QCLs, but only the above four will be addressed in this project.

1.2 Overview of This Dissertation

This dissertation aimed at addressing the four issues in GaAs-based mid-IR QCLs that have been outlined at the end of the previous section. In this research, we will show how the use of a multivalley Monte Carlo simulation (MMC) can help address and overcome problematic issues arising in the design of QCLs. This microscopic simulation can help us better understand the physics of the laser operation and pinpoint the fundamental physical limitations. Moreover, computational optimization of a laser design is much cheaper than experimental trial-and-error, and a detailed microscopic simulation such as MMC can help ahead of time identify the culprits in a particular design that lead to leakage or excessive heating, so that time and resources spent on fabrication are minimized. The work performed within this dissertation has led to one book chapter [62], seven journal papers [58, 59, 63–67], and five conference presentations [68–72]. The main accomplishments of this project are highlighted below.
1.2.1 Research accomplishments

A comprehensive Monte Carlo simulator has been developed, which includes both the Γ- and X-valley transport for the first time in the QCL simulation community [58, 59, 63]. (Reported QCL simulations [48, 49, 52] in literature so far have only focused on the Γ-valley transport.) Γ-valley electronic states are obtained by using the $\mathbf{k} \cdot \mathbf{p}$ method [73], while in most literature on QCL modeling Γ-states are obtained by solving the conduction-band effective mass equation. The $\mathbf{k} \cdot \mathbf{p}$ approach can conveniently account for the lattice-mismatch strain effect, and the band mixing between the conduction and valence bands in narrow-gap semiconductor materials [73]. X-valley states are obtained by solving the effective mass equation, since the X valleys are well above the valence bands and the effects of valence bands on the electronic states are negligible. All the relevant interaction mechanisms are included, i.e., electron-longitudinal polar optical (electron-LO) phonon, electron-electron, and intervalley scattering processes, within the same stage and between adjacent stages. The screening of electron-electron scattering is treated within the random phase approximation [74] in the static limit.

The Monte Carlo simulator has been applied to simulate the output characteristics of two well-known equivalent-design mid-IR GaAs-based QCLs. The X-valley leakage current is quantified, and the dominant X-valley leakage mechanism is identified, for the first time in the QCL research area. The X-valley leakage is shown to have a very different effect on the performance of the two QCL structures. Very good agreement with experiment is obtained at both cryogenic and room temperatures. The effects of the electron-electron (e-e) interaction in the two QCLs are studied. The e-e scattering is shown to have two crucial effects: (i) it strongly increases intersubband electron redistribution and optimizes the coupling between the active region and the injector; (ii) it drives the electron distributions in all the subbands to the heated Maxwellian distributions, and all the distributions have an effectively common electron temperature, much higher than the lattice temperature. This part of the project has led to three journal publications [58, 59, 63] and two conference presentations [68, 69].

The Monte Carlo simulator developed is then utilized to simulate and optimize the design of deep-active-well GaAs-based QCL structures, which all emit at 6.7 µm, the shortest wavelength
projected to date in GaAs mid-IR QCLs. (These QCL designs have been developed here at UW-Madison ECE department, as part of a collaboration between Prof. Dan Botez’s and Prof. Irena Knezevic’s groups.) Simulation results clearly show that the initial design is not capable of room-temperature operation, due to the strong wavefunction coupling between the injector states and the Γ-continuum states. Then a second improved structure with reduced coupling is suggested, however, its RT gain just marginally meets the total loss, and its gain at cryogenic temperature is below the loss line. The reason is a large energy separation between the injector ground level and the next-stage upper lasing level, resulting in low injection efficiency. Identification of the issues associated with the first two designs directly helps to obtain a third optimized structure that shows sufficient gain for lasing emission at both 77 K and room temperatures. Its performance is comparable to that of the GaAs/Al_{0.45}Ga_{0.55}As 9.4 µm QCL [1] that has demonstrated the best device performance among published GaAs-based mid-IR QCLs. This part of work has led to two journal papers [64, 65] and two conference papers [70, 71].

Finally, the effects of phonon confinement on the electronic transport properties in mid-IR GaAs QCLs were investigated in detail. The macroscopic dielectric continuum model is used to describe the interface (IF) and confined (CF) optical phonon modes. Dispersions of the IF modes are obtained by using the transfer matrix method with periodic boundary conditions. Normalization coefficients of the IF and CF potentials are derived in detail for MQW structures consisting of arbitrary combinations of binary and ternary alloys. Interstage and intrastage scattering rates due to all the IF and CF modes are calculated for both Γ- and X-valley electrons. The IF and CF scattering processes, in addition to the electron-electron and intervalley phonon scattering, are fully incorporated into the multivalley Monte Carlo simulation of a deep-active-well 6.7 µm GaAs-based MQW QCL [64]. At both 77 K and room temperature, we find that phonon confinement enhances the electron-polar optical phonon scattering rates to a relatively small extent, and induces minor corrections to the current, population inversion, and the electronic temperature, with respect to the results obtained in the bulk-GaAs-phonon approximation. Therefore, the bulk-phonon approximation in transport simulations of GaAs-based QCLs remains valuable due to its simplicity and high
accuracy. Based on the work on phonon confinement, two journal papers are in press [66,67], and the results were also presented at the IWCE-12 conference [72].

1.2.2 Organization of chapters

Chapter 2 describes in detail the theory behind the MMC QCL simulator [58, 59] that I have developed. First of all, the Boltzmann-like transport model for mid-IR QCL structures including both the $\Gamma$- and X-valley transport is presented (Sec. 2.1). The charge-conserving scheme is introduced and it enables one to simulate the transport in QCLs by solving the Boltzmann-like kinetic equations over two adjacent stages (Sec. 2.2). The ensemble Monte Carlo procedure for simulating QCL structures is described and the corresponding flow chart is given (Sec. 2.2.1). Details of solving the $\Gamma$- and X-valley electronic states are then presented in Sec. 2.3. Effects of the electrostatic potential on subband energy levels and wavefunctions are studied by self-consistently solving the Schrödinger-Poisson equations (Sec. 2.4). All the relevant scattering rates are evaluated in detail (Sec. 2.5). The interaction mechanisms included are electron-longitudinal polar optical (electron-LO) phonon, electron-electron, and intervalley scattering processes, within the same stage and between adjacent stages (Sec. 2.5). Calculation of the intersubband dipole matrix element is presented in Sec. 2.6.

In Chapter 3, the multivalley Monte Carlo simulator presented in Chapter 2 is applied to simulate the transport properties of two well-known equivalent-design GaAs/AlGaAs mid-IR QCLs [1, 29]. The relevant $\Gamma$- and X-subband energy levels and wavefunctions for the two lasers are illustrated and the difference in wavefunctions between the two QCLs is pointed out. The field versus current density characteristics at different temperatures for the two QCLs are analyzed, together with the field dependence of the $\Gamma$- and X-valley electron density. The analysis leads to identification of the X-valley leakage mechanism (Sec. 3.1). The effects of the X-valley leakage on the two QCLs are investigated and compared. In addition, the effect of the electron-electron interaction and electron heating are investigated and discussed in Sec. 3.2.

Chapter 4 presents the simulation and optimization of three deep-active-well QCLs, emitting at 6.7 $\mu$m. The Monte Carlo simulator is employed to characterize the output characteristics of the
three lasers. The deep-active-well QCL design is presented first, with a detailed description of its salient features (Sec. 4.1). The injector optimization of the deep-active-well QCLs follows (Sec. 4.2): transport properties of the first two structures with different injector layer sequences are simulated, and the issues associated with them are identified, which lead to the injector optimization of the third structure. The device performance of the optimized structure is compared to that of the state-of-the-art GaAs 9.4 $\mu$m QCL (Sec. 4.3).

In Chapter 5, phonon confinement is fully incorporated in the multivalley Monte Carlo simulator, and its effects are investigated in detail. Details of the phonon spectra and potentials calculation by using the transfer matrix method with periodic boundary conditions are presented in Sec. 5.1.1, and they are followed by the calculation of the phonon potential normalization coefficients in Sec. 5.1.2. The scattering rates of all the $\Gamma$- and X-valley electron states of interest by all the phonon modes are then computed and included in the multivalley simulator (Sec. 5.2). The effects of phonon confinement on the transport properties of GaAs QCLs are analyzed in Sec. 5.3.

Chapter 6 provides a summary of the research performed within this dissertation (Sec. 6.1), and presents a discussion of envisioned future work (Sec. 6.2).
Chapter 2

Theoretical Approach

2.1 Semiclassical Transport Description

Electron transport in modern semiconductor devices can be described by the Boltzmann transport equation (BTE), which is given in its common form as [75]

\[
\frac{\partial f}{\partial t} + \mathbf{v} \cdot \nabla_{\mathbf{r}} f + \frac{\partial p}{\partial t} \cdot \nabla_{\mathbf{p}} f = \sum_{k'} \left\{ S(k', k) f(k') \left[ 1 - f(k) \right] - S(k, k') f(k) \left[ 1 - f(k') \right] \right\}. \tag{2.1}
\]

The function \( f(\mathbf{r}, \mathbf{p}, t) \) is the electron distribution function, which gives the probability of finding electrons at time \( t \), located at position \( \mathbf{r} \), with momentum \( \mathbf{p} \). (Note all vectors in this section are three-dimensional.) The second term (the diffusion term) on the left-hand side (LHS) of Eq. (2.1) describes the net in-flow of electrons to an elementary volume centered at \( \mathbf{r} \) in position space. The third term (the drift term) on the LHS describes the net in-flow of electrons to an elementary volume centered at \( \mathbf{p} \) in momentum space, and is caused by external electric field. The right-hand side (RHS) of Eq. (2.1) is the collision integral, representing the result of competition between in-scattering and out-scattering. \( S(k, k') \) is the transition rate from an initial state \( | k \rangle \) to a final state \( | k' \rangle \), and can be calculated from the Fermi’s Golden Rule [75] as

\[
S(k, k') = \frac{2\pi}{\hbar} |\langle k' | H_{int} | k \rangle|^2 \delta(E(k') - E(k) \mp \hbar \omega), \tag{2.2}
\]

where \( H_{int} \) is the interaction Hamiltonian and the \( \delta \)-function reflects the conservation of energy. Solution of the BTE provides the distribution function \( f(\mathbf{r}, \mathbf{p}, t) \) from which the macroscopic quantities of interest (e.g., electron density, current density) are readily evaluated.
The BTE is a semiclassical transport description, since the LHS of the equation treats electrons as classical particles, but the RHS treats electrons quantum mechanically (as waves). The semiclassical transport description has two underlying assumptions: (i) the spatial variation of any external potential is much greater than the de Broglie wavelength at which the wavelike nature of electrons may be witnessed; (ii) the time of concern is much larger than the time between collisions that is in the range of subpicoseconds. For the two assumptions to hold, dimensions of semiconductor devices have to be relatively large and the time of concern needs to be relatively long. In addition, collisions are assumed to be mutually independent and instantaneously change the electron momentum. Any applied electric field is treated as slowly varying, and it does not interfere with the scattering processes. In the semiclassical approach, all transport calculations are based on the analytical or numerical solutions of the BTE.

2.2 Boltzmann-Like Transport Equation for QCLs

It has been shown [41, 49] that the stationary charge transport in mid-IR QCL heterostructures is incoherent in nature, and can therefore be described by a Boltzmann-like transport equation. These structures are normally grown along the (001) direction, defined as a z-axis, with the x-y plane parallel to the layers. Since the GaAs/AlGaAs QCL heterostructure is the main focus in this work, both Γ and the three X valleys are important in determining the transport properties. Electronic states near the minima of the Γ and X valleys are characterized by $|k\alpha\rangle = |k, \nu\lambda\eta\rangle$, where $k = (k_x, k_y)$ is the wave vector in the x-y plane, $\nu$ is the subband index, $\lambda$ denotes the stage, and $\eta$ is the valley index ($\eta = \Gamma, X_x, X_y, X_z$). The electron distribution function $f_{k\alpha}(t)$, measuring the probability that an electron resides in a state $|k\alpha\rangle$ at time $t$, evolves with time according to the Boltzmann-like transport equation [41]

$$\frac{d}{dt}f_{k\alpha}(t) = \sum_{k'\alpha'} \left\{ S_{\alpha\alpha'}^{\alpha'}(k', k)f_{k'\alpha'}(t) [1 - f_{k\alpha}(t)] - S_{\alpha}^{\alpha'}(k, k')f_{k\alpha}(t) [1 - f_{k'\alpha'}(t)] \right\}, \quad (2.3)$$

where $S_{\alpha\alpha'}^{\alpha'}(k, k')$ is the total transition rate from state $|k\alpha\rangle$ to $|k'\alpha'\rangle$, and $S_{\alpha}^{\alpha'} = \sum_s S_{\alpha\alpha'}^{\alpha'}$, is the sum over all possible scattering mechanisms $s$. Compared to the standard BTE [Eq. (2.1)], Eq. (2.3) does not have the in-plane drift and diffusion terms on the LHS, since there is no electric field.
applied in the $x$-$y$ plane, and the plane is large enough to be regarded as having uniform distribution
in real space. The electric field applied along $z$ does not explicitly accelerate electrons, but through
modifying the $\Gamma$ and X band profiles, affects the electronic states and therefore indirectly changes
the electron distribution. The motion of electrons along $z$ is purely hopping among subbands due
to scattering.

The translational symmetry of QCL structures (i.e., the wavefunctions in any two stages are
simple translations in space and energy) allows for simulating electron transport over a generic
central stage $\lambda$ only. Spatially remote stages have little wavefunction overlap with the central stage,
making it sufficient to limit the interstage scattering ($\lambda' \neq \lambda$) to the nearest neighbors ($\lambda' = \lambda \pm 1$).
A schematic of three adjacent stages under bias is shown in Fig.2.1.

Figure 2.1 Schematic of three adjacent stages of a QCL under an applied field, illustrating the
charge-conserving scheme.

In addition, periodicity of the structures ensures the validity of the charge-conserving scheme
[41] employed in our EMC simulation: each time an electron in a state $|k, \nu \lambda \eta \rangle$ undergoes an
interstage scattering process to a new state $|k', \nu' (\lambda \pm 1) \eta \rangle$ (e.g., processes $2$ and $3$ in Fig. 2.1),
another electron is properly injected into the central region by the corresponding processes $1$ and $4$, so that the number of electrons in the simulated stage $\lambda$ is conserved. Furthermore, the
transition rates for processes 2 and 3 can be regarded as the same as those of processes 1 and 4, respectively, thanks to the translational symmetry. As a benefit of this equivalence, it is only necessary to solve for energy levels and wavefunctions in two full adjacent stages (e.g., stages $\lambda$ and $\lambda + 1$) to compute all the intrastage ($\lambda' = \lambda$) and nearest-neighbor interstage ($\lambda' = \lambda \pm 1$) transition rates.

The current density $J$ across the whole device is defined in terms of the net electron flux through the interface between the simulated stage $\lambda$ and the next stage $\lambda + 1$,

$$J \propto \sum_{k\nu\eta} \sum_{k'\nu'\eta'} \left[ S_{\nu\lambda\eta}(k, k') f_{\nu\lambda\eta}(k) - S_{\nu'\lambda\eta'}(k, k') f_{\nu'(\lambda+1)\eta'}(k) \right].$$  \hspace{1cm} (2.4)

In the EMC implementation, $J$ is calculated by counting the interstage scattering events. Details of obtaining the energy levels and wavefunctions in stages $\lambda$ and $\lambda + 1$ are described in Sec. 2.3.

#### 2.2.1 Ensemble Monte Carlo method

The ensemble Monte Carlo (EMC) method [56] is an efficient approach for solving the standard BTE [Eq. (2.1)] and the Boltzmann-like equation [Eq. (2.3)] using random numbers. (Details for implementing an EMC simulation of bulk semiconductors or devices can be found in Refs. [56, 75, 76].) The EMC method is based on calculating the motion of an ensemble of particles during a short time $dt$. It can be applied to analyze the transient as well as steady-state electron transport in unipolar devices. Figure 2.2 shows the flight dynamics of an EMC calculation. The horizontal lines show the trajectories of particles 1 through N along the time axis. Each star-like symbol on the horizontal line represents a scattering event, where an electron is scattered by a phonon or another electron. Between two consecutive scattering events, an electron flies freely in the $x$-$y$ plane (no external field is applied in the $x$-$y$ plane). Random numbers are used to determine the time between two consecutive scattering events (also called the free-flight time), and to choose the proper scattering process among many scattering mechanisms. The vertical dotted lines indicate the sampling times, spaced by $dt$, at which the electron distribution function $f$ is evaluated from the ensemble. Macroscopic quantities of interest (e.g., electron density, current density) can be
readily evaluated from the distribution $f$. The time interval $dt$ should be chosen small enough (typically 1 femtosecond) to capture the right electron dynamics.

Figure 2.2 Flight dynamics of the ensemble Monte Carlo method. The horizontal lines show the trajectories of particles 1 through N along the time axis, and each star-like symbol on the horizontal line represents a scattering event. The vertical dotted lines indicate the sampling times, spaced by $dt$, at which the electron distribution function $f$ is evaluated from the ensemble.

Figure 2.3 shows a flow chart of the ensemble Monte Carlo transport kernel for QCL structures. Between scattering events, particles fly freely in the plane parallel to the layers ($x$-$y$ plane) and remain in a given subband in the $z$-direction. Positions of electrons in the $x$-$y$ plane are not tracked due to translational symmetry (the structure is considered unbound in the $x$-$y$ plane), and the electron transport in QCL structures is essentially vertical (scattering between subbands determines the transport). Input of the Monte Carlo transport kernel is provided by a self-consistent Schrödinger-Poisson solver, as depicted in Fig. 2.4, which solves for the electronic states in the conduction band of a QCL structure. It was found that the Poisson solver has a negligible effect on the electronic states for typical doping densities ($\leq 4 \times 10^{11}$ cm$^{-2}$) in QCLs, as investigated in Sec. 2.4, so in order to reduce the computation time, the electron states from the Schrödinger solver within the linear potential drop approximation were directly used to compute the scattering rates.
Figure 2.3 Flowchart of the ensemble Monte Carlo transport kernel for QCL structures.
in the transport kernel. Output from the transport kernel includes the current density $J$, subband electron density $n_i$, population inversion $\Delta n$, optical modal gain $G_m$, and electron temperature $T_e$.

![Flowchart of an EMC simulation for QCL structures.](image)

**Figure 2.4** Flowchart of an EMC simulation for QCL structures.

## 2.3 Electronic States

### 2.3.1 Γ-valley electronic states

In most literature on QCL modeling, Γ-valley electronic states are obtained using the conduction-band effective mass equation, whereas this work employs the $k \cdot p$ method to obtain the Γ-states. The $k \cdot p$ method provides a good way of solving the electronic band structure near the band edges, where most of the interesting physical phenomena occur in optical devices. It can conveniently account for the lattice-mismatch strain effect, and the influence of valence bands in narrow-gap semiconductor materials [73].

The standard eight-band $k \cdot p$ model [77, 78] consists of the conduction band (c), heavy-hole (hh), light hole (lh), and the spin-orbit split-off (so) band, all with their spin degeneracies. The
Hamiltonian matrix can be found for unstrained [77] and lattice-mismatched biaxially strained [78] bulk semiconductors without external perturbation. With the in-plane wave vector \( k = 0 \) and the remaining spin degeneracy, the eight-band \( k \cdot p \) model [78] reduces to a three-band problem, with the hh band factoring out. The reduced \( 3 \times 3 \) matrix takes the form

\[
H_{3\times3} = \begin{bmatrix}
E_c + (1 + 2F_K)E_z + A_e & \frac{2}{3}E_P E_z & \frac{1}{3}E_P E_z \\
\sqrt{\frac{2}{3}}E_P E_z & E_v - (\gamma_1 + 2\gamma_2)E_z - P_e + Q_e & -2\sqrt{2}\gamma_2 E_z + \sqrt{2}Q_e \\
\sqrt{\frac{1}{3}}E_P E_z & -2\sqrt{2}\gamma_2 E_z + \sqrt{2}Q_e & E_v - \Delta_{so} - \gamma_1 E_z - P_e
\end{bmatrix},
\]

where

\[
E_z = \frac{\hbar^2 k^2}{2m_0}, \\
E_P = \frac{2m_0 P_{cv}^2}{\hbar^2}, \\
P_{cv} = \frac{\hbar}{m_0} \langle iS | p_x | X \rangle, \\
\gamma_1 = \gamma_1^L - \frac{E_P^2}{3E_g}, \\
\gamma_2 = \gamma_2^L - \frac{E_P}{6E_g}, \\
A_e = a_c(\epsilon_{xx} + \epsilon_{yy} + \epsilon_{zz}), \\
P_e = -a_v(\epsilon_{xx} + \epsilon_{yy} + \epsilon_{zz}), \\
Q_e = -\frac{b}{2}(\epsilon_{xx} + \epsilon_{yy} - 2\epsilon_{zz}).
\]

\( E_c \) and \( E_v \) are the unstrained conduction- and valence-band edges, respectively, relative to a common reference energy. \( \Delta_{so} \) is the spin-orbit split-off energy. \( \gamma_1 \) and \( \gamma_2 \) are the modified Luttinger parameters [79], and \( \gamma_1^L \) and \( \gamma_2^L \) are the standard Luttinger parameters [80]. The explicit inclusion of the interaction between the valence and conduction bands in the \( k \cdot p \) model leads to the modified Luttinger parameters discussed by Pidgeon and Brown [79]. \( F_K \) [81] represents the contribution of higher conduction bands to the effective mass of the \( \Gamma_6 \) conduction-band electrons. \( P_{cv} \) is the interband momentum matrix element defined by Kane [81], and \( E_P \) is the energy parameter for \( P_{cv} \). \( a_c, a_v, \) and \( b \) are the Bir-Pikus deformation potentials [82], with the sign conventions being negative, positive, and negative, respectively.
For a strained semiconductor layer pseudomorphically grown on a (001)-oriented \((z\text{ axis})\) substrate, the strain tensor components are \([73]\)

\[
\begin{align*}
\epsilon_{xx} &= \epsilon_{yy} = \frac{a_0 - a}{a}, \\
\epsilon_{zz} &= -\frac{2c_{12}}{c_{11}}\epsilon_{xx}, \\
\epsilon_{xy} &= \epsilon_{yz} = \epsilon_{zx} = 0,
\end{align*}
\] (2.7)

where \(a_0\) and \(a\) are the lattice constants of the substrate and the layer material, respectively, and \(c_{11}\) and \(c_{12}\) are the elastic stiffness constants of the strained layer. Therefore, \(A_\epsilon\), \(P_\epsilon\), and \(Q_\epsilon\) are simplified to

\[
\begin{align*}
A_\epsilon &= 2a_c \left( 1 - \frac{c_{12}}{c_{11}} \right) \epsilon_{xx}, \\
P_\epsilon &= -2a_c \left( 1 - \frac{c_{12}}{c_{11}} \right) \epsilon_{xx}, \\
Q_\epsilon &= -b \left( 1 + \frac{2c_{12}}{c_{11}} \right) \epsilon_{xx}.
\end{align*}
\] (2.8)

The material parameters for most III-V semiconductors are documented in Ref. \([83]\).

Within the envelope function approximation (EFA) and the three-band \(k\cdot p\) model, the full \(\Gamma\)-valley electron wavefunctions near the zone-center in each layer of a heterostructure can be written as \([73, 77]\)

\[
\Psi_{n,k}^\Gamma(r, z) = \frac{1}{\sqrt{A}} e^{i\mathbf{k}\cdot\mathbf{r}} \sum_j \psi_{n,j}^\Gamma(z) u_{j0}^\Gamma(r, z),
\] (2.9)

where \(A\) is the normalization area, \(r = (x, y)\), \(j\) sums over the three bands (c, lh, so), and \(\psi_{n,j}^\Gamma(z)\) is the normalized envelope function, slowly varying with coordinate \(z\) over the elementary cells of the crystalline lattice; the periodic Bloch function \(u_{j0}^\Gamma(r, z)\) conserves its bulk properties within each layer except the small (1-2 monolayer wide) regions in the vicinity of the interfaces, and is assumed not to differ much from layer to layer. It should be noted that \(|\mathbf{k}| \ll \pi/a\) in Eq. (2.9), with \(a\) being the monolayer thickness, because only the electron states near the minimum of the \(\Gamma\) valley are of interest in this work.
In the presence of an external potential, the envelope functions $\psi_{n,j}^\Gamma(z)$ satisfy the second-order differential equations [73,77]

$$
\sum_{j'} [H_{jj'}(k = 0, k_z) + U^\Gamma(z)\delta_{jj'}] \psi_{n,j'}^\Gamma(z) = E_n^\Gamma \psi_{n,j}^\Gamma(z),
$$

(2.10)

where $H_{jj'}(k = 0, k_z)$ are the elements of the $3 \times 3$ $\mathbf{k} \cdot \mathbf{p}$ matrix in Eq. (2.5), and $k_z$ will be replaced by the differential operator $-i\partial/\partial z$; $\delta_{jj'}$ is the Kronecker delta, and $j'$ runs over the three bands (c, lh, so); the external potential $U^\Gamma(z) = -|e|F(z - d/2) - |e|\phi(z)$, where $F$ is the applied uniform electric field, and $\phi(z)$ is the electrostatic potential owing to the specific distribution of free electrons from dopants in the system. ($\phi(z)$ is the correction to the approximation of linear potential drop, and it can be obtained from solving the Poisson equation.) The effects of $\phi(z)$ on the electronic states will be investigated in Sec. 2.4, and are disregarded in this section and Sec. 2.3.2. The band discontinuity at the interfaces is taken into account in $E_c$ and $E_v$ in the $H_{jj'}$ matrix.

The coupled differential Eqs. (2.10) may be solved by using the transfer matrix method [84], the finite difference method [85], or the finite element method [86]. However, all these real-space numerical methods would generate spurious solutions (i.e., solutions outside of the first reduced Brillouin zone (BZ) of the periodic structure) [87]. The reciprocal-space numerical technique [88, 89] ensures the wave vector $k_z$ well within the first BZ to avoid spurious solutions, and is employed in our simulation. This approach relies on the periodicity of the QCL structure along the $z$ direction. The envelope function $\psi_{n,j}^\Gamma(z)$ is expanded in a Fourier series [89]

$$
\psi_{n,j}^\Gamma(z) = \sum_m a_{mn}^\Gamma e^{iG_mz},
$$

(2.11)

where $d$ denotes the length of two stages, $G_m = 2m\pi/d$ ($m = 0, \pm1, \pm2, \cdots$) are the reciprocal vectors associated with the periodic heterostructure along the $z$-axis, and index $m$ is an integer, $-N_G \leq m \leq N_G$ ($N_G = 50$ used), which runs over the number of Fourier components retained. Substituting this expression into Eq. (2.10), multiplying both sides by $e^{-iG_mz}$, and integrating over $d$, one obtains an approximation to Eq. (2.10) as the eigenvalue problem

$$
\sum_{j,m'} \sum_{j',m} a_{mn}^{\Gamma,j} \frac{1}{d} \int_0^d dz e^{i(G_m - G_{m'})z} [H_{jj'}(G_m, z) + U^\Gamma(z)\delta_{jj'}] = E_n^{\Gamma} a_{mn}^{\Gamma}. 
$$

(2.12)
The matrix element $H_{jj'}(G_m, z)$ has the same form as Eq. (2.5) for each layer at position $z$, with $k_z$ replaced by $G_m$. Eq. (2.12) is a standard eigenvalue problem for the coefficients $a_{m,j,n}^{\Gamma,j'}$ with a super-matrix of size $3(2N_G+1) \times 3(2N_G+1)$. A diagonalization of this matrix then leads to $3(2N_G+1)$ eigenenergies $E_n^{\Gamma}$ and the corresponding eigenvectors. The top $2N_G+1$ energy levels belong to the conduction subbands, and the associated eigenvectors construct the full subband wavefunctions. The remaining $2(2N_G+1)$ eigenenergies and eigenvectors characterize the valence subbands, which are not of interest in this work, since QCLs utilize electron transitions in the conduction band. As an example, Fig. 2.5 illustrates the energy levels and wavefunction moduli squared of ten $\Gamma$-valley subbands in two adjacent stages for the 45% QCL of Ref. [1] without $\phi(z)$ (linear potential drop only).

Figure 2.5 Energy levels and wavefunction moduli squared of ten $\Gamma$-valley subbands in two adjacent stages for the 45% QCL of Ref. [1] within the linear potential drop approximation. The bold black curves denote the lasing levels (3-upper, 2-lower, 1-ground), while thin black curves are the injector states. The red and green curves indicate the $\Gamma$ continuum-like states.
After obtaining $\psi_{n,j}^\Gamma(z)$, an average in-plane effective mass (to be used in the EMC calculation) is calculated for each $\Gamma$-conduction subband $n$ as

$$\frac{1}{m_n^*} = \sum_j \int_0^d \frac{d z}{m^*(j, z)} |\psi_{n,j}^\Gamma(z)|^2,$$

(2.13)

where $m^*(j, z)$ is the effective mass of band $j$ of the material at position $z$. Based on the new electron effective mass, the in-plane energy dispersion is regarded as parabolic: $E_{n,k}^\Gamma = E_n^\Gamma + \hbar^2 k^2 / 2m_n^*$.

### 2.3.2 X-valley electronic states

There are two groups of X valleys in (001)-grown QCL structures: the $X_z$ valley along the axis $z$, and two side valleys $X_x, X_y$ oriented along the $x$ and $y$ directions, respectively. The $X_x$ and $X_y$ valleys are equivalent within the effective mass framework, and are regarded as one doubly-degenerate $X_x$ valley.

In the EFA, near the X valley minima, the electron wavefunctions $\Psi_{n,k}^{X_x}(r, z)$ and $\Psi_{n,k}^{X_z}(r, z)$ can be expressed by

$$\Psi_{n,k}^{X_x}(r, z) = \frac{1}{\sqrt{A}} e^{i(k \cdot r + \pi a_x z)} \psi_n^{X_x}(z) u_{X_x}^{\ell}(r, z),$$

(2.14a)

$$\Psi_{n,k}^{X_z}(r, z) = \frac{1}{\sqrt{A}} e^{i(k \cdot r)} \psi_n^{X_z}(z) u_{X_z}^{\ell}(r, z),$$

(2.14b)

where $\psi_n^{\ell}(z) (\ell = X_x, X_z)$ are the envelope functions along $z$, $u_{X}^{\ell}(r, z)$ are the Bloch functions, and $a$ is the monolayer width.

To solve for $\psi_n^{\ell}(z)$, the effective-mass equations are sufficient, since the X valleys are well above the valence bands and the effect of the valence bands on the electronic states is negligible. $\psi_n^{X_x}(z)$ and $\psi_n^{X_z}(z)$ in each layer satisfy the effective-mass Eqs. (2.15a) and (2.15b), respectively.

$$\left[ \frac{-\hbar^2}{2m_{X_x}^*(z)} \frac{d^2}{d z^2} + U_X(z) \right] \psi_n^{X_x}(z) = E_n^{X_x} \psi_n^{X_x}(z),$$

(2.15a)

$$\left[ \frac{-\hbar^2}{2m_{X_z}^*(z)} \frac{d^2}{d z^2} + U_X(z) \right] \psi_n^{X_z}(z) = E_n^{X_z} \psi_n^{X_z}(z),$$

(2.15b)
where \( m^{*}_t(z) \) and \( m^{*}_l(z) \) are the corresponding transverse and longitudinal X-valley electron effective mass; \( U_X(z) = E_X(z) - |e|F(z - d/2) \) is the external potential, where \( E_X(z) \) contains the X-valley conduction-band offset relative to a common energy reference level.

In the reciprocal-space approach [89], following a similar procedure as described for the \( \Gamma \)-subband envelope function \( \psi_{\Gamma, n,j}^{\Gamma}(z) \), Eqs. (2.11) and (2.12), one obtains the eigenvalue problem

\[
\sum_{m',m} a_{mn}^{Xx} \frac{1}{d} \int_0^d dz e^{i(G_m - G_{m'})z} \left[ \frac{\hbar^2 G_m^2}{2m^{*}_t(z)} + U_X(z) \right] = E_n^{Xx} a_{mn}^{Xx},
\]

for the Fourier coefficients \( a_{mn}^{Xx} \). The eigenvalue problem for \( a_{mn}^{Xz} \) is similar to Eq. (2.16) with \( m^{*}_t(z) \) replaced by \( m^{*}_l(z) \). Solutions to the eigenvalue problems provide \( 2N_{G+1} \) energy levels and the corresponding eigenvectors for the \( X_x \) (\( X_z \)) subbands, where \( X_x \) subbands are doubly-degenerate. Figures 2.6 and 2.7 show the energy levels and wavefunction moduli squared of the \( X_x \)- and \( X_z \)-subbands, respectively, in two adjacent stages for the 45% QCL [1], without \( \phi(z) \) (linear potential drop only).

Figure 2.6 Energy levels and wavefunction moduli squared of six \( X_z \)-subbands (doubly-degenerate) in two adjacent stages for the 45% QCL [1] within the linear potential drop approximation.
Figure 2.7 Energy levels and wavefunction moduli squared of nine Xz-subbands in two adjacent stages for the 45% QCL [1] within the linear potential drop approximation.

With the envelope function $\psi^\ell_n(z)$, one can compute the average in-plane longitudinal ($m_{ln}^\ell$) and transverse ($m_{tn}^\ell$) electron effective mass for each X-subband $n$,

$$\frac{1}{m_{ln}^\ell} = \int_0^d \frac{|\psi_n^\ell(z)|^2}{m_n^\ell(z)} \, dz, \quad i = l, t. \quad (2.17)$$

For GaAs, X-valley effective masses are $m_l^* = 1.3$ and $m_t^* = 0.23$, while for AlAs, $m_l^* = 0.97$ and $m_t^* = 0.22$. A parabolic in-plane dispersion relation is assumed, with the averaged effective mass:

$$E_{n,k}^{Xz} = E_n^{Xx} + \frac{\hbar^2 k^2}{2m_{tn}^*}, \quad (2.18a)$$

$$E_{n,k}^{Xz} = E_n^{Xx} + \frac{\hbar^2 k^2}{2m_{tn}^*}. \quad (2.18b)$$

The above wavefunctions $\Psi^\eta_{n,k}$ and energy dispersions $E_{n,k}^{\eta}$ ($\eta = \Gamma, X_x, X_z$) are obtained by treating two adjacent stages as one period. They need to be properly assigned to each stage according to the localization of wavefunctions. Then the lowest $N_\eta$ conduction subbands in each stage are selected for transport simulation. $N_\eta$ values depend on the QCL structure and applied field, and is significantly smaller than the actually obtained levels ($2N_G + 1$ or so). One can relabel
the chosen electronic states in stage \( \lambda \) as \( |k, \nu \lambda \eta \rangle \) (\( \nu \)th subband, \( \lambda \)th stage, and \( \eta \)th valley), the associated effective mass as \( m_{\nu \lambda}^{\eta} \), and the energy dispersion \( E_{\nu \lambda}^{\eta}(k) \), which satisfies the following relation
\[
E_{\nu \lambda}^{\eta}(k) = \frac{\hbar^2 k^2}{2m_{\nu \lambda}^{\eta}} + E_{\nu \lambda}^{\eta}.
\]
(2.19)
The states \( |k, \nu \lambda \eta \rangle \) are rewritten in the form
\[
|k, \nu \lambda \Gamma \rangle = \frac{1}{\sqrt{A}} e^{i k \cdot r} \sum_j \psi_{j, \nu \lambda}^{\Gamma}(z) u_{\Gamma j 0}(r, z),
\]
(2.20a)
\[
|k, \nu \lambda X_x \rangle = \frac{1}{\sqrt{A}} e^{i (k \cdot r + \pi a_x)} \psi_{\nu \lambda}^{X_x}(z) u_{X_x}(r, z),
\]
(2.20b)
\[
|k, \nu \lambda X_z \rangle = \frac{1}{\sqrt{A}} e^{i k r} \psi_{\nu \lambda}^{X_z}(z) u_{X_z}(r, z).
\]
(2.20c)
The subband index \( \nu = 1, 2, \cdots, N_{\eta} \) for valley \( \eta \). Similar notation is used for electronic states in stage \( \lambda+1 \).

2.4 Self-Consistent Schrödinger-Poisson Solver

The last two subsections have focused on solving the Schrödinger equations under the assumption that the electrostatic potential \( \phi(z) = 0 \) (potential drop is linear). This subsection accounts for \( \phi(z) \), a correction to the linear potential drop which would imply charge neutrality everywhere, by solving the Schrödinger equations self-consistently with the Poisson equation. Also, we investigate the influence of \( \phi(z) \) on the subband energies and wavefunctions, and explore the validity of disregarding \( \phi(z) \) in the simulation to reduce the computational burden.

\( \phi(z) \) has the same profile in each stage due to the translational symmetry of QCL structures, and satisfies the one-dimensional Poisson equation
\[
\frac{\partial}{\partial z} \left( \varepsilon_s \frac{\partial \phi}{\partial z} \right) = -|e| \left[ N_D^+(z) - n(z) \right],
\]
(2.21)
where \( \varepsilon_s \) is the static dielectric permittivity. An effective constant value for \( \varepsilon_s \) is assumed throughout the heterostructure, which is a reasonable approximation for GaAs/AlGaAs systems. \( N_D^+(z) \) and \( n(z) \) are the ionized donor and electron concentrations, respectively, and the hole concentration \( p(z) \) is neglected since QCL structures are \( n \)-type doped. Assuming conservation of charge in
each stage (i.e., the charge-conserving scheme in Sec. 2.2), one can define a position-independent quasi-Fermi level $E_F$ in each stage [44, 90]. The electron concentration $n(z)$ in stage $\lambda$ is related to the electronic wavefunctions by

$$n(z) = \sum_{\nu=1}^{N_{\Gamma}} \sum_j |\psi_{j,\nu\lambda}(z)|^2 N_{\nu s} + \sum_{\ell} \sum_{\nu=1}^{N_{\ell}} |\psi_{\ell\nu\lambda}(z)|^2 N_{\ell s},$$

(2.22)

where the sum over $j$ is over the three (c, lh, so) bands, $\ell$ sums over the $X_x$ and $X_z$ valleys, and $N_{\nu s} (\eta = \Gamma, X_x, X_z)$ is the sheet electron density in subband $\nu$. $N_{\nu s}$ can be calculated as [73]

$$N_{\nu s} = \frac{2}{A} \sum_k \frac{1}{1 + \exp \left( \frac{E_{\nu\lambda}(k) - E_F}{k_B T} \right)},$$

(2.23)

where the Fermi-Dirac distribution is used for the in-plane electron distribution in each subband, $E_{\nu\lambda}(k)$ is the electronic dispersion relation given by Eq. (2.19), and $T$ is the lattice temperature. (The electron ensemble and the lattice are assumed to have the same temperature here.) Making use of the “sum-to-integral” rule

$$\frac{1}{A} \sum_k = \frac{1}{(2\pi)^2} \int d^2 k = \int_0^{\infty} \frac{2\pi k dk}{(2\pi)^2},$$

(2.24)

and the integral identity

$$\int \frac{dx}{1 + e^x} = \int \frac{e^{-x}}{1 + e^{-x}} dx = -\ln(1 + e^{-x}),$$

(2.25)

the sheet electron density is obtained as

$$N_{\nu s} = \frac{k_B T m_{\nu s}^*}{\pi \hbar^2} \ln \left[ 1 + \exp \left( \frac{E_F - E_{\nu\lambda}^*}{k_B T} \right) \right].$$

(2.26)

For $\eta = X_x$, Eq. (2.26) needs to be multiplied by 2 to give $N_{\nu s}^{X_x}$, since the $X_x$ valley is doubly-degenerate. The sum of $N_{\nu s}^\eta$ in one stage (active region + injector) is equal to the total sheet electron density $N_s$ from the charge neutrality condition [73, 90, 91]

$$N_s = \sum_{\eta} \sum_{\nu=1}^{N_{\eta}} N_{\nu s}^\eta = \int_{\text{one stage}} N_{\eta s}^+ (z) dz,$$

(2.27)

where $\eta$ sums over all valleys ($\eta = \Gamma, X_x, X_z$). For a given $N_s$, the quasi-Fermi level $E_F$ is obtained by iteratively solving Eq. (2.27). When including $\phi(z)$ in Eqs. (2.10), (2.15a), and
Figure 2.8 Flow chart of a self-consistent Schrödinger-Poisson solver.
(2.15b), it is required to solve these equations self-consistently with Eqs. (2.21)-(2.22) and (2.26)-(2.27). The flow chart of a self-consistent calculation is given in Fig. 2.8.

The results from the self-consistent Schrödinger-Poisson solver in two adjacent stages for the 45% QCL [1] at $F = 60$ kV/cm and $T = 77$ K are presented in Figs. 2.9-2.10. Figure 2.9 shows the electrostatic potential energy profile $-\epsilon|\phi(z)$ from the self-consistent Schrödinger-Poisson solver. The Poisson equation is solved using the total $n(z)$ in the stage defined by the red $\times$ marks with periodic boundary conditions, and then the potential is translated to the whole simulated region. For this particular example, the potential energy $-\epsilon|\phi(z)$ has a peak-to-peak value of about 25 meV, but the external potential energy drop is $-\epsilon FL_p = 270$ meV ($L_p = 45$ nm, the length of one stage), the former being less than 10% of the applied potential energy drop. This small band bending owing to the specific electron distribution slightly changes the electronic structure (energies and wavefunctions), as shown in the right panel of Fig. 2.10.

![Figure 2.9](image)

Figure 2.9 Electrostatic potential energy profile $-\epsilon|\phi(z)$ from the self-consistent Schrödinger-Poisson solver in two adjacent stages. The $\times$ marks define the stage where the Poisson equation is solved with periodic boundary conditions, and the resulting potential is then translated to the whole simulated region. It is noticed that the potential is periodic between stages.

It is seen from Fig. 2.10 that $\Gamma$-subband wavefunctions remain nearly the same and the subband energy levels are modified within a few meVs when the band bending is included. This result holds
true for all doping levels less than $4.0 \times 10^{11}$ cm$^{-2}$, in agreement with the calculation in Ref. [46].

Thanks to the small effect of potential $\phi(z)$ on the electronic states, $\phi(z)$ is neglected due to low doping levels ($< 4 \times 10^{11}$ cm$^{-2}$) in this work.

![Graph](image)

**Figure 2.10** Ten $\Gamma$-subband energy levels and wavefunction moduli squared for the 45% QCL [1] with $\phi(z) = 0$ (a) and with $\phi(z)$ (b) obtained from the self-consistent Schrödinger-Poisson solver.

### 2.5 Scattering Mechanisms

The various scattering mechanisms included in the Monte Carlo simulation are listed in Table 2.1. The intervalley scattering mechanism is written in the form of $A \rightarrow B$ (e.g., $\Gamma \rightarrow X_x$) in the table, with $A$ being the initial valley and $B$ the final valley. The electron-LO phonon scattering is included for all the valleys, but the electron-electron (e-e) interaction is implemented only for the $\Gamma$ states. (Electron population of the X valleys is relatively low under typical operating conditions, so the e-e scattering is not important.) The intervalley $X_x \rightarrow X_x$ scattering takes place due to the double-degeneracy of the $X_x$ states. For all the scattering mechanisms, both the intrastage and interstage scattering events are included, with the latter yielding the current flow through the QCL. The interstage $\lambda \rightarrow \lambda - 1$ scattering rates are equal to those for the interstage $\lambda + 1 \rightarrow \lambda$, due
to the translational symmetry. The Pauli exclusion principle of final states is included in the EMC code following the rejection technique of Lugli and Ferry [92].

Table 2.1 Scattering mechanisms included in the Monte Carlo simulator for the $\Gamma$- and $X$-valley states.

<table>
<thead>
<tr>
<th>Mechanism</th>
<th>$\Gamma$ valley</th>
<th>$X_x$ valley</th>
<th>$X_z$ valley</th>
</tr>
</thead>
<tbody>
<tr>
<td>intrastage</td>
<td>electron-LO</td>
<td>electron-LO</td>
<td>electron-LO</td>
</tr>
<tr>
<td>$(\lambda \to \lambda)$ &amp; electron-electron &amp; $\Gamma \to X_x$ &amp; $X_x \to \Gamma$ &amp; $X_z \to \Gamma$</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>&amp; interstage       &amp; $\Gamma \to X_z$ &amp; $X_z \to X_x$ &amp; $X_z \to X_x$</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$(\lambda \to \lambda \pm 1)$ &amp; electron-LO &amp; $X_x \to X_x$ &amp; $X_x \to X_x$</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

2.5.1 **Electron-longitudinal optical (LO) phonon scattering**

The scattering of electrons with longitudinal polar optical (LO) phonons in a multiple-quantum-well (MQW) system is usually described by the Fröhlich interaction [93]. Though electrons have been widely accepted as quasi-two-dimensional (2D) particles in such systems, the correct model for the lattice vibrational modes is still an area of active research. The assumption of dispersionless unscreened bulk phonon modes has been shown [94] to reproduce the total scattering rates reasonably well in quantum-well systems, and is adopted in our calculation of the electron-LO phonon scattering rates in Chapters 2-4. The effect of spatial phonon confinement on the electron-LO scattering strength is studied in Chapter 5, where it will be shown that the bulk phonon approximation is sufficiently good.

The transition rate from a state $|k, \nu \lambda \eta\rangle$ to a new state $|k', \nu' \lambda' \eta\rangle$ ($\lambda'$ could be $\lambda$, $\lambda + 1$, or $\lambda - 1$) is given by Fermi’s Golden Rule

$$S_{\nu' \lambda' \eta}^{\nu \lambda \eta}(k, k') = \frac{2\pi}{\hbar} |\langle k', \nu' \lambda' \eta | H_{e-LO} | k, \nu \lambda \eta \rangle|^2 \delta(E' - E \mp \hbar \omega_0),$$

(2.28)

where $\hbar \omega_0$ is the LO phonon energy, $\delta(\cdot)$ is the delta function, and the upper and lower signs correspond to phonon absorption and emission, respectively (this convention will be used throughout
this section). The electron-LO interaction Hamiltonian \( H_{e-LO} \) takes the following form

\[
H_{e-LO} = \left[ \frac{e_0^2 \hbar \omega_0}{2V} \left( \frac{1}{\varepsilon_\infty} - \frac{1}{\varepsilon_0} \right) \right]^{1/2} \sum_Q \frac{1}{iQ} \left( \hat{a}_Q e^{iQ \cdot R - i\omega_0 t} + \hat{a}_Q^+ e^{-iQ \cdot R + i\omega_0 t} \right),
\]

(2.29)

for bulk LO phonons, where \( e_0 \) is the universal electron charge, \( V \) is the generic volume, \( \varepsilon_0 \) and \( \varepsilon_\infty \) are the static and high-frequency dielectric permittivity, respectively, \( Q \) is the bulk phonon wave vector, \( R = (r, z) \), \( \hat{a}_Q \) and \( \hat{a}_Q^+ \) are the phonon destruction and creation operators, respectively. The total scattering rate for an electron initially in subband \( \nu \), stage \( \lambda \), to a final subband \( \nu' \), stage \( \lambda' \), can obtained as \([56, 93]\)

\[
A_{\nu, \nu'}^{\lambda, \lambda'}(k) = \sum_{k'} C_{\nu, \nu'}^{\lambda, \lambda'}(k, k') = \frac{e_0^2 \hbar \omega_0 \mu_{\nu, \nu'}^s}{8\pi \hbar^3 \varepsilon_\infty} \left( n_0 + \frac{1}{2} \mp \frac{1}{2} \right) \left( \frac{1}{\varepsilon_\infty} - \frac{1}{\varepsilon_0} \right) \times \int_0^{2\pi} d\theta F_{\nu, \nu'}^{\lambda, \lambda'}(q^\pm) \theta (E_k + E_{\nu, \nu'}^\mp - \hbar \omega_0),
\]

(2.30)

where \( n_0 \) is the equilibrium phonon number, \( q = |k' - k| \), \( \theta \) is the angle between \( k \) and \( k' \), and \( \theta(\cdot) \) is the Heaviside step function.

The form factor function \( F_{\nu, \nu'}^{\lambda, \lambda'}(q) \) depends on the valleys. For the \( \Gamma \) valley, since the electron wavefunctions include the influence of the valence bands, the form factor function is calculated by averaging over the elementary cell and employing the orthogonality of the zone-center Bloch functions \( u_{j0}(r, z) \) \((j = c, lh, so)\). Then one obtains

\[
F_{\nu, \nu'}^{\lambda, \lambda'}(q) = \sum_{j, j'} \int_0^d dz \int_0^d dz' \psi_{j, \nu, \lambda}(z) \psi_{j', \nu', \lambda'}(z') \psi_{j, \nu, \lambda}^*(z) \psi_{j', \nu', \lambda'}^*(z') e^{-q|z - z'|}.
\]

(2.31)

For \( \ell = X_x, X_z \) valleys, the form factor is calculated using the envelope functions as

\[
F_{\nu, \nu'}^{\lambda, \lambda'}(q) = \int_0^d dz \int_0^d dz' \psi_{\nu, \lambda}(z) \psi_{\nu', \lambda'}(z') \psi_{\nu, \lambda}^*(z) \psi_{\nu', \lambda'}^*(z') e^{-q|z - z'|}.
\]

(2.32)

The allowed \( q \)'s are fixed by the in-plane momentum conservation \((k' = k \pm q)\) and the energy conservation \([Eq. (2.28)]\) laws. Taking into account the difference of the electron effective mass between the initial and final states, one obtains

\[
q^\pm = \sqrt{\frac{2}{\hbar}} \left\{ \left( m_{\nu, \lambda}^{\nu, \lambda} + m_{\nu, \lambda'}^{\nu, \lambda'} \right) E_k + m_{\nu, \lambda}^{\nu, \lambda} \left( \hbar \omega_{\nu, \nu'}^\pm \right) \right\}^{1/2} - 2 \left[ m_{\nu, \lambda}^{\nu, \lambda} m_{\nu', \lambda}^{\nu', \lambda} E_k \left( E_k + \hbar \omega_{\nu, \nu'}^\pm \right) \right]^{1/2} \cos \theta,
\]

(2.33)
with $\hbar\omega_{\nu,\nu'} = E_{\nu,\lambda}^{\eta} - E_{\nu',\lambda'}^{\eta'} \pm \hbar\omega_0$. Figure 2.11 plots the form factors $F_{3\Gamma}^{3\Gamma}(q)$ ($3 \rightarrow 3$), $F_{3\Gamma}^{2\Gamma}(q)$ ($3 \rightarrow 2$), and $F_{3\Gamma}^{1\Gamma}(q)$ ($3 \rightarrow 1$) as a function of $q$ for the $\Gamma$-subbands 3, 2, 1 in the left panel of Fig. 2.10.

Figure 2.11 Selected form factors for intrasubband ($3 \rightarrow 3$) and intersubband ($3 \rightarrow 2$, $3 \rightarrow 1$) electron-LO scattering within the same stage $\lambda$ for the $\Gamma$-subbands 3, 2, 1 shown in the left panel of Fig. 2.10.

### 2.5.2 Intervalley scattering

It has been shown [95] that the phonon-assisted intervalley $\Gamma - X_x$ and $\Gamma - X_z$ electron transitions can be modeled through an intervalley deformation potential (DP). For bulk intervalley phonons, the total intervalley scattering rate for an electron from a state $|k, \nu,\lambda,\eta\rangle$ to a state of subband $\nu'$, stage $\lambda'$, and valley $\eta'$ is given by [93]

$$\Lambda_{\nu',\lambda',\eta'}^{\nu,\lambda,\eta}(k) = \frac{Z_{\eta'} D_{\eta,\eta'}^{2} m_{\nu,\lambda} n_{\eta,\eta'}^{*}}{2\hbar \rho E_{\eta,\eta'}} \left( n_{\eta,\eta'}^{*} + \frac{1}{2} \mp \frac{1}{2} \right) I_{\nu,\lambda,\eta} \vartheta \left( E_{k} + E_{\nu,\lambda}^{\eta} - E_{\nu',\lambda'}^{\eta'} \pm E_{\eta,\eta'}^{*} \right), \quad (2.34)$$

where $D_{\eta,\eta'}$ is the intervalley $(\eta - \eta')$ deformation potential constant, $\rho$ is the mass density, and $E_{\eta,\eta'}$ is the intervalley phonon energy; $n_{\eta,\eta'}$ is the equilibrium intervalley phonon occupancy, and $Z_{\eta'}$ is the degeneracy of the final valley ($Z_\Gamma = 1$, $Z_{X_x} = 1$, and $Z_{X_z} = 1$ for intervalley $X_x - X_x$, 2
for others). The overlap integral $I^{\nu'\lambda'\eta}_\nu^\nu$ is given by
\begin{equation}
I^{\nu'\lambda'\eta}_\nu^\nu = \sum_j \int_0^d dz \left| \psi_{j,\nu\lambda}(z) \right|^2 \left| \psi_{\nu'\lambda'}(z) \right|^2,
\end{equation}
where the first equation is for the scattering between $\Gamma$ and $\ell$ ($\ell = X_x, X_z$) valleys, and the second one is for the intervalley scattering among the X valleys. All the material constants necessary for calculating scattering rates have been taken from GaAs [75].

2.5.3 Electron-electron scattering

The electron-electron (e-e) scattering among the $\Gamma$-subbands has a significant effect [41] on QCL structures. The e-e scattering is included for the $\Gamma$ subbands, but not for the X subbands, since the electron densities in the X valleys are fairly low, except at high fields (above threshold). The e-e interaction may be divided into the binary e-e scattering and electron-plasmon coupling, with the latter being important in highly-doped systems and neglected in our calculation. The binary quasi-2D e-e scattering is treated by taking into account both the antiparallel-spin and parallel-spin e-e collisions, with the exchange effect [96] included for the parallel-spin collisions.

One can consider a “principal” $\Gamma$-electron in subband $\nu$, stage $\lambda$, with an in-plane wave vector $k$, and a “partner” $\Gamma$-electron in subband $\mu$, stage $\lambda_0$, with a wave vector $k_0$. Since the e-e scattering involves only the $\Gamma$ valley, the valley index will be omitted in the following. The final states of these two electrons are, respectively, $|k', \nu'\lambda'\rangle$ and $|k'_0, \mu'\lambda'_0\rangle$. The total electron scattering rate from the state $|k, \nu\lambda\rangle$ into a final state of subband $\nu'$, stage $\lambda'$, is given by [97]
\begin{equation}
\Lambda^{\nu'\lambda'}_{\nu\lambda}(k) = \frac{e_0^4 m^*_{\nu\lambda}}{32\pi \hbar^3 \varepsilon_{\infty} A} \sum_{k_0,\sigma} \sum_{\mu'\lambda_0} \sum_{\lambda_0} f_{\mu\lambda_0}(k_0) \times \int_0^{2\pi} d\theta \left[ \frac{|F(q)|^2}{q^2 \epsilon^2(q)} + \frac{|F(q')|^2}{q'^2 \epsilon^2(q')} - \frac{|F(q)||F(q')|}{q \epsilon(q) q' \epsilon(q')} \right],
\end{equation}
where $\sigma$ denotes the spin of electrons, $f_{\mu\lambda_0}(k_0)$ is the $\Gamma$-valley electron distribution, $\theta$ is the angle between $g$ ($g = k_0 - k$) and $g'$ ($g' = k'_0 - k'$), $q = |g - g'|/2$, and $\epsilon(q)$ is the static dielectric...
function; each of the three indices \((\lambda', \lambda_0, \lambda'_0)\) could be \(\lambda, \lambda + 1, \) or \(\lambda - 1.\) The form factor function \(F(q)\) (with indices omitted for compactness) satisfies

\[
F_{\Gamma_{\nu\mu'}}^{\lambda\lambda_0\lambda'_0}(q) = \sum_{i,j} \int_0^d \int_0^d \mathrm{d}z \int_0^d \mathrm{d}z' \psi_{i,\nu}(z) \psi_{i,\nu'}(z) \psi_{j,\mu}(z') \psi_{j,\mu'}(z') e^{-q|z-z'|},
\]

with \(i\) and \(j\) both running over the three bands (c, lh, so) at the zone-center. The values of \(q\) and \(q'\) are determined by

\[
q = \frac{1}{2} \left[ 2g^2 + g_0^2 - 2g \sqrt{g^2 + g_0^2 \cos \theta} \right]^{1/2},
\]

\[
q' = \frac{1}{2} \left[ 2g^2 + g_0^2 + 2g \sqrt{g^2 + g_0^2 \cos \theta} \right]^{1/2},
\]

where

\[
g = |g|, \quad g_0 = \frac{2}{\hbar} \left[ m_{\nu\lambda}^\star \left( E_{\nu\lambda}^\Gamma + E_{\mu\lambda_0}^\Gamma - E_{\mu'\lambda'}^\Gamma - E_{\mu'\lambda_0}^\Gamma \right) \right]^{1/2}.
\]

Figure 2.12 shows the form factors as a function of \(q\) for intrasubband and intersubband electron-electron scattering within the same stage \(\lambda,\) for \(\Gamma\)-subband wavefunctions of Fig. 2.5.

Analysis of the screening phenomena in multisubband systems, such as QCLs, is a formidable task. The effect of multisubband screening is treated within the static random-phase approximation (RPA) [98]. In the long-wavelength limit \(q \to 0,\) for intrasubband \((\nu = \nu', \lambda = \lambda', \mu = \mu', \lambda_0 = \lambda'_0)\) e-e transitions, the static dielectric function may be written as [74]

\[
\epsilon(q) = 1 + \sum_{\nu=1}^{N_\Gamma} f_{\nu\lambda}(0) \frac{q_{\nu\lambda}}{q}, \quad q_{\nu\lambda} = \frac{e_0^2 m_{\nu\lambda}^\star}{2 \pi \hbar^2 \epsilon_\infty},
\]

where \(f_{\nu\lambda}(0)\) is the occupancy of the bottom of the subband \(\nu\) in the central stage \(\lambda.\) The occupancy \(f_{\nu\lambda}(0)\) is evaluated at each time step in the EMC simulation, allowing for the screening function to be adjusted according to the actual electron distribution.

For intersubband e-e scattering, the effect of screening is expected to be much smaller than for the intrasubband case, since the dielectric function is weighted by the form factor functions within the RPA, and the intersubband form factors are much lower in magnitude and vanish for \(q = 0,\) as seen in Fig. 2.12, due to the orthogonality of the envelope functions. Therefore, to first-order, one can treat the intersubband e-e scattering as unscreened, which is also validated by Fig. 3 of
Figure 2.12  Selected form factors for intrasubband [(3, 3) → (3, 3), (3, 2) → (3, 2)] and intersubband [(3, 2) → (2, 3), (3, 2) → (2, 1)] electron-electron scattering within the same stage $\lambda$ for $\Gamma$-subband wavefunctions shown in Fig. 2.5, where $(\nu, \mu) \rightarrow (\nu', \mu')$ denotes that the principal electron in subband $\nu$ is scattered to subband $\nu'$ and the partner electron in subband $\mu$ is scattered to subband $\mu'$ due to the electron-electron scattering.
Ref. [44], where the unscreened intersubband Coulomb potential is very close to their screened potential.

The implementation of the e-e interaction in the EMC code follows the technique of Goodnick and Lugli [99]: precalculate the form factors as a function of q according to Eq. (2.37) (note that one can make use of the symmetry to reduce the computation), then compute the scattering rate \( \Lambda_{\nu'\lambda'}(k) \) in Eq. (2.36) by replacing the integrand with the square of the maximum value of \( F_{\nu\nu'\mu'\mu}(q) \) for given \( \nu, \lambda, \nu', \lambda' \), divided by \( q_{\nu\lambda}^2 \) given in (2.40); the actual value of the integrand function is accounted for through a rejection method when the final state is chosen. The scattering rate employed in the simulation is computed using half of the rate Eq. (2.36), since each electron from the simulated ensemble is scattered both as a principal electron and as a scattering partner [100]. In addition, it is necessary to update both the principal electron and its partner electron after the e-e collisions, in order to conserve the energy and momentum in the system [100].

2.6 Intersubband Dipole Matrix Element

One of the most important parameters that characterize the performance of a QCL is the intersubband dipole matrix element \( \langle z_{32} \rangle \), which represents the optical coupling strength between the upper lasing level 3 and the lower lasing level 2. The definition of \( \langle z_{ba} \rangle \) between any two subbands \( |b\rangle \) and \( |a\rangle \) is related to the intersubband dipole moment given by [73]

\[
\mathbf{u}_{ba} = \langle b|e(\hat{r}r + \hat{z}z)|a\rangle,
\]

(2.41)

where \( e \) is the electron charge, \( \hat{r} \) and \( \hat{z} \) are the unit vectors for the \( r \) (in plane) and \( z \) directions. If we consider two \( \Gamma \)-subbands in the \( \lambda \)th stage of a QCL, \( |k_b, \nu_b\lambda\Gamma\rangle \) and \( |k_a, \nu_a\lambda\Gamma\rangle \), and substitute Eq. (2.20a) into Eq. (2.41), we obtain

\[
\mathbf{u}_{ba} = \frac{e}{A} \left< e^{ik_b\cdot r} \sum_j \psi_{j,\nu_b\lambda}(z) u_{j0}^\Gamma(r,z) |(\hat{r}r + \hat{z}z)| e^{ik_a\cdot r} \sum_j \psi_{j,\nu_a\lambda}(z) u_{j0}^\Gamma(r,z) \right>.
\]

(2.42)

To simplify Eq. (2.42), we have to make use of the assumption that, on the length scale of an underlying crystal unit cell, only the Bloch functions, \( u_{j0}^\Gamma(r,z) \) and \( u_{j0}^\Gamma(r,z) \) vary, but any other functions in Eq. (2.42) remain constant. Then, after integrating both sides of Eq. (2.42) over a unit
cell and utilizing the orthonormality of the Bloch functions, we obtain

\[
\mathbf{u}_{ba} = \frac{e}{A} \sum_j \left( e^{i \mathbf{k}_b \cdot \mathbf{r}} \psi_{\Gamma_j,\nu_b,\lambda}(z) \right| \left( \mathbf{rr} + \hat{z}z \right) e^{i \mathbf{k}_a \cdot \mathbf{r}} \psi_{\Gamma_j,\nu_a,\lambda}(z) \right) .
\] (2.43)

Since the envelope functions of any two different \(\Gamma\)-subbands \((\nu_a \neq \nu_b)\) are mutually orthogonal, i.e.,

\[
\langle \psi_{\Gamma_j,\nu_b,\lambda}(z) | \psi_{\Gamma_j,\nu_a,\lambda}(z) \rangle = 0,
\] (2.44)

Eq. (2.43) can be simplified to

\[
\mathbf{u}_{ba} = \frac{e}{A} \left( e^{i \mathbf{k}_b \cdot \mathbf{r}} | e^{i \mathbf{k}_a \cdot \mathbf{r}} \right) \sum_j \langle \psi_{\Gamma_j,\nu_b,\lambda}(z) | \hat{z}z | \psi_{\Gamma_j,\nu_a,\lambda}(z) \rangle .
\] (2.45)

Since the plane waves are normalized as

\[
\langle e^{i \mathbf{k}_b \cdot \mathbf{r}} | e^{i \mathbf{k}_a \cdot \mathbf{r}} \rangle = \int d^2 \mathbf{r} e^{i (\mathbf{k}_a - \mathbf{k}_b) \cdot \mathbf{r}} = A \delta_{\mathbf{k}_a,\mathbf{k}_b},
\] (2.46)

the final expression for \(\mathbf{u}_{ba}\) has the following form

\[
\mathbf{u}_{ba} = e \delta_{\mathbf{k}_a,\mathbf{k}_b} \hat{z} \langle z_{ba} \rangle,
\] (2.47)

where the dipole matrix element is defined as

\[
\langle z_{ba} \rangle = \sum_j \langle \psi_{\Gamma_j,\nu_b,\lambda}(z) | \psi_{\Gamma_j,\nu_a,\lambda}(z) \rangle
\]

\[
= \sum_j \int_0^d z \psi_{\Gamma_j,\nu_b,\lambda}(z) \psi_{\Gamma_j,\nu_a,\lambda}(z) \, dz,
\] (2.48)

with \(j\) summing over the three (c, lh, so) bands. Note that \(\langle z_{ba} \rangle\) has the unit of length with ‘nm’ generally used in the QCL community. When designing a QCL, \(\langle z_{32} \rangle\) between the upper (3) and lower (2) lasing levels is a critical design parameter, and its value must be sufficiently large to achieve lasing.
Chapter 3

X-valley Leakage in GaAs/AlGaAs Quantum Cascade Lasers

In this chapter, the multivalley Monte Carlo simulator presented in Chapter 2 is applied to simulate two GaAs-based mid-infrared QCLs with equivalent designs: the GaAs/Al$_{0.33}$Ga$_{0.67}$As QCL of Refs. [29] and [30] (referred to as the 33% QCL), and the GaAs/Al$_{0.45}$Ga$_{0.55}$As QCL of Ref. [1] (referred to as the 45% QCL). Designs of these two QCLs are equivalent [1], since they have similar emitting wavelengths ($\sim$ 9.4 $\mu$m), threshold fields (48 kV/cm), dipole matrix elements (1.6 nm for the 33% QCL and 1.7 nm for the 45% QCL), and lifetimes in the upper lasing level (1.5 ps and 1.4 ps, respectively).

Figure 3.1 shows the $\Gamma$-subband energy levels and wavefunction moduli squared in two adjacent stages for the 33% QCL (Fig. 3.1a) and the 45% QCL (Fig. 3.1b) at the above-threshold field $F = 53$ kV/cm and the lattice temperature $T = 77$ K. For the 33% QCL [29], the thicknesses of the layers in one stage (in Å), starting from the injection barrier (the widest barrier in a stage), are 58, 15, 20, 49, 17, 40, 34, 32, 20, 28, 23, 23, 25, 25, 21. For the 45% QCL [1], the thicknesses of the layers in one stage (in Å), starting from the injection barrier (the widest barrier in a stage), are 46, 19, 11, 54, 11, 48, 28, 34, 17, 30, 18, 28, 20, 30, 26, 30. The bold script denotes the barriers, the normal script are the wells, and the underlined script denotes the n-type doped regions with a sheet density of $N_s = 3.9 \times 10^{11}$ cm$^{-2}$ for the 33% QCL, $N_s = 3.8 \times 10^{11}$ cm$^{-2}$ for the 45% QCL.

In each stage, ten $\Gamma$-subbands, including two $\Gamma$-continuum states ($\Gamma_c$), are used in the simulation of both lasers. In the 33% QCL, the upper lasing level and the injector localized states ($\Gamma_l$) are quite close to the $\Gamma$ band edge, and there is appreciable wavefunction overlap between $\Gamma_l$ and the next-stage $\Gamma_c$. On the other hand, in the 45% case, the $\Gamma$-point conduction band offset between the wells and the barriers increases by about 90 meV due to the higher Al content, so in order to achieve a
Figure 3.1 Γ-subband energy levels and wavefunction moduli squared in two adjacent stages for the 33% QCL (a) and the 45% QCL (b) at F = 53 kV/cm (above-threshold) and T = 77 K. The bold black lines denote the active lasing levels (3 - upper lasing level, 2 - lower lasing level, 1 - active ground level). The black thin lines are the localized injector states (Γ_l), while the top two (red and green) are the continuum states (Γ_c). The radiative transition occurs between levels 3 and 2.
similar emission wavelength (\(\sim 9.4 \mu\text{m}\)), the active lasing levels and the \(\Gamma_l\) levels are farther away from the \(\Gamma\) band edge than in the 33\% QCL. Consequently, the wavefunction overlap between \(\Gamma_l\) and next-stage \(\Gamma_c\) is negligible in the 45\% QCL.

The X-subbands under the same conditions are displayed in Fig. 3.2, with the \(\Gamma_c\) states also plotted, where Fig. 3.2a is for the 33\% QCL and Fig. 3.2b for the 45\% case. The number of X states in both stages is the same for each laser and chosen such that the highest X subband is right above the second \(\Gamma_c\) state and below other higher X levels. Since the X band edge is about 83 meV above the \(\Gamma\) band edge in the 33\% QCL, but \(\sim 30\) meV below in the 45\% QCL, more X subbands are needed to properly simulate the latter device. In particular, at \(F = 53 \text{kV/cm}\) and \(T = 77 \text{K}\), nine (ten) \(X_z\) subbands and four (six) \(X_x\) subbands are used in the 33\% (45\%) QCL simulation. It is clear from the figures that in both lasers, the \(\Gamma_c\) states are strongly coupled to the X states in the same stage.
Figure 3.2 X-subband energy levels and wavefunction moduli squared in two adjacent stages for the 33% QCL (a) and the 45% QCL (b) at $F = 53$ kV/cm and $T = 77$ K. The blue lines denote the doubly-degenerate $X_x$ states, and the yellow ones are the $X_z$ states, while the red and green lines are the $\Gamma_c$ subbands.
3.1 X-Valley Leakage Mechanism

The applied field vs. current density characteristics at the lattice temperatures of 77 K and 300 K are shown in Fig. 3.3 for the 33\% QCL (Fig. 3.3a) and the 45\% QCL (Fig. 3.3b), with and without the X-valley transport included. (Note that the leakage to \(\Gamma\)-continuum states [53] is accounted for as part of the \(\Gamma\)-valley current, so it is present in both curves.) The calculated threshold current densities (\(J_{th}\)) at the threshold field \(F = 48 \text{kV/cm}\) are summarized in Table 3.1, together with the available experimental data. It can be seen that the calculated \(J_{th}\) values are in very good agreement with the experimental data, considering that the simulation does not account for the losses at the lateral waveguide (ridge) edges. Subsequent experimental work [23] on 45\% QCLs, where the ridge edges had smooth surfaces due to wet chemical etching and no absorbing material, reported \(J_{th} (77 \text{K}) = 3 \text{kA/cm}^2\), in excellent agreement with 2.9 kA/cm\(^2\) obtained in our calculation.

![Figure 3.3](image_url)

Figure 3.3 Electric field vs current density characteristics for the 33\% QCL (a) and the 45\% QCL (b) at the lattice temperatures of 77 K and 300 K, with and without X-valley transport.

At the cryogenic temperature, the inclusion of X-valley transport has a very negligible effect on the current density up to above-threshold fields in both QCLs, but the 33\% QCL shows about 1 kA/cm\(^2\) higher \(J_{th}\) than the 45\% QCL in both theory and experiment. This difference is primarily
Table 3.1 Calculated threshold current densities $J_{th}$ with and without the X-valley transport.

<table>
<thead>
<tr>
<th></th>
<th>$J_{th}$ (kA/cm$^2$), 77 K</th>
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<th>$J_{th}$ (kA/cm$^2$), 300 K</th>
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<tr>
<td></td>
<td>w/o X</td>
<td>w/ X</td>
<td>Exper.</td>
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<tr>
<td>33% QCL</td>
<td>4.1</td>
<td>4.4</td>
<td>5.0$^1$</td>
</tr>
<tr>
<td>45% QCL</td>
<td>2.9</td>
<td>2.9</td>
<td>4.0$^2$</td>
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due to the strong interstage leakage from the injector localized states ($\Gamma_l$) (thin black curves in Fig. 3.4a) to the next-stage $\Gamma$-continuum states ($\Gamma_c$) (green curves in Fig. 3.4a) in the 33\% QCL. Ideally, all electrons in the $\Gamma_l$-states are expected to inject into the next-stage upper lasing level 3 (bold black curves in Fig. 3.4) for lasing transitions. In reality, however, the overlap between $\Gamma_l$ and the next-stage $\Gamma_c$ wavefunctions causes some electrons to leak into $\Gamma_c$, and these electrons do not contribute to the lasing process. In the 33\% QCL, this overlap is large, due to poor localization of the $\Gamma_l$-states (Fig. 3.4a), while in the 45\% case, the overlap is much smaller, thanks to the higher $\Gamma$-valley barriers, as seen in Fig. 3.4b. So it is the relatively large leakage from the $\Gamma_l$-states to the next-stage $\Gamma_c$-states that results in higher threshold current in the 33\% QCL. This mechanism of leakage to the $\Gamma_c$-states has been well recognized \[1, 23, 101\] and studied \[60\].

At room temperature, the simulated threshold current in the 33\% structure with the X-valley transport included is very high ($J_{th} = 25.5$ kA/cm$^2$), which can help explain why the 33\% QCL could not lase at 300 K. The increase in current density due to the inclusion of X-valley transport (X-valley leakage current) in the 33\% QCL is very large, even at fields significantly below threshold, equaling 8.3 kA/cm$^2$ at threshold. In contrast, the X-valley leakage current in the 45\% QCL is rather low in the whole simulated field range.

Figure 3.5 presents a variation of electron population in the $\Gamma$- and X-valleys with the field at different temperatures. By comparing the trends in Fig. 3.3 and Fig. 3.5 at 300 K, one can observe that both the X-valley leakage current and the X-valley electron population increase with the field in both lasers, which suggests that the increase in the X-valley leakage current follows the increase in the X-valley population (also true at 77 K, although somewhat less obvious). This is due to
Figure 3.4 Blow-up of the wavefunction moduli squared of the injector states ($\Gamma_l$) and the next-stage $\Gamma$-continuum ($\Gamma_c$) states in the 33% QCL (a) and the 45% QCL (b). Thin black curves denote the injector states, while the green ones are the next-stage $\Gamma_c$-states. The bold black curve 3 is the upper lasing level in the next stage, and the dotted black line indicates the $\Gamma$-valley band edge.
the fact that interstage X→X intervalley scattering is the dominant X-leakage mechanism in both QCLs, more efficient than direct interstage scattering between Γ and X. The dominant X-valley leakage takes place through the three-step mechanism sketched in Fig. 3.6. First of all, the coupling between the injector states (Γ₁) and the next-stage Γₑ states results in the leakage current $J_c$, indicated by the red horizontal arrows. Secondly, once those continuum states are populated, a portion of the electrons will efficiently scatter into the same-stage X valleys (provided there are enough intervalley phonons to enable the transition), indicated by the green vertical arrows, because the X states have larger effective masses and are strongly coupled to the same-stage Γₑ states in both QCLs, as seen from their large wavefunction overlap in Fig. 3.2. The inverse scattering is significantly less efficient, because of the Γ-valley lower mass, so carriers scattered into the X-valleys tend to remain there rather than go back to Γ. Thirdly, with enough population in the X-subbands and the fact that the plane-wave-like tails of the X wavefunctions in one stage couple to the X states in the next stage (also seen in Fig. 3.2), the X→X intervalley scattering between adjacent stages occurs, giving rise to the leakage current $J_X$, indicated by the blue arrows. Other
leakage paths, e.g., from $\Gamma_l$ to the next-stage X states, are less efficient. Therefore, a large overlap between $\Gamma_l$ and the next-stage $\Gamma_c$ causes not only leakage to the $\Gamma$-continuum, $J_c$, but also significant parallel X-to-X leakage, $J_X$.

![Diagram of leakage paths](image)

Figure 3.6 Schematic of the dominant X-valley leakage mechanism in both the 33% and 45% QCLs.

After identifying the dominant X-valley leakage mechanism, we are ready to explain why there is a large X-valley leakage current $J_X$ in the 33% QCL at room temperature even at low fields, but not in the 45% device. The reason is illustrated in Fig. 3.7. The value of $J_X$ relies on the X-valley population and the number of intervalley X-X phonons. In the 33% QCL, at room temperature, the leakage $J_c$ is very strong, due to both the large wavefunction overlap between $\Gamma_l$ and $\Gamma_c$ and the increase in the number of polar optical phonons, which leads to high $\Gamma_c$ population. At the same time, the number of intervalley $\Gamma$-to-X phonons increases. The combination of these two factors results in highly efficient scattering from $\Gamma_c$ to the same-stage X states (green arrows in Fig. 3.6), implying high X-valley population, since the scattering process efficiency depends on the number of phonons as well as the initial occupation. In addition, the number of intervalley (large phonon wave vector $q$) X-X phonons is also raised. Consequently, intervalley X-X scattering between
adjacent stages is greatly enhanced, producing a large $J_X$. In the 45% QCL, in contrast, $\Gamma_c$ does not get filled through $\Gamma_l \rightarrow \Gamma_c$ scattering because their overlap is very small (Fig. 3.4b), which explains the near-zero $J_X$ below threshold (Fig. 3.3b). Only at very high fields, $\Gamma_c$ gets filled by intrastage scattering (i.e., high fields lead to high electronic temperatures), which then augments the leakage current $J_X$ to some extent. The mechanism of X-valley leakage described above holds at 77 K as well, but leakage is much less pronounced because of few active phonons that enable intervalley scattering.

Figure 3.7 Block diagram illustrating the large X-valley leakage current at 300 K in the 33% QCL.

### 3.2 Effect of the Electron-Electron Interaction

The electron-electron (e-e) interaction plays a minor role in affecting the electron distribution for low electron sheet densities, and electrons relax their energy via a cascade of successive optical-phonon emissions ($\hbar\omega_{LO} \approx 36$ meV). However, for typical sheet densities in GaAs-based mid-IR QCLs (e.g., $N_s = 3.9 \times 10^{11}$ cm$^{-2}$), the e-e scattering has a significant effect on the steady-state electron distribution.
Figure 3.8 shows the time evolution of the electron density in six Γ-subbands at the above-threshold field $F = 53 \text{kV/cm}$ and $T = 77 \text{K}$, without the e-e interaction (a) and with the e-e interaction (b), for the 33% QCL. One can see the e-e interaction dramatically enhances the upper lasing level population, which was also observed in Ref. [41]. Inclusion of the e-e scattering strongly increases intersubband electron redistribution, thus reducing the electron accumulation in the lowest two injector levels (subbands inj. 1 and inj. 2) and optimizing the coupling between the active region and the injector.

Figure 3.9 displays the electron distributions in various Γ-subbands as a function of the in-plane electron kinetic energy at the above-threshold field $F = 53 \text{kV/cm}$ and $T = 77 \text{K}$, without the e-e interaction (a), and with the e-e interaction (b), for the 33% QCL with a sheet density of $3.9 \times 10^{11} \text{cm}^{-2}$. It is clear that the e-e scattering drives the distribution in each subband to a heated Maxwellian profile [i.e., $f(E_k) \sim e^{-E_k/k_BT_e}$]. This agrees with the findings by Iotti and Rossi [102]: for high electron densities, the e-e scattering is very effective in setting up a drifted Maxwellian distribution with a high electron temperature, and for typical operating conditions, electrons in QCLs thermalize within each subband. The slopes of the electron distributions in different subbands are fairly similar in Fig. 3.9b, indicating that the various subbands have an effective common electron temperature, as also found in Ref. [103]. The role of the e-e scattering is again crucial in setting up this behavior. As pointed out in Ref. [103], it can be mainly ascribed to the bi-intrasubband e-e scattering processes, i.e., two electrons in different subbands interact and stay in their own original subbands. These processes provide a very efficient way of redistributing excess kinetic energy among the electrons in order to achieve a common electron temperature. At high bias, such temperature is usually much higher than the lattice temperature. For example, in the case of Fig. 3.9b, the distribution function slope of the injector ground level (inj. 1) gives an approximate electron temperature of 221 K, much higher than the lattice temperature of 77 K. This estimated value of 221 K agrees well with the steady-state averaged electron temperature (244 K) obtained using $T_e = \langle E_k \rangle / k_B$ for a two-dimensional electron system, with $\langle E_k \rangle$ being the ensemble-averaged electron kinetic energy and $k_B$ the Boltzmann constant.
Figure 3.8 Time evolution of the electron density in six $\Gamma$-subbands at the above-threshold field $F = 53$ kV/cm and $T = 77$ K, without the e-e interaction (a) and with the e-e interaction (b), for the 33% QCL. In the legend of the figures, the first three indicate the electron densities in the lowest three injector states ($\Gamma_l$), while the remaining three are in the lasing levels (3 - upper lasing level, 2 - lower lasing level, 1 - active ground level) indicated in Fig. 3.1a.
Figure 3.9 Steady-state electron distributions in various Γ-subbands as a function of the in-plane kinetic energy at the above-threshold field $F = 53 \text{ kV/cm}$ and $T = 77 \text{ K}$, without the e-e interaction (a) and with the e-e interaction (b), for the 33% QCL. In the legend of the figures, the first three indicate that the distributions are in the lowest three injector states ($\Gamma_l$), while the remaining three are in the lasing levels (3 - upper lasing level, 2 - lower lasing level, 1 - active ground level) indicated in Fig. 3.1a.
The electron temperature is a direct measure of electron heating in QCL devices, and it is important to study how the electron temperature varies with the operating conditions. The elevated electron temperature is a clear fingerprint of a strong hot-electron regime: the electron system is not able to dissipate (via optical-phonon emission) the relatively large amount of energy provided by the applied bias. This electron heating in QCL devices has already been observed in experiment [61]. In Fig. 3.10, the electron temperature is plotted as a function of the current density for the 33\% (a) and 45\% (b) QCLs. It is clear that, in both QCLs, the temperature of the electron ensemble is much higher than the lattice temperature. At the low lattice temperature of 77 K, the electron temperature shows good linear relation with the current density in both lasers. The linearity can be described in the form of \( T_e = T_l + \alpha_{e-l} J \), where \( T \) is the lattice temperature, and \( \alpha_{e-l} \) is the electron temperature-current density coupling constant and can be obtained by least-square linear fits to the data. In the 33\% QCL, the coupling constant \( \alpha_{e-l} \) is calculated to be 14.0 K cm\(^2\)/kA for the case without X-valley transport, and 12.2 K/kA cm\(^{-2}\) for the case with X-valley transport. The latter is slightly smaller because the electrons lost to the X valleys have a heavier effective mass and thus a smaller kinetic energy. In the 45\% QCL, the coupling constants are identical for the two

![Figure 3.10 Electron temperature vs current density at the lattice temperatures of 77 K and 300 K, with and without X-valley transport, for the 33\% (a) and 45\% (b) QCLs. The magenta lines are least-square linear fits (\( T_e = T_l + \alpha_{e-l} J \)) to the data. The blue curves are guides for the eyes.](image-url)
cases (i.e., with and without X-valley transport) and equal to 14.4 K/kA cm$^{-2}$, because the electron loss to the X valleys is so small in the considered current density range that the heavier X-valley electrons are only a negligible portion of the whole electron ensemble. The linear dependence of the electron temperature on the current density was also obtained by Harrison et al. [104]. At the high lattice temperature of 300 K, the increase of the electron temperature with the current density becomes more non-linear in both QCLs. It is noted that at 300 K, in the 33% QCL, the difference in electron temperature between the two cases of with and without X-valley transport becomes more evident, but this difference is still negligible in the 45% case. The reason is that the 33% QCL has large carrier leakage to the X valleys at room temperature, as explained in Sec. 3.1, and once carriers leak into the X valleys, they become heavier due to the larger X-valley effective mass, so that the average carrier kinetic energy decreases, meaning a lower electron temperature.
Chapter 4

Design and Optimization of Deep-Active-Well GaAs-based 6.7 µm QCLs

The GaAs/Al$_{0.45}$Ga$_{0.55}$As quantum cascade laser by Page et al. [1], emitting at 9.4 µm, has shown the best device performance so far among GaAs-based mid-infrared QCLs. It can achieve pulsed room-temperature operation and operate in continuous mode up to 150 K [36]. As described in the previous section, the superior performance of this particular design is due to the relatively large (370 meV) Γ-point conduction band offset between the wells and barriers, which minimizes the leakage arising from the scattering of carriers from the injector states to the next-stage Γ-continuum states [23], and also indirectly results in very small leakage to the X valleys even at room temperature [57, 58].

To decrease the wavelength of GaAs/Al$_x$Ga$_{1-x}$As QCLs below 9 µm, one alternative is increasing the Al content in order to increase the Γ-point conduction band offset between the wells and the barriers. It is known that using 100% Al in the barriers (i.e., AlAs barriers) maximizes the conduction band discontinuity (∼1 eV). Wilson et al. [39] have shown that, in GaAs/AlAs QCLs incorporating a single injection barrier, once the upper lasing level becomes aligned with the lowest X-valley state of the injection barrier, lasing is suppressed due to intervalley (Γ-X) electron transfer, which limits the emission wavelengths to above 8 µm. Utilizing a double injection barrier [39], however, reduced the emission wavelength to 7.3 µm, with pulsed lasing observed only at cryogenic temperatures. In order to increase the operating temperature of GaAs/AlAs QCLs at these shorter wavelengths, Carder et al. [40] deposited InAs monolayers in the active quantum wells of a GaAs/AlAs QCL, and achieved room-temperature pulsed lasing at 8.5 µm. This technique of depositing InAs monolayers was also used in reducing the emission wavelength of the
GaAs/Al$_{0.33}$Ga$_{0.67}$As QCL from 9.4 $\mu$m [29] to 7.4 $\mu$m [26], and the lasing was observed at 15 K in pulsed mode.

This chapter presents the simulation and optimization of a deep-active-well GaAs QCL design that emits at 6.7 $\mu$m [64,65], the shortest room-temperature lasing wavelength projected to date for GaAs-based QCLs. This design utilizes compressively-strained In$_{0.1}$Ga$_{0.9}$As in the active wells, GaAs in the injector wells, Al$_{0.45}$Ga$_{0.55}$As in the barriers, and tensile-strained GaAs$_{0.6}$P$_{0.4}$ in a layer adjacent to the injection barrier. The multivalley Monte Carlo (MMC) simulator developed in Chapter 2 was employed to simulate the performance of three deep-active-well QCL structures (referred to as A, B, C), which are all designed to lase around 6.7 $\mu$m, but have different injector layers thicknesses. The unique inclusion of both $\Gamma$- and X-valley transport allows for identifying the issues associated with a particular structure. Simulation results from the first two structures A and B directly provide physical insights into obtaining an optimized injector layer sequence for both low- and room-temperature lasing operation. The optimized structure C was predicted to have threshold-current densities of 5 kA/cm$^2$ at 77 K and 14 kA/cm$^2$ at 300 K, similar to the experimental values obtained for the 9.4 $\mu$m GaAs QCL [1]. Furthermore, the electron temperature at 300 K lattice temperature is similar to that of the 9.4 $\mu$m device.

4.1 The Deep-Active-Well Design

The concept of utilizing deep wells in the active region (strained In$_x$Ga$_{1-x}$As active wells in GaAs-based devices) in order to significantly shorten the emission wavelength was demonstrated on single-stage In$_{0.3}$Ga$_{0.7}$As/GaAs/Al$_{0.7}$Ga$_{0.3}$As devices, which provided strong room-temperature electroluminescence at 4.7 $\mu$m [105] and were not strain compensated. A similar concept was applied to the not-strain-compensated, deep-active-well In$_{0.04}$Ga$_{0.96}$As/GaAs/Al$_{0.33}$Ga$_{0.67}$As devices [106], which resulted in a rather modest emission-wavelength decrease (i.e., from 10 $\mu$m [107] to 9.5 $\mu$m [106]) while the maximum operating temperature was only 200 K.

Our proposed laser structure employs the conventional three-well active region design [1], but utilizes strained In$_{0.1}$Ga$_{0.9}$As in the two wide active QWs to achieve a room-temperature emission wavelength below 7 $\mu$m, with the strain compensated in the injector of each stage. The calculated
The conduction band profile and the moduli squared of the relevant Γ- and X-valley wavefunctions in two adjacent stages for the optimized structure C are depicted in Fig. 4.1, where the Γ-states were solved for using the \textbf{k}·\textbf{p} method and the X-states were obtained within the effective mass framework, as described in Chapter 2. The layer sequence in one stage (in Å) of structure C starting from the GaAs$_{0.6}$P$_{0.4}$ barrier step on the left is 20, 32, 12, 11, 48, 11, 40, 28, 34, 15, 30, 16, 28, 18, 25, 20, 19. The bold italic script denotes the GaAs$_{0.6}$P$_{0.4}$ barrier step, the bold script the Al$_{0.45}$Ga$_{0.55}$As barriers, the normal script the GaAs wells, and the values in curly brackets indicate the In$_{0.1}$Ga$_{0.9}$As wells. The underlined layers are n-type doped with a sheet doping density of $N_s = 3.8 \times 10^{11}$ cm$^{-2}$. Structures A and B have the same doping density and active-region layers thicknesses as structure C, but with different injector layer sequence. The layer sequence of structure A is 20, 28, 12, 11, 48, 11, 40, 28, 38, 13, 30, 17, 17, and structure B has 20, 28, 12, 11, 48, 11, 40, 28, 38, 13, 30, 14, 30, 16, 27, 17, 25. As will be shown in Sec. 4.2, simulation results from structures A and B directly help to optimize the injector layer sequence of structure C.

To compensate the compressive strain in the In$_{0.1}$Ga$_{0.9}$As layers, one GaAs$_{0.6}$P$_{0.4}$ layer is added just before the injection barrier. The parallel component ($\epsilon_{xx}$, in the plane perpendicular to the growth direction) of the strain tensor for the compressively-strained In$_{0.1}$Ga$_{0.9}$As well is $\epsilon_w = -0.71\%$, and for the tensilely-strained GaAs$_{0.6}$P$_{0.4}$ barrier it is $\epsilon_b = +1.46\%$. The ratio of the thicknesses of the In$_{0.1}$Ga$_{0.9}$As layers to the GaAs$_{0.6}$P$_{0.4}$ barrier is designed such that the average in-plane lattice constant ($a_\parallel$) in one stage is nearly equal to that of the GaAs substrate (5.653 Å). $a_\parallel$ is calculated within the model solid theory [108], i.e., $a_\parallel = \sum_i a_i G_i d_i / \sum_i G_i d_i$, where $a_i$ and $d_i$ are the unstrained lattice constant and the thickness of the $i$th layer, respectively, and $G_i$ is the $i$th layer elastic shear modulus. For the optimized structure C, the calculated $a_\parallel$ equals to 5.657 Å, resulting in a net compressive strain of 0.07% in one stage.

The Γ-point conduction band offset between the strained In$_{0.1}$Ga$_{0.9}$As well and the Al$_{0.45}$Ga$_{0.55}$As barrier (the strain-induced band offset is included using the model-solid theory [108]) is 45 meV larger than that between the GaAs well and the Al$_{0.45}$Ga$_{0.55}$As barrier. The radiative transition energy (3→2) is 185 meV, about 54 meV larger than 131 meV for the 9.4 μm QCL of Page et
Figure 4.1 Calculated conduction band profile and the moduli squared of the relevant $\Gamma$- and $X$-valley wavefunctions in two adjacent stages for the optimized structure C. (a) The bold black lines denote the active lasing levels 3, 2, and 1. The thin black lines are the injector miniband states, and the bold green lines are the $\Gamma$-continuum states (levels 4 and 5). The vertical arrow denotes the lasing transition. (b) The thin blue lines are the (degenerate) $X_x$- and $X_y$-states, and the yellow ones are the $X_z$-states, together with the $\Gamma$-continuum states.
While the upper lasing level of our proposed structure is 20 meV closer to the $\Gamma$-band edge due to the thinner active-region wells, the remaining 34 meV increase in the transition energy is thanks to the deep wells in the active region. The thinner active wells also enable the lowest $\Gamma$-continuum level (level 4 in Fig. 4.1) to shift up by 15 meV, which helps to reduce the leakage to the continuum [58]. The calculated lifetime due to the electron-LO interaction for level 3 is $\tau_3 = 1.5$ ps, where $1/\tau_3 = 1/\tau_{32} + 1/\tau_{31}$, $\tau_{32} = 2.5$ ps, and $\tau_{31} = 3.8$ ps. The lifetime of level 2 is $\tau_2 \approx \tau_{21} = 0.3$ ps. The dipole matrix element $\langle z_{32} \rangle$ is calculated to be 1.5 nm. These calculations were done at $F = 55$ kV/cm, close to the estimated threshold field of $F = 58$ kV/cm, and $T = 77$ K. The lifetimes and matrix element are similar to those of the 9.4 $\mu$m QCL (though $\tau_3 = 1.4$ ps, $\tau_2 = 0.3$ ps, and $\langle z_{32} \rangle = 1.7$ nm) [1].

As mentioned earlier, the three structures (A, B, C) all have the same active-region layer sequence, designed to obtain the lifetimes of the lasing levels and the dipole matrix element similar to those of the state-of-the-art 9.4 $\mu$m GaAs QCL [1], but with a much shorter wavelength of 6.7 $\mu$m enabled by the use of deep active wells. Apart from designing the active region, it is essential to design and optimize the thicknesses of layers in the injector for desired device performance. The goal was to obtain an optimized injector layer sequence so that the resulting structure can achieve sufficient optical gain for lasing emission at both low and room temperatures. This is where the MMC simulator plays a crucial role. The injector of the initial structure A was modified from the 9.4 $\mu$m QCL. The MMC simulation of this structure clearly reveals that structure A is incapable of lasing action at 300 K. With an improved injector design, structure B shows better room-temperature performance, but sacrifices the gain at the low temperature of 77 K. Identification of issues associated with structures A and B directly helps to optimize the injector of structure C, which demonstrates adequate gain at both 77 K and 300 K.

### 4.2 Injector Optimization of Deep-Active-Well QCLs

This section will show how we can take advantage of the MMC simulator to optimize the injector layer sequence to obtain the desired structure C. The MMC simulation can theoretically characterize the output characteristics of GaAs QCL structures, namely, it yields the current density
$J$, the electron density in each subband, and the electron temperature $T_e$ for a given electric field. The modal gain $G_m$ can then be calculated from the population inversion (i.e., the electron density difference between the upper and lower lasing levels) and the waveguide confinement factor $\Gamma_w$. Based on the estimate of the waveguide losses $\alpha_w$ and mirror losses $\alpha_m$, a realistic threshold-current density can be determined.

The modal gain $G_m$ is proportional to the population inversion $\Delta n$ and is given under the steady-state conditions by [54]

$$G_m = \frac{4\pi e^2}{\varepsilon_0 n} \frac{\langle z_{32} \rangle^2}{2\gamma_{32} L_p \lambda} \Gamma_w \Delta n = g \Gamma_w J,$$

where $e$ is the electron charge, $\varepsilon_0$ is the vacuum dielectric permittivity, $n$ is the optical mode refractive index, $2\gamma_{32}$ is the full width at half maximum of the electroluminescent spectrum below threshold, $L_p$ is the length of one stage (active region + injector), $\lambda$ is the laser emission wavelength, $\langle z_{32} \rangle$ is the dipole matrix element, $\Gamma_w$ is the waveguide confinement factor, and $g$ is the local gain coefficient.

The waveguide for the deep-active-well 6.7 $\mu$m GaAs QCLs was designed by Mithun D’Souza, a graduate student of Prof. Dan Botez. It was designed such that a 25-stage QCL structure is sandwiched between two 1.3 $\mu$m-thick, low-doped ($4 \times 10^{16}$ cm$^{-3}$) GaAs layers. (Only 25 stages are expected to be grown considering that the residual strain may cause dislocations in the crystal lattice.) The GaAs layers are followed by 0.3 $\mu$m-thick Al$_{0.9}$Ga$_{0.1}$As cladding layers (doped to $1 \times 10^{18}$ cm$^{-3}$) and 1 $\mu$m-thick GaAs layers (doped to $4 \times 10^{16}$ cm$^{-3}$). On one side of the above layer sequence is a 1 $\mu$m-thick, highly doped ($5 \times 10^{18}$ cm$^{-3}$) GaAs contact layer, and on the other side is a 1 $\mu$m-thick, highly doped ($5 \times 10^{18}$ cm$^{-3}$) GaAs cladding layer and the GaAs substrate. The parameters of the designed waveguide were calculated to be $\Gamma_w = 33\%$, $n = 3.21$, and $\alpha_w = 15$ cm$^{-1}$. Even with fewer stages, the addition of Al$_{0.9}$Ga$_{0.1}$As cladding layers allows for a $\Gamma_w$ value higher than 28\%, the value used in the 36-stage 9.4 $\mu$m QCL [1]. The calculated waveguide losses are lower than those of the 9.4 $\mu$m QCL (20 cm$^{-1}$), due to a reduction in the free-carrier absorption with decreasing wavelength [109]. This same waveguide was used in the following three structures (A, B, and C).
4.2.1 Initial structure A

The design of initial structure A was done by Mithun D’Souza. Without the MMC simulation, it was unknown whether this structure has the desired threshold current density or optical modal gain, so it was indispensable to simulate the performance of structure A. Figure 4.2 shows the electric field vs current density characteristics for the QCL structure A. The computed $J$ values with the X-valley transport included account for the leakage currents through the next-stage $\Gamma_c$ states and the X-valley states [58]. At the low lattice temperature of 77 K, the current density is nearly the same for the two cases of with and without the X-valley, whereas the increase in the current density due to the X-valley leakage is dramatic at room temperature.

![Electric field vs current density characteristics at the lattice temperatures of 77 K and 300 K, with and without the X-valley transport included, for the QCL structure A. Dashed and solid curves are least-square polynomial fits to the calculated data, intended to guide the eyes.](image)

The $\Gamma$- and X-valley electron density for the case with the X-valley transport is shown in Fig. 4.3 as a function of electric field. At 77 K, the X-valley occupation is low up to very high fields, and the $\Gamma$- and X-valley electron densities cross at about 69 kV/cm. The large high-field X-valley population results from the enhanced scattering to the X山谷es, due to the elevated electronic temperature (Fig. 4.4). However, at 300 K, electron loss to the X valleys becomes significant even at low fields, and the $\Gamma$- and X-valley electron densities cross at a lower field of 59 kV/cm.
Furthermore, from Fig. 4.4, the X-valley electron density as a function of electron temperature follows the Maxwellian-Boltzmann profile, which was observed for all the QCL structures simulated in this dissertation.

![Graph of electron density vs. electric field](image)

Figure 4.3 Γ- and X-valley electron density as a function of electric field at the lattice temperatures of 77 K and 300 K.

The modal gain as a function of the current density is shown in Fig. 4.5, where the gain was calculated with $L_p = 39.4 \text{ nm}$, $\lambda = 6.7 \mu\text{m}$, $\langle z_{32} \rangle = 1.4 \text{ nm}$, $2\gamma_{32} = 12 \text{ meV}$ at 77 K, and $2\gamma_{32} = 22 \text{ meV}$ at 300 K ($2\gamma_{32}$ values were taken from Ref. [1]). In order to observe lasing in a QCL device, the modal gain must be higher than the total waveguide losses. Structure A shows very high gain at 77 K, much higher than the losses. However, at 300 K, although the gain obtained without the X valleys is above the loss line, in reality, carrier loss to the X valleys largely reduces the population inversion, as shown in Fig. 4.6, so that the actual gain with the X valleys saturates below the total waveguide losses. Overall, structure A has two problems, namely, large X-valley leakage current and insufficient modal gain at room temperature.

The X-valley leakage was shown to take place through the three-step process described in Chapter 3, and significant overlap between the injector miniband states and the next-stage $\Gamma_c$-states results in not only direct carrier loss to the $\Gamma_c$-states, but also indirect high current through the X-valley states by the three-step process. As can be seen from Fig. 4.7a, in structure A, the
Figure 4.4 Γ- and X-valley electron density as a function of electronic temperature \( (T_e) \) at the lattice temperatures of 77 K and 300 K. Solid curves are least-squared Maxwellian-Boltzmann fits.

Figure 4.5 Modal gain vs current density at the lattice temperatures of 77 K and 300 K, with and without the X-valley transport included, for the QCL structure A. Dashed and solid lines are least-square linear fits to the data, and the horizontal dash-dotted line indicates the calculated total losses \( (\alpha_w + \alpha_m = 19 \text{ cm}^{-1}) \) of the improved waveguide.
next-stage $\Gamma_c$-states (green) are indeed strongly coupled to the injector miniband (including level 2) (black), especially to the top two injector states. In fact, in the field range of 50-70 kV/cm, the two next-stage $\Gamma_c$-states become nearly resonant with the top two injector states; this leads to a large X-valley leakage current in that range of fields at 300 K, as seen in the $F$ vs $J$ characteristics (Fig. 4.2), and consequently to large carrier loss to the X valleys. Therefore, the strong coupling between the injector states and the next-stage $\Gamma_c$-states directly results in the large X-valley leakage and inadequate gain at room temperature in structure A.

### 4.2.2 Improved structure B

In order to reduce the coupling of the injector states with the $\Gamma_c$-states, the injector layer sequence of structure A needs to be modified. The first improved QCL design was done by Mithun D’Souza, referred to as structure B. In structure B, the thicknesses of three injector wells on the left of the injection barrier were increased by a few angstroms with respect to structure A, so that the injector top two levels are brought down in energy and their wavefunction maxima are shifted away from the continuum, as shown in Fig. 4.7b. By comparing Fig. 4.7b with Fig. 4.7a, one can
Figure 4.7 Zoom-in of the wavefunction moduli squared of the injector miniband and the next-stage $\Gamma$-continuum ($\Gamma_c$) states in the initial structure A (a) and the improved structure B (b). Thin black curves denote the injector states, while the green ones are the next-stage $\Gamma_c$-states. The lasing levels are shown in bold black, where 2 indicates the lower lasing level, 1 the active ground state, and 3 the upper lasing level in the next stage.
immediately see that structure B indeed shows smaller overlap between the injector and the $\Gamma_c$ wavefunctions than in structure A.

In Fig. 4.8, the electric field vs current density is compared for structures B and A at the lattice temperature of 300 K. The X-valley leakage current in structure B, i.e., the difference between the blue solid and dashed curves, does become smaller than in structure A, due to a decreased coupling of the injector states with the $\Gamma_c$-states. The modal gain vs current density at 300 K is shown in Fig. 4.9 for the two structures. The modal gain of structure B was calculated with $L_p = 40.8$ nm, $\lambda = 6.8 \mu$m, $\langle z_{32} \rangle = 1.6$ nm. The waveguide parameters are identical to those used for structure A. The dipole matrix element $\langle z_{32} \rangle$ in structure B is larger than in A [$\langle z_{32} \rangle = 1.4$ nm] thanks to better localization of the lasing levels, which further enhances the modal gain. Structure B indeed shows improved room-temperature gain compared to A, and the actual gain with the X-valley transport is just enough to overcome the total losses.

![Figure 4.8 Electric field vs current density characteristics at 300 K lattice temperature, with and without the X-valley transport included, for structures A and B. Dashed and solid curves are least-square polynomial fits to the calculated data, intended to guide the eyes.](image)

However, the low-temperature output characteristics of structure B is not as good as that of structure A. The electric field vs current density at 77 K is plotted in Fig. 4.10 for structures A and B. The current density in structure B is rather small even at fields above 70 kV/cm. The low current
Figure 4.9 Modal gain vs current density at 300 K lattice temperature, with and without the X-valley transport included, for structures A and B. Dashed and solid lines are least-square linear fits to the data, and the horizontal dash-dotted line indicates the calculated waveguide losses ($\alpha_w + \alpha_m = 19 \text{ cm}^{-1}$).

Figure 4.10 Electric field vs current density at 77 K lattice temperature, with and without the X-valley transport included, for structures A and B. Dashed and solid curves are least-square polynomial fits to the calculated data, intended to guide the eyes.
leads to a very low population inversion, resulting in insufficient modal gain ($< 15 \text{ cm}^{-1}$) in B at 77 K. The reason is that, in structure B, the next-stage upper lasing level (level 3 in Fig. 4.7b) is more weakly coupled to the injector ground state (level g) and far above in energy level (Fig. 4.7b), compared to structure A (Fig. 4.7a). At the low temperature of 77 K, the weak coupling and the large energy separation between levels 3 and g cause most electrons to stay in level g (injector ground state), e.g., the electron density in level 3 is only 3% of that in level g at $F = 70 \text{kV/cm}$, but this number is 78% in structure A. This electron accumulation in the injector ground state results in very poor injection efficiency. In contrast, at 300 K, the electrons are energetic and distribute among all the $\Gamma$-subbands due to active phonons, and thus the modal gain is still sufficient for lasing.

### 4.2.3 Optimized structure C

Identification of issues associated with structures A and B directly helps the optimization of structure C. The injector layer sequence of structure C was designed such that not only the coupling between the injector states and the next-stage $\Gamma_c$-states is reduced, but also the upper lasing level has a good wavefunction overlap and a small energy difference with the injector ground state, as seen in Fig. 4.11, to ensure adequate electron population in the upper lasing level. (The injector design of structure C was done by myself.) The X-valley leakage current ($\Delta J$), which is the difference between the current densities obtained with and without the X-valley transport, as a function of electric field, is shown in Fig. 4.12 at the lattice temperatures of 77 K and 300 K for structures A and C. The leakage current at 77 K is very similar in both structures, whereas structure C shows much smaller leakage current at 300 K than structure A, thanks to the reduced coupling of the injector states with the next-stage $\Gamma_c$-states.

The electric field vs current density behavior is shown in Fig. 4.13 at the lattice temperatures of 77 K and 300 K, with the X-valley transport, for the three structures A, B, and C. At a given field, the current density in structure C is lower than in structure A due to the reduced injector-$\Gamma_c$ coupling, but higher than in structure B due to the improved coupling between levels g and 3 in Fig. 4.11. The modal gain with the X-valley transport included is compared in Fig. 4.14 for the
Figure 4.11 Blow-up of the wavefunction moduli squared of the injector miniband and the next-stage $\Gamma$-continuum ($\Gamma_c$) states in the optimized structure C. Notation is the same as in Fig. 4.7.

Figure 4.12 X-valley leakage current vs electric field at the lattice temperatures of 77 K and 300 K for structures A and C. The leakage current $\Delta J$ is the difference between the current densities obtained with and without the X-valley transport included at a given field.
three structures A, B, and C. It is clear that structure C shows the highest modal gain among the three structures at 300 K and also excellent gain at 77 K. Similar to structures A and B, the modal gain of structure C at 300 K saturates in the high-\( J \) range (above 17 kA/cm\(^2\)) due to carrier loss to the X valleys \cite{59} that results in reduced population inversion. Nevertheless, the saturation gain (\( \approx 25 \) cm\(^{-1}\)) is sufficiently high with respect to the total losses of 19 cm\(^{-1}\) to deem room-temperature operation of the optimized structure C quite feasible.

### 4.3 Performance Comparison with the State-of-the-Art 9.4 \( \mu \)m QCL

In this section, the performance of the optimized 6.7 \( \mu \)m structure C is compared with that of the state-of-the-art 9.4 \( \mu \)m QCL \cite{1} (the 45\% QCL discussed in Chapter 3). Figure 4.15 presents the electric field vs current density behavior for the optimized 6.7 \( \mu \)m QCL structure C (a) and the 9.4 \( \mu \)m QCL of Ref. \cite{1} (b), at the lattice temperatures of 77 K and 300 K, with and without the X-valley leakage. At both 77 K and 300 K, the X-valley leakage current in the 6.7 \( \mu \)m structure C is higher than in the 9.4 \( \mu \)m QCL. (At 77 K, the leakage in both structures is fairly low, predominantly due to a low number of intervalley phonons.) As seen in Fig. 4.16, the reason is the difference in the
wavefunction overlap between the injector miniband (black) and the next-stage $\Gamma$-continuum states (green): the structure of Page et al. has a remarkably low overlap (Fig. 4.16b) between the injector miniband and the next-stage $\Gamma_c$-states; in the 6.7 $\mu$m structure C, however, this overlap is greater (Fig. 4.16a) due to the wavelength reduction, which results in a higher X-valley leakage current for a given electric field, especially at 300 K, through the X-leakage mechanism presented in Chapter 3. However, even with more X-valley leakage, the total current at a given field is significantly lower in the 6.7 $\mu$m structure C than in the 9.4 $\mu$m QCL, owing to the design modification necessary for the wavelength shortening.

Furthermore, the 6.7 $\mu$m structure C demonstrates gain coefficients and threshold current densities similar to those of the 9.4 $\mu$m QCL. The gain coefficient $g$ in Eq. (4.1) can be obtained for the structure C: $g = 11.8$ cm/kA at 77 K and $g = 7.6$ cm/kA at 300 K, values close to the calculations by Indjin et al. [60] for the 9.4 $\mu$m QCL ($g = 11$ cm/kA at 77 K and $g = 5$ cm/kA at 300 K). Given the calculated total waveguide losses ($\alpha_m + \alpha_w = 19$ cm$^{-1}$, $\alpha_m = 4$ cm$^{-1}$), since the threshold field was unknown for the structure C, the threshold-current density $J_{th}$ can be defined at which the linear fit of optical modal gain intersects with the loss line in the $G_m$ vs $J$ relation. From Fig. 4.14, $J_{th}$ is found to be 5 kA/cm$^2$ at 77 K and 14 kA/cm$^2$ at 300 K. (Structure B has
Figure 4.15 Electric field vs current density for the optimized 6.7 $\mu$m structure C (a) and the 9.4 $\mu$m QCL from Ref. [1] (b), at the lattice temperatures of 77 K and 300 K, with and without the X-valley transport included. Dashed and solid curves are polynomial fits to the data points.
Figure 4.16 Zoom-in of the wavefunction moduli squared of the injector miniband and the next-stage $\Gamma$-continuum ($\Gamma_c$) states in the optimized 6.7 $\mu$m structure C (a) and the 9.4 $\mu$m QCL from Ref. [1] (b), at the field of 63 kV/cm (above threshold for both structures). Thin black curves denote the injector states, while the green ones are the next-stage $\Gamma_c$-states. The lasing levels are shown in bold black, where 2 indicates the lower lasing level, 1 the active ground state, and 3 the upper lasing level in the next stage.
$J_{th} = 9.5$ kA/cm$^2$ at 300 K due to the smaller currents in Fig. 4.13 and higher $\langle z_{32} \rangle = 1.6$ nm.)

For the 9.4 $\mu$m QCL, the calculated $J_{th}$ has the values of 2.9 kA/cm$^2$ at 77 K and 14.4 kA/cm$^2$ at 300 K, which are listed in Table 3.1, while the experimental values are 4 kA/cm$^2$ (77 K) and 16.7 kA/cm$^2$ (300 K), respectively. By comparison, it is clear that the threshold-current densities of the optimized 6.7 $\mu$m structure C are comparable to those of the 9.4 $\mu$m QCL.

The electron temperature $T_e$, generally higher than the lattice temperature, characterizes the degree of electron heating in a QCL device. With relatively high carrier densities per subband, distribution function within each subband $i$ becomes Maxwellian [$f(E_{k,i}) \sim \exp(-E_{k,i}/k_B T_e)$] as a result of efficient carrier-carrier intrasubband scattering (intersubband scattering, both electron-electron and electron-phonon, are less efficient). The electronic temperature $T_e$ is approximately the same for all subbands [102, 103]. In the Monte Carlo simulation, the electron temperature can be obtained either from the thermalized distribution function or from the ensemble average of the carrier kinetic energy $T_e = \langle E_k \rangle / k_B$. These two approaches are equivalent as long as the distribution is Maxwellian, as discussed in Sec. 3.2, and in this work the latter approach was used.

Figure 4.17 shows the electron temperature $T_e$ as a function of the current density $J$ (a) and as a function of the input electrical power density $P_e$ (b), for the optimized 6.7 $\mu$m structure C and the 9.4 $\mu$m QCL [1], at the lattice temperature of 300 K. It is clear that the reduction in the room-temperature emission wavelength, provided by the proposed deep-active-well design, is achieved at no penalty in the electron heating: at threshold, the electron temperatures are $T_e = 530$ K ($J_{th} = 14$ kA/cm$^2$) for the 6.7 $\mu$m structure C, and $T_e = 544$ K ($J_{th} = 16.7$ kA/cm$^2$) for the 9.4 $\mu$m QCL. (We find very little influence of the X-valley transport on the electron temperature up to very high electric fields, and therefore do not separate the curves with and without the X-valley transport. The reason is that the X-valley population is generally low and these X-subbands have low kinetic energy due to their larger effective mass up to high electric fields, as discussed in Sec. 3.2.)

In both structures, the variation of the electronic temperature with the current density is linear only for very low current densities (Fig. 4.17a), with the low-current electron-lattice coupling constant being $\lim_{J \to 0} \frac{T_e - T_L}{J} \approx 14$ K cm$^2$/kA for both devices. However, the variation of $T_e$ with the power density ($P_e = JF$) obeys a quasi-linear law, even up to high power densities (Fig.
Figure 4.17 Electron temperature $T_e$ vs current density $J$ (a) and vs input electrical power density $P_e (= JF)$ (b), for the optimized 6.7 $\mu$m structure C and the 9.4 $\mu$m QCL, at the lattice temperature of 300 K. The lines are polynomial fits, intended to guide the eye.

4.17b). This agrees with a simple energy-balance equation

$$nk_B \frac{dT_e - T_L}{dt} = JF - nk_B \frac{(T_e - T_L)}{\tau_E},$$

where $n$ is the total carrier density, and $\tau_E$ is an effective (ensemble averaged) energy relaxation time [75]. In the steady state, the above equation yields

$$T_e - T_L = \frac{\tau_E}{nk_B} JF,$$

which is a simple, intuitive description of the linear behavior that we observe in the simulation (Fig. 4.17b).

For the structure of Page et al., the University of Bari group recently reported [61] a projected electronic temperature of about 800 K at threshold, with the heatsink temperature of 243 K and an estimated lattice temperature of 300 K. The temperature they reported is about 250 K higher than what we obtain for the same lattice temperature and at threshold (Fig. 4.17a). There are several reasons for the discrepancy. First, we perform all the simulations at a given lattice temperature, rather than at a given heatsink temperature, because a specified lattice temperature is necessary for
the calculation of the scattering rates that enter our simulator. Therefore, we do not account for the thermal resistance \([11, 34, 110]\), which is a non-trivial thing to model in superlattices. Accounting for confined bulk-like and interface phonons, as well as for the nonequilibrium phonon distribution at high fields, is important for the computation of the thermal resistance, but would also alter the scattering rates and further change the output of the electronic-transport simulation. Secondly, our calculation accounts for the transport through the laser’s active region alone, and thus no effect of the actual waveguide is incorporated in the simulation. Therefore, it is not trivial to perform a direct comparison of our simulation results with the experimental data of Spagnolo et al \([61]\).

In addition to the device optimization presented in this chapter, the MMC simulator is also used to simulate and optimize two GaAs-based 7.9 μm QCLs. Details of the 7.9 μm QCL structures and their simulations are presented in Appendix A.
Chapter 5

Phonon Confinement in GaAs QCLs

In the chapters presented above, the electron-LO phonon scattering rates were calculated using the bulk GaAs phonon approximation. In general, transport simulations [43, 44, 48, 52, 59, 111] of QCLs assume electrons interacting with dispersionless bulk phonons of the well material, due to the simplicity of the scattering rate calculation under this approximation. However, it has been unclear whether the spatial confinement of phonons has a strong influence on the electron-LO phonon scattering strength in cascaded structures: on the one hand, Rücker et al. [94, 112] showed that phonon confinement has an insignificant effect on the scattering rate in single-well GaAs/AlGaAs structures, but is critical for interpreting the signals of time-resolved Raman spectroscopy [113]. Williams and Hu [114] also showed that the total phonon scattering rates including confinement in one stage (three quantum wells) of two GaAs/Al$_{0.3}$Ga$_{0.7}$As THz QCLs are very close to those obtained using GaAs bulk phonons, thanks to wide wells and thin, low-Al barriers. On the other hand, calculations by Menon et al. [115] indicated that the scattering rate $\tau_{21}$ (levels 2 to 1) in a similar three-QW THz structure can be strongly enhanced due to phonon confinement, and Ref. [116] reported that, in step quantum-well structures using GaAs/Al$_{0.25}$Ga$_{0.75}$As/Al$_{0.4}$Ga$_{0.6}$As materials, the scattering rates with phonon quantization are more than an order of magnitude greater than the bulk GaAs rate.

Furthermore, very little work has been done on incorporating phonon confinement in QCL transport calculations (beyond the scattering rate computation): notable work in this direction came from the University of Rome group [42, 117, 118], who focused on the electroluminescence spectra calculation in InGaAs/InAlAs superlattice QCLs (each period containing one barrier and one well) lattice-matched to InP substrates. The same group also reported calculation of the electron-LO
phonon scattering rates with phonon quantization [119] for a GaAs/Al$_{0.45}$Ga$_{0.55}$As 6.9 $\mu$m QCL structure consisting of triple-QW active regions, designed by Strasser et al. [28]. However, so far there has been no systematic account of the phonon confinement on the transport properties of GaAs-based QCLs, such as the current-field characteristics or population inversion.

In this chapter, we provide a detailed account of the effects that phonon confinement has on the electron transport properties of mid-infrared, multiple-quantum-well, GaAs-based QCLs [66, 67]. The phonon quantization due to spatial confinement is treated within the macroscopic dielectric continuum model [120], where the LO phonons are classified as confined (CF) bulk-like modes and interface (IF) modes. The IF phonon spectra are obtained by using the transfer matrix method [121] with periodic boundary conditions [66]. The calculated CF and IF potentials are then utilized to evaluate both $\Gamma$- and X-valley electron transition rates due to the interaction with these phonon modes. Intravalley electron-IF, electron-CF, and electron-electron scattering, as well as intervalley scattering, are implemented in the multivalley Monte Carlo simulation of the deep-active-well optimized 6.7 $\mu$m structure C presented in Chapter 4.

## 5.1 Phonon Confinement Modeling

There are four macroscopic models that can treat the interaction of confined electrons with confined polar-optical phonons in III-V heterostructures: the dielectric continuum (DC) model [120, 122], the hydrodynamic (HD) model [123], the reformulated (RM) DC model [124, 125], and the hybrid model [126–128]. It was shown by Nash [127] that the first three models all produce the same scattering rates in a single GaAs/AlAs quantum well in the limit of dispersionless bulk phonons, provided that the modes in each model are mutually orthogonal and constitute a complete set. Constantinou and Ridley [128] found that the total scattering rates obtained by the hybrid model are reproduced to an excellent degree by the DC model, and are virtually insensitive to the bulk dispersion. Moreover, Rücker et al. [94] and Insook et al. [129] showed that the DC model accurately predicts the total scattering rates obtained by microscopic calculations. In view of the accuracy of the DC model in the scattering rate calculation and the relative simplicity of
its implementation, the DC model has been widely used \([114–116, 119, 121, 130]\) to treat phonon confinement in multiple-quantum-well (MQW) structures.

### 5.1.1 Phonon dispersions

Within the DC model, the optical phonon modes are described by the electrostatic potential \(\Phi(\mathbf{r}, z)\) (\(\mathbf{r}\) is the in-plane position vector, \(z\) the growth direction) resulting from the polarization field created by atomic displacements in a polar semiconductor. In regions devoid of free charges, the phonon potential \(\Phi(\mathbf{r}, z)\) satisfies the Poisson equation \(\varepsilon(\omega) \nabla^2 \Phi(\mathbf{r}, z) = 0\), where \(\varepsilon(\omega)\) is the dielectric function.

Using the generalized Lyddane-Sachs-Teller relation, \(\varepsilon(\omega)\) takes the form of a single-pole function,

\[
\varepsilon(\omega) = \varepsilon_\infty \frac{\omega^2 - \omega_{\text{LO}}^2}{\omega^2 - \omega_{\text{TO}}^2},
\]

for a binary alloy, and obeys a double-pole relation \([131]\),

\[
\varepsilon(\omega) = \varepsilon_\infty \left( \frac{\omega^2 - \omega_{\text{LO}1}^2}{\omega^2 - \omega_{\text{TO}1}^2} \right) \left( \frac{\omega^2 - \omega_{\text{LO}2}^2}{\omega^2 - \omega_{\text{TO}2}^2} \right),
\]

for a ternary alloy, where \(\text{LO}\) (\(\text{TO}\)) stands for the longitudinal (transverse) optical mode, and 1 and 2 denote the material type. \(\varepsilon_\infty\) is the high-frequency dielectric constant of the alloy, found as a linear combination of the constituents’ constants. The dielectric constants and phonon frequencies of the alloys used in the 6.7 \(\mu\)m QCL \([64]\) are listed in Tables 5.1-5.2, where for ternary alloys \(A_xB_{1-x}C\), \(\hbar\omega_{\text{LO}1}/(\hbar\omega_{\text{TO}1})\) is the BC-like LO (TO) frequency, and \(\hbar\omega_{\text{LO}2}/(\hbar\omega_{\text{TO}2})\) is the AC-like LO (TO) frequency. The strain-induced frequency shifts in GaAs\(_{1-x}\)P\(_x\) and In\(_x\)Ga\(_{1-x}\)As grown on GaAs substrates are less than 1 meV \([132–134]\), so they were not included here.

Since phonons are unconfined in the \(x\)-\(y\) plane in a multiple quantum well system, the potential \(\Phi(\mathbf{r}, z)\) can be written as

\[
\Phi(\mathbf{r}, z) = \sum_{\mathbf{q}} f(q) \phi(q, z) e^{i\mathbf{q} \cdot \mathbf{r}},
\]

where \(\mathbf{q}\) and \(\mathbf{r}\) are the in-plane phonon wave and position vectors, \(q = |\mathbf{q}|\), \(\phi(q, z)\) is the so-called functional form, and \(f(q)\) is the normalization coefficient (calculation of \(f(q)\) can be found in Sec. 5.1.2). Substituting Eq. (5.3) into the Poisson equation, we obtain the equation that \(\phi(q, z)\) must
Table 5.1 Dielectric constants and optical phonon frequencies for the binary alloys used in the optimized 6.7 \( \mu \)m QCL structure C.

<table>
<thead>
<tr>
<th></th>
<th>GaAs</th>
<th>AlAs</th>
<th>InAs</th>
<th>GaP</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \varepsilon_\infty )</td>
<td>10.89</td>
<td>8.16</td>
<td>12.3</td>
<td>9.11</td>
</tr>
<tr>
<td>( \hbar \omega_{LO1} ) (meV)</td>
<td>36.25</td>
<td>50.09</td>
<td>29.79</td>
<td>50.19</td>
</tr>
<tr>
<td>( \hbar \omega_{TO1} ) (meV)</td>
<td>33.29</td>
<td>44.88</td>
<td>27.18</td>
<td>45.57</td>
</tr>
</tbody>
</table>

Table 5.2 Dielectric constants and optical phonon frequencies for the ternary alloys used in the optimized 6.7 \( \mu \)m QCL structure C.

<table>
<thead>
<tr>
<th></th>
<th>Al(<em>x)Ga(</em>{1-x})As [135]</th>
<th>In(<em>x)Ga(</em>{1-x})As [136]</th>
<th>GaAs(_{1-x})P(_x) [132, 137]</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \varepsilon_\infty )</td>
<td>10.89-2.73( x )</td>
<td>10.89+1.41( x )</td>
<td>10.89-1.78( x )</td>
</tr>
<tr>
<td>( \hbar \omega_{LO1} ) (meV)</td>
<td>36.22-4.67( x )-0.27( x )^2</td>
<td>36.32-3.75( x )-2.27( x )^2</td>
<td>36.17-3.17( x )</td>
</tr>
<tr>
<td>( \hbar \omega_{TO1} ) (meV)</td>
<td>33.16+0.58( x )-2.46( x )^2</td>
<td>33.5-3.64( x )</td>
<td>33.28+0.8( x )</td>
</tr>
<tr>
<td>( \hbar \omega_{LO2} ) (meV)</td>
<td>45.09+6.66( x )-1.72( x )^2</td>
<td>29.66-2.1( x )+2.08( x )^2</td>
<td>43.5+8.97( x )-2.49( x )^2</td>
</tr>
<tr>
<td>( \hbar \omega_{TO2} ) (meV)</td>
<td>44.93-1.19( x )+1.18( x )^2</td>
<td>29.53-2.45( x )</td>
<td>41.9+3.4( x )</td>
</tr>
</tbody>
</table>

\( \varepsilon(\omega) \left( \frac{\partial^2}{\partial z^2} - q^2 \right) \phi(q, z) = 0. \quad (5.4) \)

Eq. (5.4) has two types of solutions: interface (IF) modes, for which \( [(\partial^2/\partial z^2) - q^2] \phi(q, z) = 0 \) and \( \varepsilon(\omega) \neq 0 \), and confined (CF) modes, for which \( \varepsilon(\omega) = 0 \).

For interface (IF) modes, \( \varepsilon(\omega) \neq 0 \) and the modes should have frequencies \( \omega(q) \neq \omega_{LO} \). In region \( R_i \) of Fig. 5.1 \((i = 1, 2, ..., N) \), where \( N \) is the number of layers in one stage), \( \phi_i(q, z) \) is given by

\[ \phi_i(q, z) = c_{i,+}e^{q(z-z_i)} + c_{i,-}e^{-q(z-z_i)}. \quad (5.5) \]

In the DC model, the potential \( \phi_i(q, z) \) and the tangential component of the electric field \( \varepsilon_i(\partial/\partial z)\phi_i(q, z) \) (where \( \varepsilon_i = \varepsilon_i(\omega) \), with \( \omega \) omitted for brevity) must be continuous at each interface. It then follows
Figure 5.1 Schematic of a MQW QCL heterostructure. The $z$ axis is chosen as the growth direction.

that, at the interface between regions $R_{i+1}$ and $R_i$ (at $z = z_{i+1}$),

$$
\phi_{i+1}(q, z_{i+1}) = \phi_i(q, z_{i+1}),
$$

(5.6)

$$
\varepsilon_{i+1} \frac{\partial}{\partial z} \phi_{i+1}(q, z_{i+1}) = \varepsilon_i \frac{\partial}{\partial z} \phi_i(q, z_{i+1}).
$$

(5.7)

After some manipulations, we obtain the following compact matrix-form equation,

$$
\begin{bmatrix}
  c_{i+1,+} \\
  c_{i+1,-}
\end{bmatrix} = Q_i(d_i) \begin{bmatrix}
  c_{i,+} \\
  c_{i,-}
\end{bmatrix},
$$

(5.8)

where $d_i$ denotes the thickness of the $i$th layer, and the transfer matrix $Q_i(d_i)$ is defined as

$$
Q_i(d_i) = \frac{1}{2} \begin{bmatrix}
  (1 + \frac{\varepsilon_i}{\varepsilon_{i+1}}) e^{qd_i} & (1 - \frac{\varepsilon_i}{\varepsilon_{i+1}}) e^{-qd_i} \\
  (1 - \frac{\varepsilon_i}{\varepsilon_{i+1}}) e^{qd_i} & (1 + \frac{\varepsilon_i}{\varepsilon_{i+1}}) e^{-qd_i}
\end{bmatrix}.
$$

(5.9)

By applying the chain rule, the coefficients in the last ($N$th) and the first layer of the same stage are related by

$$
\begin{bmatrix}
  c_{N,+} \\
  c_{N,-}
\end{bmatrix} = Q_{N-1}(d_{N-1}) ... Q_i(d_i) ... Q_1(d_1) \begin{bmatrix}
  c_{1,+} \\
  c_{1,-}
\end{bmatrix}.
$$

(5.10)

Next, by imposing the electrostatic boundary conditions at the interface between two adjacent stages, i.e., at $z = z_1$, we obtain

$$
\begin{bmatrix}
  c_{1,+} \\
  c_{1,-}
\end{bmatrix} = Q_{N'}(d_{N'}) \begin{bmatrix}
  c_{N'+,+} \\
  c_{N'-,}
\end{bmatrix},
$$

(5.11)
with \( Q_{N'} \) defined as
\[
Q_{N'}(d_{N'}) = \frac{1}{2} \begin{pmatrix}
(1 + \frac{\varepsilon_{N'}}{\varepsilon_1}) e^{qd_{N'}} & (1 - \frac{\varepsilon_{N'}}{\varepsilon_1}) e^{-qd_{N'}} \\
(1 - \frac{\varepsilon_{N'}}{\varepsilon_1}) e^{qd_{N'}} & (1 + \frac{\varepsilon_{N'}}{\varepsilon_1}) e^{-qd_{N'}}
\end{pmatrix}.
\] (5.12)

To obtain the IF dispersions in a MQW QCL, the isolated-stage boundary conditions (BCs) are generally assumed [114, 115, 121], namely, the IF potentials purely decay in the \( N' \)th and \( N \)th layers of Fig. 5.1, i.e., \( c_{N,+} = 0 \) and \( c_{N',-} = 0 \). However, this assumption leads to a discontinuity in the phonon potentials at the interfaces between adjacent stages [138], which violates the BCs of the DC model. In order to properly account for the periodicity of QCL structures, periodic BCs [117] are used here in obtaining the IF modes, namely, \( c_{N',+} = c_{N,+} \) and \( c_{N',-} = c_{N,-} \). It is also evident that \( Q_{N'}(d_{N'}) = Q_{N}(d_N) \). Then we end up with the following eigenvalue equation,
\[
(M - I) \begin{pmatrix}
c_{1,+} \\
c_{1,-}
\end{pmatrix} = 0,
\] (5.13)
where \( I \) is the identity matrix, and
\[
M = Q_N(d_N)Q_{N-1}(d_{N-1})...Q_1(d_1)...Q_1(d_1).
\] (5.14)
For Eq. (5.13) to have a nontrivial solution, \( \det(M - I) = 0 \). Since \( M = M(q, \omega) \), the solutions to \( \det(M - I) = 0 \) yield the dispersion relations of IF modes. The number of IF modes is determined by the number of interfaces in one stage and the constituents of each interface. Each binary/binary interface contributes with 2 IF modes, while the number of contributed modes per interface is 3 for a binary/ternary (each ternary has two LO-like frequencies), and 4 for a ternary/ternary interface.

The IF dispersions for the 6.7 \( \mu m \) QCL are shown in Fig. 5.2. One stage of this structure has 17 interfaces, among which 5 are ternary/ternary and 12 are ternary/binary, giving rise to a total of 56 IF modes. The mode frequencies at each \( q \) are obtained by numerically solving \( \det(M(q, \omega) - I) = 0 \). Due to the many interfaces and the multiplication in Eq. (5.14), \( \det(M(q, \omega) - I) \) rapidly oscillates as a function of \( \omega \), making it increasingly difficult to resolve between consecutive zeroes for larger chosen \( q \)'s. Therefore, for the 6.7 \( \mu m \) QCL considered, we explicitly calculate the frequencies up to \( q_{lim} = 0.25 \, \text{Å}^{-1} \), while at higher \( q \)'s we simply extrapolate \( \omega(q > q_{lim}) = \omega(q_{lim}) \) for each mode obtained.
Figure 5.2 Dispersion relations of the 56 IF modes for the 6.7 µm QCL. Bottom to top, the dispersions include 4 InAs-like modes around 29.4 meV, 17 GaAs-like modes within 32.8-33.6 meV, a second set of 17 GaAs-like modes within 34-36.4 meV, and 18 modes (2 GaP-like and 16 AlAs-like) within 43-48 meV.
Once the IF dispersions have been obtained, the potential $\phi_i(q,z)$ in region $R_i$ for each IF mode, can be deduced by setting $c_{1,+} = 1$. [The real value of $c_{1,+}$ will be taken care of by the normalization coefficient $f(q)$ (Sec. 5.1.2).] The IF potential in one stage consists of the different layer $\phi_i(q,z)$ in that stage, i.e., $\phi_i(q,z) = \phi_i(q,z)$, $z \in R_i$. Then the IF potential in the entire computational domain (ECD) (two or more stages), $\phi(q,z)$, is obtained by repeating the one-stage potential, $\phi_i(q,z)$, with the proper shift in the $z$-axis.

For confined (CF) bulk-like modes, $\varepsilon(\omega) = 0$, so $\omega = \omega_{LO}$ for a binary alloy, and $\omega = \omega_{LO1}$ or $\omega_{LO2}$ for a ternary alloy. The LO phonons propagate in a chosen layer to the interfaces, and are fully backscattered from them because the neighboring layers have different LO frequencies; the incident and backscattered waves interfere and result in zero potential at the interfaces [139, 140]. Therefore, in each layer $i$ ($i = 1, 2, \cdots, N_{tot}$, where $N_{tot}$ is the total number of layers in the ECD), the following functional form of the CF potential holds:

$$\phi_i(q,z) = \sin \left[ \frac{m\pi}{d_i} (z - z_i) \right], \quad m = 1, 2, \cdots, (5.15)$$

where $z_i \leq z < z_{i+1}$ and $d_i = z_{i+1} - z_i$ is the layer thickness. In the case of the 6.7 $\mu$m QCL, there are 7 CF LO modes: InAs-like from InGaAs (29.47 meV), GaP-like from GaAsP (46.69 meV), AlAs-like from AlGaAs (47.74 meV), GaAs (36.25 meV), GaAs-like from InGaAs (35.92 meV), GaAs-like from AlGaAs (34.06 meV), and GaAs-like from GaAsP (34.9 meV) (see Table 5.1).

### 5.1.2 Phonon mode normalization

The normalization coefficient $f(q)$ of the IF and CF potentials for each phonon mode can be determined from the orthonormality and completeness conditions imposed on the phonon eigenfunctions [120, 141]. In polar crystals, optical vibrations of the lattice, characterized by a relative displacement $u(r,z)$, result in macroscopic electric and polarization fields ($E(r,z)$ and $P(r,z)$, respectively). These quantities are related to each other by the macroscopic Born-Huang theory.
Using two-dimensional Fourier transforms, we can rewrite \( u(\mathbf{r}, z) = \sum_q u(q, z)e^{iq\mathbf{r}} \), similarly for \( E(\mathbf{r}, z) \) and \( P(\mathbf{r}, z) \). For binary layers, the Fourier transforms are related by

\[
\rho(\omega_{TO}^2 - \omega^2)u(q, z) = e^*E(q, z),
\]

(5.16a)

\[
P(q, z) = \varepsilon_0(\varepsilon_{\infty} - 1)E(q, z) + e^*u(q, z),
\]

(5.16b)

where \( \rho \) is the reduced mass density, \( e^* \) the effective ionic charge density, and \( \varepsilon_0 \) is the vacuum permittivity. In Eq. (5.16b), the first term on the right-hand side represents the electronic polarization, while the second term describes the ionic polarization. Making use of the relation

\[
D(q, z) = \varepsilon_0\varepsilon(\omega)E(q, z) = \varepsilon_0E(q, z) + P(q, z),
\]

(5.17)

and Eq. (5.1), it is straightforward to obtain

\[
(e^*)^2 = \rho\varepsilon_0\varepsilon_{\infty}(\omega_{LO}^2 - \omega_{TO}^2),
\]

(5.18)

and then

\[
|\sqrt{\rho}u(q, z)|^2 = \frac{\varepsilon_0}{2\omega}\frac{d\varepsilon(\omega)}{d\omega}|E(q, z)|^2,
\]

(5.19)

where the relation that \( d\varepsilon(\omega)/d\omega = \varepsilon_{\infty}2\omega(\omega_{LO}^2 - \omega_{TO}^2)/(\omega^2 - \omega_{TO}^2)^2 \) has been used.

In the case of ternary layers \( A_xB_{1-x}C \ (0 < x < 1) \), we have [117, 129]

\[
\rho_1(\omega_{TO1}^2 - \omega^2)u_1(q, z) = e_1^*E(q, z),
\]

(5.20a)

\[
\rho_2(\omega_{TO2}^2 - \omega^2)u_2(q, z) = e_2^*E(q, z),
\]

(5.20b)

\[
P(q, z) = \varepsilon_0(\varepsilon_{\infty} - 1)E(q, z) + (1 - x)e_1^*u_1(q, z) + xe_2^*u_2(q, z),
\]

(5.20c)

with 1 and 2 referring to the BC-like and AC-like alloy modes, respectively. Combination of Eqs. (5.17) and (5.20c) results in

\[
\varepsilon_0[\varepsilon(\omega) - \varepsilon_{\infty}]E(q, z) = (1 - x)e_1^*u_1(q, z) + xe_2^*u_2(q, z).
\]

(5.21)
Substituting Eqs. (5.20a), (5.20b) and (5.2) into Eq. (5.21) leads to

\[
\left(1 - x\right) \frac{(e^*_1)^2}{\rho_1} \left(\omega^2_{TO2} - \omega^2\right) + x \frac{(e^*_2)^2}{\rho_2} \left(\omega^2_{TO1} - \omega^2\right)
= \varepsilon_0 \varepsilon_\infty \left[ (\omega^2 - \omega^2_{LO1}) (\omega^2 - \omega^2_{LO2}) \right. \\
\left. - (\omega^2 - \omega^2_{TO1}) (\omega^2 - \omega^2_{TO2}) \right].
\] (5.22)

From Eq. (5.22), we can derive the expressions for \((e^*_1)^2\) (let \(\omega = \omega_{TO1}\)) and \((e^*_2)^2\) (let \(\omega = \omega_{TO2}\)),

\[
(e^*_1)^2 = \frac{\rho_1 \varepsilon_0 \varepsilon_\infty (\omega^2_{LO1} - \omega^2_{TO1}) (\omega^2_{LO2} - \omega^2_{TO1})}{(1 - x) (\omega^2_{TO2} - \omega^2_{TO1})},
\] (5.23a)

\[
(e^*_2)^2 = \frac{\rho_2 \varepsilon_0 \varepsilon_\infty (\omega^2_{LO1} - \omega^2_{TO2}) (\omega^2_{LO2} - \omega^2_{TO2})}{x (\omega^2_{TO1} - \omega^2_{TO2})}.
\] (5.23b)

By combining Eqs. (5.20) and (5.23), we obtain

\[
(1 - x) \left| \sqrt{\rho_1} u_1(q, z) \right|^2 + x \left| \sqrt{\rho_2} u_2(q, z) \right|^2
= \varepsilon_0 \frac{d\varepsilon(\omega)}{d\omega} \left| E(q, z) \right|^2,
\] (5.24)

where the first derivative of the dielectric function has the following form for ternary alloys,

\[
\frac{d\varepsilon(\omega)}{d\omega} = \varepsilon_\infty 2\omega \left[ \frac{(\omega^2_{LO1} - \omega^2_{TO1}) (\omega^2_{LO2} - \omega^2_{TO1})}{(\omega^2 - \omega^2_{TO1})^2 (\omega^2_{TO2} - \omega^2_{TO1})} + \frac{(\omega^2_{LO1} - \omega^2_{TO2}) (\omega^2_{LO2} - \omega^2_{TO2})}{(\omega^2 - \omega^2_{TO2})^2 (\omega^2_{TO1} - \omega^2_{TO2})} \right].
\] (5.25)

The generalized normalization condition for each optical (IF and CF) phonon mode \(\nu\) in MQW structures consisting of arbitrary combinations of binary and ternary layers is given by [120, 131]

\[
\sum_i A \int_{R_i} dz \left[ (1 - x) \left| \sqrt{\rho_1} u_1(q, z) \right|^2 + x \left| \sqrt{\rho_2} u_2(q, z) \right|^2 \right] = \frac{\hbar}{2\omega_\nu},
\] (5.26)

where the summation goes over all the layers of interest, \(A\) is the in-plane area, \(x = 0, 1\) for binary alloys, and \(0 < x < 1\) for ternary ones. Substituting Eqs. (5.19) and (5.24) into Eq. (5.26), the normalization condition can be rewritten as

\[
\sum_i A \int_{R_i} dz \frac{\varepsilon_0}{2\omega_\nu} \frac{d\varepsilon(\omega)}{d\omega} \left| E_\nu(q, z) \right|^2 = \frac{\hbar}{2\omega_\nu}.
\] (5.27)
On the other hand, the electric field is related to the potential through

$$E^\nu(r, z) = -\nabla \Phi^\nu(r, z) = -\sum_q e^{i\mathbf{q} \cdot \mathbf{r}} \left( i q \hat{q} + \hat{z} \frac{\partial}{\partial z} \right) f^\nu(q) \phi^\nu(q, z), \quad (5.28)$$

where $\hat{q}$ and $\hat{z}$ are the unit vectors for the $q$ and $z$ directions, respectively, hence,

$$E^\nu(q, z) = \left( -i q \hat{q} - \hat{z} \frac{\partial}{\partial z} \right) f^\nu(q) \phi^\nu(q, z). \quad (5.29)$$

Putting Eqs. (5.29) into (5.27), we obtain the final expression for the normalization coefficient $f^\nu(q)$,

$$f^\nu(q) = \left( \frac{\hbar}{2A\omega_\nu} \right)^{1/2} \left\{ \sum_i \left[ \frac{\varepsilon_0}{2\omega_\nu} \frac{d\varepsilon_i(\omega)}{d\omega} I_i(q) \right] \right\}^{-1/2}, \quad (5.30)$$

where the summation over the layers ‘$i$’ should be carried out in one stage for IF modes, and $I_i(q)$ is defined as

$$I_i(q) = \int_{R_i} dz \left[ q^2 |\phi^\nu_i(q, z)|^2 + \left| \frac{\partial \phi^\nu_i(q, z)}{\partial z} \right|^2 \right]. \quad (5.31)$$

For IF modes, $I_i(q)$ is simplified to be

$$I_i(q) = q \left[ c_{i,+}^2 (e^{2qd_i} - 1) - c_{i,-}^2 (e^{-2qd_i} - 1) \right], \quad (5.32)$$

and for CF modes,

$$I_i(q) = \left[ q^2 + \left( \frac{m\pi}{d_i} \right)^2 \right] \frac{d_i}{2}. \quad (5.33)$$

The IF potentials are calculated for all $q$’s of interest for the 6.7 $\mu$m QCL. Arbitrarily selected normalized IF potentials, $f^\nu(q)\phi^\nu(q, z)$, at $q = 0.03$ Å$^{-1}$ are given in Fig. 5.3. It is clearly seen that the IF potentials peak at the layer interfaces and are periodic between stages. The normalized GaAs-like CF potentials for $m = 1$ and $q = 0.03$ Å$^{-1}$ are shown in Fig. 5.4. The CF potentials in each layer are independent and calculated separately, and they are plotted together for comparison. The layers consisting of the same alloy have the same confined GaAs-like LO frequencies, so there are 4 different frequencies in Fig. 5.4. It is noted that for the layers that have the same GaAs-like LO frequency within one stage, the normalized CF potentials show different amplitudes, which is
because the normalization coefficients directly depend on the layer thickness as given in Eq. (5.33). The InAs-like, GaP-like, and AlAs-like CF potentials are similar to those in Fig. 5.4, but with different amplitudes.

Figure 5.3 Selected normalized IF potentials at $q = 0.03 \ \text{Å}^{-1}$ for the 6.7 $\mu$m QCL, illustrating the continuity of the IF potentials and their periodicity between stages. The arbitrarily selected IF modes are 4 of the 18 modes within 43-48 meV from Fig. 5.2. The vertical dotted lines indicate interfaces between layers.

5.2 Electron-LO Scattering with Phonon Confinement

The obtained IF dispersions and the potentials of the IF and CF modes are utilized to evaluate the electron-longitudinal polar optical (electron-LO) phonon scattering rates, which are then incorporated in the MMC simulator [59]. The electron-LO phonon interaction Hamiltonian can be written as

$$H_{e-LO}(r, z) = -e \Phi(r, z)$$

$$= -e \sum_{q, \nu} f_{\nu}(q) \phi_{\nu}(q, z) e^{i \mathbf{q} \cdot \mathbf{r}} (b_q + b_q^+), \quad (5.34)$$
Figure 5.4 Normalized GaAs-like CF potentials at $m = 1$ and $q = 0.03 \, \text{Å}^{-1}$ for the 6.7 $\mu$m QCL. The dash-dotted red curves are the confined potentials in the GaAs layers, the dashed green ones in the GaAsP layers, the bold black curves in the AlGaAs layers, and the thin blue ones in the InGaAs layers. The vertical dotted lines indicate the interfaces between layers.

where $b_q$ and $b^+_q$ are the phonon destruction and creation operators, respectively, inserted to indicate the destruction and creation of phonons, $\nu$ goes over all IF and CF modes, while $f^{\nu}(q)$ and $\phi^\nu(q, z)$ depend on a particular IF or CF mode. Following Fermi’s Golden Rule, the transition rate of an electron from an initial state $|j\mathbf{k}\rangle$ (in-plane wave vector $\mathbf{k}$ and subband $j$) to a final state $|j'\mathbf{k}'\rangle$ is given by

$$ S_{jj'}(\mathbf{k}, \mathbf{k}') = \frac{2\pi}{\hbar} |M_{jj'}(\mathbf{k}, \mathbf{k}')|^2 \delta \left[ E' - E \pm \hbar \omega_{\nu}(q) \right], \quad (5.35) $$

with the upper and lower signs corresponding to phonon emission and absorption, respectively (this convention will be used throughout this section). The delta function guarantees energy conservation in the process, where $E' = E_{j'} + \hbar^2 k'^2 / 2m^*_j$, and $E = E_j + \hbar^2 k^2 / 2m^*_j$, with $E_j (E_{j'})$ and $m^*_j (m^*_j)$ being the electron subband energy and effective mass obtained as detailed in Chapter
2. The matrix element of the electron-LO interaction is

\[ M_{jj'}(k, k') = \langle n_{q\nu} \pm 1, j' | H_{e-LO}(r, z) | n_{q\nu}, j \rangle \]

\[ = -e \sum_{q\nu} \sqrt{n_{q\nu} + 1} \frac{1}{2} \int_V \Psi_{j'k'}^*(r, z) \times f^{\nu}(q) \phi^{\nu}(q, z) e^{\mp i q \cdot r} \Psi_{jk}(r, z), \quad (5.36) \]

where \( n_{q\nu} \) denotes the number of equilibrium phonons in mode \( \nu \), the integration is carried out in the whole volume of computation, and the electronic states take the form

\[ \Psi_{jk}(r, z) = \frac{1}{\sqrt{A}} e^{ikr} \psi_j(z). \quad (5.37) \]

Details of solving for \( \psi_j(z) \) are given in Chapter 2. By performing the integration, we obtain

\[ M_{jj'}(k, k') = -e \sum_{\nu} \sqrt{n_{q\nu} + 1} \frac{1}{2} g^{\nu}_{jj'}(q) \times \delta(k' - k \pm q), \quad (5.38) \]

where the summation over \( q \) is killed due to the delta function that ensures the in-plane momentum conservation. Given the electron initial \( (k) \) and final \( (k') \) wave vectors, the exchanged phonon wave vector must have the following magnitude,

\[ q = |k' - k| = k'^2 + k^2 - 2k'k \cos \theta, \quad (5.39) \]

with \( \theta \) being the angle between vectors \( k' \) and \( k \). The electron-phonon coupling function, \( g^{\nu}_{jj'}(q) \), is defined as

\[ g^{\nu}_{jj'}(q) = \int_0^L dz \psi_j^*(z) f^{\nu}(q) \phi^{\nu}(q, z) \psi_j(z), \quad (5.40) \]

with \( L \) being the total length of the ECD along \( z \). The total scattering rate, \( \Gamma_{jj'}(k) \), is obtained by summing over all final wave vectors \( k' \), i.e.,

\[ \Gamma_{jj'}(k) = \sum_{k'} S_{jj'}(k, k') \frac{2\pi e^2}{\hbar} \frac{A}{(2\pi)^2} \times \sum_{\nu} \int d^2k' \left( n_{q\nu} + 1 \right) \frac{1}{2} |g^{\nu}_{jj'}(q)|^2 \times \delta(k' - k \pm q) \delta (E' - E \pm \hbar \omega_{\nu}(q)). \quad (5.41) \]
where the “sum-to-integral” rule, \( \sum k' = A/(2\pi)^2 \int d^2k' \), is used. By performing the \( k' \) integration in polar coordinates and utilizing properties of the delta functions, we reach the final expression for \( \Gamma_{jj'}(k) \),

\[
\Gamma_{jj'}(k) = \frac{A m_j^* e^2}{2\pi \hbar^3} \sum \int_0^{2\pi} d\theta \left( n_{q\nu} + \frac{1}{2} \pm \frac{1}{2} \right) \\
\times \left| g_{jj'}(q) \right|^2 \vartheta \left( E_k + \hbar \omega_{\nu}^\mp \right),
\]

with \( E_k = \hbar^2 k^2 / 2m_j^* \), \( \hbar \omega_{\nu}^\mp = E_j - E_{jj'} \mp \hbar \omega_{\nu}(q) \), and \( \vartheta \) being the Heaviside step function. For the IF modes, \( n_{q\nu} \) and \( \hbar \omega_{\nu}(q) \) depend on the wave vector \( q \), but there is no \( q \)-dependence for the CF modes. The \( q \) value in Eq. (5.42) is determined from the in-plane momentum and energy conservation laws, taking into account the different effective masses in the initial and final subbands,

\[
q = \frac{\sqrt{2}}{\hbar} \left\{ \left( m_j^* + m_{jj'}^* \right) E_k + m_{jj'}^* \left( \hbar \omega_{\nu}^\mp \right) \right. \\
- 2 \cos \theta \left[ m_j^* m_{jj'}^* E_k \left( E_k + \hbar \omega_{\nu}^\mp \right) \right]^{1/2} \right\}^{1/2}.
\]

Since the right-hand side of Eq. (5.43) has a complicated \( q \)-dependence for the IF modes due to their dispersions, it is necessary to numerically solve Eq. (5.43) for a required \( q \) during the IF rate calculations. If \( q \leq q_{\text{lim}} \), the exact IF frequency \( \omega_{\nu}(q) \), is taken; otherwise, the IF frequency at \( q_{\text{lim}}, \omega_{\nu}(q_{\text{lim}}) \), is used to compute \( q \). The calculated IF emission rates from the upper lasing level 3 to the lower lasing level 2 are shown in Fig. 5.5 for the 6.7 \( \mu \)m QCL, at the field of 55 kV/cm and the lattice temperature of 77 K. The cumulative rate for the 17 GaAs-like IF modes within the 32.9-33.6 meV window is indistinguishable from the horizontal axis, implying their negligible contribution. The 4 InAs-like IF modes also have a small contribution due to the low In content (10\%) and the smaller phonon energies. The dominant contributions come from the higher-energy IF modes, where the GaAs-like modes within 34-36.4 meV contribute the most, as expected for transitions in GaAs-rich wells.

The CF scattering rates can be calculated in the zero-dispersion limit of the bulk-like CF modes, since it was shown \([128]\) that these rates are insensitive to the bulk dispersions. For each mode
Figure 5.5 IF emission rates from the upper to lower lasing levels ($3 \rightarrow 2$) for the 6.7 $\mu$m QCL at $F = 55$ kV/cm and $T = 77$ K. The numbers in the brackets denote the energy ranges (in meV) of the corresponding IF modes shown in Fig. 5.2. Each curve represents the sum of the IF rates for all the modes in the specified energy range.

Figure 5.6 CF emission rates of $3 \rightarrow 2$ for the 6.7 $\mu$m QCL at $F = 55$ kV/cm and $T = 77$ K. The GaAs-like modes include GaAs LO and 3 GaAs-like LO modes that originate from InGaAs, AIGaAs, and GaAsP.
$\hbar \omega_{LO}$, the CF rates are computed separately with $m = 1, 2, \ldots, 20$ (the contribution of higher-order modes is negligible) for all the layers that have the same frequency, and are summed up to yield the total rate for the particular mode. Figure 5.6 shows the CF emission rates of $3 \rightarrow 2$ for the 6.7 $\mu$m QCL at $F = 55$ kV/cm and $T = 77$ K. The GaP-like CF mode shows a negligible contribution to the total CF rate, since each stage has only one GaAsP layer and the electronic wavefunction magnitudes in that layer are very small. The 4 GaAs-like CF modes completely dominate the total rate, among which the GaAs-like mode originating from InGaAs contributes to the CF rate about an order of magnitude more than the other 3 modes.

5.3 Effect of Phonon Confinement

Next, the effects of phonon confinement on the electron-LO scattering rates and the transport properties of the optimized 6.7 $\mu$m structure C are investigated in detail. In Figs. 5.7 and 5.8, the sum of the IF and CF emission rates for the $3 \rightarrow 2$ and $2 \rightarrow 1$ transitions are compared to the rates obtained using bulk GaAs and AlAs phonons. In both transitions, it can be seen that the total IF+CF rates fall between the bulk GaAs and AlAs rates, consistent with the sum rule for scattering rate calculation in quantum-well systems [94]. And the total IF+CF rates are very close to the bulk GaAs rates, which justifies the use of bulk GaAs phonon approximation in Chapters 2-4. This result also agrees with other work [94, 114] on phonon confinement in quantum-well systems.

In order to explore the effects of phonon confinement on the transport properties of GaAs QCLs, we compute the scattering rates of all the $\Gamma$- and X-valley electronic states of interest by all the 56 IF and 7 CF modes for the optimized 6.7 $\mu$m structure C, and fully incorporate them in the multivalley Monte Carlo simulator [58, 59]. The $\Gamma$-valley electronic states are calculated using a three-band $k \cdot p$ method within the envelope function approximation, while the X-valley states are obtained by solving the single-band effective mass equation [59], as detailed in Chapter 2. For sheet doping densities $N_s < 4 \times 10^{11}$ cm$^{-2}$ (in our structure, $N_s = 3.8 \times 10^{11}$ cm$^{-2}$) and typical operating conditions in GaAs-based mid-IR QCLs, it has been shown [46, 62] that the electronic states obtained by self-consistently solving the coupled 1D Schrödinger-Poisson equations are virtually the same as those obtained from the Schrödinger solver alone within the linear potential drop
Figure 5.7 Emission rates of $3 \rightarrow 2$ for the 6.7 $\mu$m QCL at $F = 55$ kV/cm and $T = 77$ K using the bulk phonon approximation and the IF+CF modes.

Figure 5.8 Emission rates from the lower lasing level 2 to the active ground level 1 ($2 \rightarrow 1$) for the 6.7 $\mu$m QCL at $F = 55$ kV/cm and $T = 77$ K using the bulk phonon approximation and the IF+CF modes.
approximation (see Sec. 2.4). Therefore, in order to reduce the computation burden (exacerbated by the inclusion of the IF and CF modes), electronic states from the Schrödinger solver with a linear potential drop are directly used to calculate the scattering rates in the Monte Carlo simulation, and remain fixed during the simulation for a given electric field.

Due to the enormous memory requirement of storing the electron-IF and electron-CF rates for all phonon modes, only the total IF+CF rates between any two subbands are pre-calculated and stored. During the Monte Carlo simulation, the IF and CF rates between two specified subbands by all phonon modes are calculated in real time, and random numbers are used to choose a particular phonon mode for scattering of an electron. Besides the electron-IF and electron-CF scattering in both Γ- and X-valleys, the electron-electron and various intervalley scattering mechanisms are also included for the same stage and between adjacent stages (for details, see Chapter 2).

The output characteristics of the 6.7 µm QCL from the above-described Monte Carlo simulator are presented in Figs. 5.9-5.11 at 77 K and 300 K lattice temperatures, where the results with the phonon confinement are compared to those obtained by using the bulk GaAs phonon approximation. From Fig. 5.9, it is clear that phonon confinement induces a minor correction to the current density for a given electric field at both 77 K and 300 K. The population inversion, with phonon confinement included, remains nearly identical to the case with bulk phonons at both temperatures (Fig. 5.10). On the other hand, the electron temperature $T_e$ (Fig. 5.11), which characterizes the degree of electron heating, is somewhat reduced at high currents due to phonon confinement. This reduction occurs because, in the case of including phonon confinement, high-energy (> bulk GaAs LO) IF and CF phonons come into play, and through emitting those high-energy phonons, electrons are able to dissipate more energy to the lattice, leading to a lower electron temperature. Overall, simulation results demonstrate that phonon confinement turns out to have a minor impact on the device performance of the 6.7 µm QCL even at room temperature, and thus the GaAs bulk phonon approximation remains valid in transport simulation of GaAs QCLs.

Since the 6.7 µm GaAs QCL is a pretty generic mid-IR GaAs structure: it uses 45% Al and has a typical layer sequence and layer thickness used in such a device, so the above conclusion should hold for any mid-IR GaAs QCL that uses Al content equal to or less than 45% (since, in a
Figure 5.9 Electric field vs current density at 77 K and 300 K lattice temperatures using the bulk GaAs phonon approximation and the IF+CF modes. Dashed and solid curves are polynomial fits to the data points to guide the eyes.

Figure 5.10 Population inversion vs current density at 77 K and 300 K using the bulk GaAs phonon approximation and the IF+CF modes. Dashed and solid curves are polynomial fits to the data points to guide the eyes.
Figure 5.11 Electron temperature vs current density at 77 K and 300 K using the bulk GaAs phonon approximation and the IF+CF modes. Dashed and solid curves are polynomial fits to the data points to guide the eyes.

nutshell, the less Al in the barriers, the better the GaAs bulk phonon approximation). As for the QCLs with 100% Al in the barriers, the effect of phonon confinement is expected to increase the electron-LO scattering rates to a larger degree due to the high Al content, but the increase should not be dramatic and the rates should be still much lower than the bulk AlAs rates due to the wide GaAs wells (generally wider than the barriers). Becker et al. [130] showed that the experimental thermal behavior of a 11.4 µm GaAs/AlAs QCL is better explained by a theoretical calculation including phonon confinement; however, the calculated difference between the two cases (with and without optical phonon confinement) is not dramatic (Fig. 2 in Ref. [130]). Therefore, our conclusion that the bulk phonon approximation suffices is certainly applicable to any mid-IR GaAs QCL that uses Al% ≤ 45%. The bulk phonon approximation should be a decent estimate even for 100% Al in the barriers (i.e., for GaAs/AlAs QCLs). Furthermore, since terahertz (THz) GaAs QCLs in general utilize low-Al content (15% or 30%) in the AlGaAs barriers and wide GaAs wells to achieve terahertz (small energy) transitions, phonon confinement should not be important in them. Williams and Hu [114] showed that the total electron-LO scattering rates, including phonon confinement in one stage of two GaAs/Al0.3Ga0.7As THz QCLs, are very close to those obtained
using GaAs bulk phonons (Fig. 5 in Ref. [114]). Therefore, our conclusion also holds for THz GaAs QCLs.
Chapter 6

Summary and Future Work

6.1 Summary

At the core of this work was the development of a multivalley ensemble Monte Carlo simulator of electron transport in QCLs, which includes both Γ- and X-valley transport for the first time in the QCL simulation community. The details of solving the Γ- and X-valley electronic states were presented. The Γ-states were calculated using the \( \mathbf{k} \cdot \mathbf{p} \) method within the envelope function approximation, while the X-states were obtained within the effective mass framework. The Γ- and X-states were used to compute all the relevant scattering rates in detail. The included interaction mechanisms were electron-longitudinal polar optical phonon, electron-electron, and intervalley scattering processes, within the same stage and between adjacent stages (the LO and intervalley phonons were treated as bulk phonons). The effects of the electrostatic potential on the subband energy levels and wavefunctions have been studied by self-consistently solving the Schrödinger-Poisson equations. The electrostatic potential was shown to have a minor effect on the electronic states for typical sheet doping densities in QCLs (below \( 4 \times 10^{11} \text{ cm}^{-2} \)). The following three major accomplishments are resulted from this work [58, 59, 64, 66].

6.1.1 X-valley leakage in GaAs/AlGaAs QCLs

The multivalley Monte Carlo (MMC) simulator developed has been applied to simulate the transport properties of the two well-known equivalent-design GaAs/Al\(_{0.33}\)Ga\(_{0.67}\)As (33% QCL) and GaAs/Al\(_{0.45}\)Ga\(_{0.55}\)As (45% QCL) mid-infrared QCLs [58, 59, 63]. The effect of the X-valley transport on the performance of the two QCLs has been investigated for the first time in the QCL
simulation community. The dominant X-valley leakage path in both QCLs was found to be the interstage $X \rightarrow X$ intervalley scattering. The magnitude of the $X \rightarrow X$ leakage current $J_X$ depends on the temperature and occupation of the X-valley subbands, which are populated primarily by the same-stage scattering from the $\Gamma$-continuum states ($\Gamma_c$). At the low lattice temperature of 77 K, $J_X$ is small up to very high fields in both QCLs. However, at room temperature, the 33\% QCL shows a much higher $J_X$ than the 45\% QCL even at low fields, which helps explain why it never lased at this temperature. The reason is that in the 33\% QCL the coupling between the $\Gamma$-localized states ($\Gamma_l$, also called the injector states) and the next-stage $\Gamma_c$ is strong, which leads not only to the well-recognized $\Gamma_l \rightarrow \Gamma_c$ leakage, but also facilitates filling of the X states through efficient intrastage $\Gamma_c \rightarrow X$ scattering; with high X-valley population and high temperature, the efficient interstage $X \rightarrow X$ scattering yields a large $J_X$. In contrast, in the 45\% QCL, coupling of $\Gamma_l$ with the next-stage $\Gamma_c$ is weak, due to good localization of the $\Gamma_l$ states coming from the high (45\% Al) barriers. Consequently, occupation of the X states through the same-stage $\Gamma_c \rightarrow X$ scattering is low, leading to small $J_X$ up to high fields. The simulated threshold current density $J_{th}$ with the X-valley leakage included is in very good agreement with experiment for both QCLs at both cryogenic and room temperatures.

In addition, the effect of the electron-electron (e-e) interaction and electron heating in the two QCLs have been studied. The e-e scattering was shown to have two important effects: (i) it strongly increases intersubband electron redistribution; (ii) it drives the electron distributions in both the active region and the injector subbands to the heated-Maxwellian distributions, and all the electron distributions have a common effective electron temperature, higher than the lattice temperature. The elevated electron temperature directly reflects that, in QCLs, the electron system is not able to dissipate the large amount of energy provided by the external bias, which leads to strong internal heating.

### 6.1.2 Simulation and optimization of deep-active-well GaAs QCLs

Three deep-active-well QCL structures (A, B, and C) that utilize the strain-compensated
In$_{0.1}$Ga$_{0.9}$As/GaAs/Al$_{0.45}$Ga$_{0.55}$As materials system grown on GaAs substrates have been analyzed. The three QCLs were all designed to emit at 6.7 $\mu$m, the shortest wavelength to be achieved in GaAs-based mid-infrared QCLs at room temperature, but have different injector designs. The MMC simulator was employed to simulate the output characteristics of the three lasers and optimize the injector design for room-temperature operation [64, 65].

The initial QCL (structure A), designed by Mithun D’Souza, shows good performance at the low lattice temperature of 77 K, namely, negligible X-valley leakage current, large modal gain, and a reasonably low threshold current density $J_{th} = 6.0$ kA/cm$^2$, given the total waveguide loss of 19 cm$^{-1}$. Nevertheless, the room-temperature performance of structure A is rather poor: it shows large X-valley leakage current and insufficient modal gain. This is because the injector layer sequence of structure A was not optimized, so there is a strong coupling between the injector states and the next-stage $\Gamma$-continuum states, which leads to a large X-valley leakage current and a dramatic electron loss to the X-valleys at room temperature.

In order to achieve room-temperature laser emission, an improved structure B was suggested by Mithun D’Souza, which utilizes the same active-region design as structure A, but increases the thicknesses of the injector wells. The wider wells bring down the injector top state and shift its wavefunction maximum farther away from the continuum. Consequently, the coupling of the injector states with the next-stage $\Gamma_c$-states is reduced, leading to a smaller X-valley leakage. This smaller X-valley leakage, together with the improved dipole matrix element, enhances the room-temperature modal gain, but it just marginally meets the total loss line. Moreover, structure B does not have enough modal gain for lasing at 77 K. Both situations can be traced back to the fact that the next-stage upper lasing level is weakly coupled to the injector ground level and far above it in energy, which results in poor injection efficiency and consequently low population inversion.

Identification of issues associated with structures A and B by using the MMC simulator plays a direct and important role in obtaining the optimized structure C. The injector layer sequence of this structure was designed by myself. In structure C, not only is the coupling between the injector states and the next-stage $\Gamma_c$-states reduced, but the next-stage upper lasing level also has a good wavefunction overlap and a small energy difference with the injector ground level. The optimized
structure C shows the best device performance among the three structures, namely, it shows adequate optical gain for lasing at both 77 K and 300 K lattice temperatures, in conjunction with an efficient waveguide design (due to Mithun D’Souza) that all three structures share. Although the optimized structure C shows a higher X-valley leakage current than the state-of-the-art 9.4 \( \mu m \) GaAs QCL of Page et al. [1] due to the shorter wavelength, its threshold current density \( J_{th} \) was calculated to be 5 kA/cm\(^2\) at 77 K and 14 kA/cm\(^2\) at 300 K, close to the values for the 9.4 \( \mu m \) QCL (\( J_{th} = 4 \) kA/cm\(^2\) at 77 K and 16.7 kA/cm\(^2\) at 300 K) [1]. Furthermore, electron heating at room temperature is similar to that of the 9.4 \( \mu m \) device. Therefore, structure C demonstrates that utilization of deep active quantum wells appears to be an attractive design approach for high performance GaAs-based QCLs at wavelengths below 7 \( \mu m \).

### 6.1.3 Investigation of phonon confinement in mid-IR GaAs QCLs

Phonon confinement has been implemented in detail in the MMC simulator and its effects on the electronic transport properties in mid-infrared, GaAs-based multiple-quantum-well (MQW) QCLs were investigated [66, 67]. The macroscopic dielectric continuum model was used to describe the interface (IF) and confined (CF) optical phonons. The IF-mode dispersions were obtained by using the transfer matrix method with periodic boundary conditions. Normalization coefficients of the IF and CF potentials were thoroughly derived for MQW structures consisting of arbitrary combinations of binary and ternary alloys. The resulting IF dispersions and phonon potentials were utilized to compute the scattering rates for all the relevant \( \Gamma \)- and X-valley electron states. Electron-IF, electron-CF, electron-electron, and intervalley scattering mechanisms were fully incorporated in the MMC simulation of the optimized deep-active-well 6.7 \( \mu m \) QCL structure C. At both 77 K and 300 K lattice temperatures, simulation results demonstrated that inclusion of phonon confinement enhances the electron-LO phonon scattering rates only to a small extent and induces very minor correction to the device performance, with respect to the results obtained in the bulk-GaAs-phonon approximation. Therefore, the bulk phonon approximation in GaAs-based QCLs remains meritorious due to its simplicity and high accuracy. This conclusion ought to be applicable to virtually any mid-IR GaAs-based QCL, due to the fact that GaAs wells are generally
thicker than AlGaAs barriers, and it should also hold for THz GaAs QCLs, thanks to both wide GaAs wells and low-Al content barriers.

6.2 Future Work

The MMC simulator developed has demonstrated diverse application capabilities: quantifying the X-valley leakage current and electron heating, and optimizing new GaAs QCL designs for desired performance. It is also opening several avenues for future research, which are outlined in the following:

6.2.1 Extension to InP-based QCLs

Mid-IR QCLs are generally fabricated on two types of substrates: InP and GaAs, with GaAs-based QCLs being the focus of this research. InP-based QCLs have demonstrated CW RT lasing emission in the 3.8-9.5 \( \mu \text{m} \) range [11–18], and pulsed operation around 3 \( \mu \text{m} \) up to 150 K [21,22]. Great research effort is on pushing the wavelengths of InP QCLs to below 3 \( \mu \text{m} \) with CW RT operation capability. To date, InP QCLs lasing around 3 \( \mu \text{m} \) can only work in pulsed mode and at low temperatures. A systematic simulation of this type of lasers would provide insight into optimizing InP QCL designs for high performance below 3 \( \mu \text{m} \).

There have only been a few references in the literature on electron transport simulation of InP QCLs. Ref. [143] reported the simulation of the InGaAs/InAlAs QCL lattice-matched to an InP substrate of Beck et al. [11], through a self-consistent solution of the subband population rate equations. A Monte Carlo simulation [118] was also performed for lattice-matched InGaAs/InAlAs QCLs. No published work has been found on transport simulation of strain-compensated InP-based QCLs, which have demonstrated the best device performance in the range of 3-9.5 \( \mu \text{m} \) wavelengths.

Extension of the MMC simulator developed in this work to strain-compensated InP QCLs would be an intriguing research subject. The \( \mathbf{k} \cdot \mathbf{p} \) approach employed in the simulator can conveniently account for the strain effect, which is very important in these strain-compensated devices. Moreover, the multivalley nature of the simulator allows for quantifying the satellite-valley leakage
that limits the high-temperature performance of InP QCLs [144], so it can provide insight for device optimization. However, there are a few issues that require careful attention, when adapting the MMC simulator to InP-based QCLs. First of all, the nonparabolicity in InGaAs/InAlAs and other InP-based materials systems can be significant [19], so the parabolic in-plane dispersion may no longer be a good assumption, and a full-band $k \cdot p$ calculation would be required instead. Secondly, since the parameters of InP-based materials systems are not well characterized and can vary in a wide range, careful tuning of certain parameters in the simulator may be required to obtain good agreement between theory and experiment. In addition, due to the very high barriers in InP QCLs, there are many more electronic states in each stage than in GaAs/AlGaAs QCLs, which increases the computation time dramatically.

6.2.2 Inclusion of thermal transport

As pointed out at the end of Chapter 4, the calculated electronic temperature from the MMC simulator cannot be directly compared to the experimentally measured value. For a direct comparison, thermal transport needs to be included. Enormous internal (electron and lattice) heating is one of the main reasons for rapid degradation of the QCL performance with temperature. Several experimental measurements of both electronic and lattice temperatures in GaAs- and InP-based QCLs [61, 145, 146] have been reported. The measured thermal behavior tends to be theoretically explained by solving the heat diffusion equations [145, 147, 148].

However, thermal and electronic transport in a QCL device are inherently coupled. The electron system in a QCL is strongly heated, as shown in this work (Chapter 3), due to the excess input electrical power. The hot electrons eventually lose the energy to the crystal lattice through multiple-phonon emission processes, which lead to nonequilibrium optical phonons [149]. These optical phonons do not conduct heat because of their near-zero group velocity, but they decay to acoustic phonons, which have large group velocity and can thus efficiently transfer the heat, contributing to the heat conductivity. In this process, the electron and phonon dynamics interact with each other in complicated ways, so a systematic model, which can simultaneously explain the electronic and thermal properties of a QCL device, should be a self-consistent solution of both
the electron and phonon (thermal) transport equations. Ref. [150] presented a simplified model that self-consistently solves the temperature-dependent threshold current density equation and heat equation. A more accurate model would be self-consistently solving the Boltzmann transport equations for both electrons and phonons using the ensemble Monte Carlo approach [151]. With the Monte Carlo electron transport simulator already developed in this work, one would need to develop the phonon transport simulator and couple it to the electron transport part.

Nevertheless, this kind of fully-coupled microscopic electron and phonon simulation will face serious challenges when applied to complex heterostructures like QCLs. A trade-off between accuracy and feasibility can be made by replacing the Boltzmann phonon transport equation with the heat diffusion equation, resulting in a model similar to the electrothermal Monte Carlo method by Sadi et al. [152]. Even with this simplification, there is another immediate difficulty that has to be addressed: the incompatibility of computation domains. The electron transport is simulated in a very small fraction of a QCL device, namely, two or two and half stages, while the thermal transport needs to be considered in a whole device including the waveguide, so a clever way to connect the electron and thermal transport simulations is ultimately necessary.

6.2.3 Development of the IQB transport simulator

To date, all QCLs have a relatively low (< 5%) wallplug efficiency (the ratio of the output optical power to the input electrical power), with one exception [18] that achieved 9% wallplug efficiency at room temperature [18], but showed a rapidly decreasing slope efficiency due to carrier leakage to the continuum. To obtain high wallplug efficiency, the concept of intersubband quantum box (IQB) lasers was proposed by Prof. Dan Botez’s group at the University of Wisconsin-Madison [153–155]. This type of laser was predicted to have wallplug efficiency as high as 20-30% and superior device performance in the 3-5 μm wavelength range [155].

Figure 6.1 shows the schematic of an IQB structure taken from Ref. [156]. An IQB laser is a single-stage intersubband-transition laser with a core region composed of a 2D array of small, box-like regions (called “active boxes”) separated by current-blocking material (CBM). The 2D array of active boxes, together with low-doped, n-type GaAs layers on either side, constitutes the core
of an optical waveguide with heavily doped, $n$-type GaAs cladding layers. As shown in Fig. 6.2, each active box has a superlattice-type electron injector followed by an active region composed of two deep InGaAs quantum wells (QW) where the electron transition occurs, and an electron Bragg-mirror region. Since the optical gain is created in a 6.6 nm-thick “box” with 30 nm-wide sides, it is called a quantum box design.

Figure 6.1 Schematic of an IQB structure (CBM = Current Blocking Material).

IQB lasers are expected to be able to operate at room-temperature (RT) in continuous-wave (CW) mode with high CW output power and the CW wallplug efficiency $\geq 20\%$ [155, 156]. The expected high performance of the IQB structure [153–155] relies on two important factors, namely the phonon bottleneck effect [157] and the deep-well design [64, 155]. The phonon bottleneck effect is a confinement-induced reduction in the optical-phonon scattering efficiency. The “box” design in an IQB structure leads to the quantization of electron and phonon states in all three directions, which increases the LO-phonon-assisted electron relaxation times to $\geq 50$ ps [158,159]. These times are long enough to strongly favor the radiative transitions with respect to the phonon-assisted relaxation, so the gain per radiative stage can be increased to the point that only a single radiative stage is needed for lasing, thus eliminating the need for a cascade process. The long
phonon-assisted relaxation time was shown [155] to increase the wallplug efficiency by 20-30 times. Moreover, the deep InGaAs QWs design in the optical gain region has been demonstrated [105] to result in negligible carrier leakage to the continuum with increasing temperature, which promises low threshold currents, high output powers and temperature-insensitive characteristics [58, 59].

Although IQB lasers have been predicted to have high device performance, the research on IQB lasers is still in its initial stage. A systematic IQB transport simulator is crucial in advancing the IQB research. However, strong confinement in all three directions in these structures imposes several great challenges in developing an IQB transport simulator. First, electronic states become discrete in all three directions, which requires a proper generalization of the $k \cdot p$ method used in the present QCL simulator for efficient and accurate calculation of electronic levels. To our knowledge,
this kind of generalization has not been done. It will take a significant effort to generalize the $k \cdot p$
method.

The second challenge lies in the efficient implementation of the electron-electron (e-e) interaction among many discrete electron subbands. The e-e scattering has been shown [49, 62] to play a crucial role in determining the performance of QCLs. It is also expected to be very important in IQB lasers. Direct implementation of the e-e interaction in these 0D systems will be very computationally demanding. Novel algorithms need to be developed for efficient implementation of the e-e scattering in IQB lasers.

The third challenge is the calculation of fully confined optical and interface phonon spectra. The underlying idea of the IQB laser superior performance with respect to the QCLs in the continuous-wave (CW) regime is the phonon bottleneck effect [157], resulting in phonon-assisted relaxation times greater than 50 ps, which is a major improvement with respect to the lifetimes of 1-2 ps in QCLs. Therefore, accurate characterization of phonon dynamics in IQB structures is extremely important. Confined optical and interface phonon spectra in 2D and 1D systems have been calculated in literature [66], but 0D phonon modes have not been well studied. Calculation of 0D optical and interface phonon modes is technically and computationally difficult.

Last but not least, a self-consistent calculation of scattering rates among the fully confined electronic states due to the many fully confined phonon modes is a challenge. Both the electron and phonon states are fully quantized, which will lead to an incredible demand for computation resources. Novel algorithms need to be implemented for efficient calculation.

The Monte Carlo simulator developed in this work would be very useful in helping build the first comprehensive transport simulator for IQB lasers. Although the present simulator is developed particularly for quantum-well systems (1D confinement) and cannot be directly applied to quantum-box systems (3D confinement), its results can be used to test the IQB simulation code in the quantum-well limit (the two directions perpendicular to the active layers in an IQB laser are large enough so that the IQB laser behaves like a QCL). Furthermore, understanding of techniques used in the present QCL simulator will definitely be helpful in development of the IQB simulation.
LIST OF REFERENCES


[71] X. Gao, M. D’Souza, D. Botez, and I. Knezevic, presented at the 7th International Conference on Numerical Simulation of Optoelectronics Devices (NUSOD’07), Newark, Delaware (September 24-28, 2007 (BEST STUDENT PAPER AWARD)).

[72] X. Gao, D. Botez, and I. Knezevic, presented at the 12th International Workshop on Computational Electronics (IWCE-12), Amherst, Massachusetts (October 8-10, 2007).


Appendix A: Simulation of GaAs-based 7.9 µm QCLs

In this Appendix, the Monte Carlo simulator presented in Chapter 2 was utilized to simulate the performance of two GaAs-based 7.9 µm QCLs, which was also a part of my PhD work. Simulation results of the initial 7.9 µm QCL structure demonstrated that the X-valley transport has a negligible effect on the performance at the low lattice temperature of 80 K, as expected, but the calculated threshold current density was two times of the desired value. To analyze the source of the large current, the various current components were calculated separately, and the current through the Γ-continuum states showed the dominant contribution in the total current, leading to poor injection efficiency. This large Γ-continuum leakage comes from the fact that, in the initial 7.9 µm QCL, the Γ-continuum states are strongly coupled to the injector states. In order to reduce this coupling, the structure was modified, resulting in an improved design. The performance of the improved design was simulated and the results were discussed.

A.1 The initial Design

The initial design of the 7.9 µm QCL was done by Mithun D’Souza, a graduate student of Prof. Dan Botez. My work was to simulate the performance of the structure and provide a suggestion for design improvement according to the simulation results. Figure A.1 shows the conduction band profile and the Γ-valley wavefunction moduli squared for the initial 7.9 µm QCL, where the electrostatic potential from the Poisson equation was included self-consistently with the Schrödinger solver, as described in Chapter 2, so there is additional band bending superimposed on the linear potential energy drop in the band profile. The reason for including the self-consistent electrostatic potential is that the structure is doped to have a sheet density of $6.62 \times 10^{11}$ cm$^{-2}$ (Fig. A.1), beyond the limit of $4 \times 10^{11}$ cm$^{-2}$ (see Chapter 2.4), so it is necessary to include the effect of the electrostatic potential on the electronic states. The layer materials composition, thickness, and doping concentration used in one stage (defined as active region + Bragg reflector + injector) starting from the injection barrier are shown in Table A.1.
Figure A.1 Fifteen $\Gamma$-valley subbands in two adjacent stages for the initial 7.9 $\mu$m QCL. In each stage, the three green curves are the $\Gamma$-continuum states, the three bold black curves are the lasing levels, and the nine thin black curves are the injector states. The electronic states were obtained from the self-consistent Schrödinger-Poisson solver, as described in Chapter 2, so there is band bending due to the electrostatic potential in addition to the applied linear potential energy drop in the conduction band profile.
<table>
<thead>
<tr>
<th>Material</th>
<th>Layer Thickness (Angstrom)</th>
<th>Doping (cm(^{-3}))</th>
</tr>
</thead>
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<td>13 (injection barrier)</td>
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Table A.1 Layer material, thickness, and doping concentration in one stage (defined as active region + Bragg reflector + injector) starting from the injection barrier for the initial 7.9 \(\mu\)m QCL. The doping profile gives rise to a sheet density of \(6.62 \times 10^{11}\) cm\(^{-2}\) per stage.
A.1.1 Low-Temperature Characteristics

The simulator developed in Chapter 2 was employed to simulate the performance of the initial 7.9 \( \mu \)m QCL (the phonons were treated in the bulk-GaAs-phonon approximation). Figure A.2 shows its electric field vs current density behavior at the lattice temperature of 80 K, with and without the X-valley transport. At this low temperature, the X-valley transport has a small effect on the current density at any given field in the range of interest, due to the low number of phonons.

![Electric field vs current density at the lattice temperature of 80 K, with and without the X-valley transport, for the initial 7.9 \( \mu \)m QCL.](image)

Figure A.2 Electric field vs current density at the lattice temperature of 80 K, with and without the X-valley transport, for the initial 7.9 \( \mu \)m QCL. Electronic states used in the simulation were obtained from the self-consistent Schrödinger-Poisson solver, as described in Chapter 2.

The modal gain as a function of the current density is shown in Fig. A.3 at the lattice temperature of 80 K, with and without the X-valley transport, for the initial 7.9 \( \mu \)m QCL. The modal gain was calculated using Eq. (4.1), with the waveguide parameters \( n = 3.27, \Gamma_w = 0.465 \), and the estimated total losses \( \alpha_w + \alpha_m = 20 \) cm\(^{-1} \), provided by Mithun D’Souza, and \( L_p = 62.2 \) nm, \( \langle z_{32} \rangle = 1.7 \) nm, \( 2\gamma_{32} = 12 \) meV. Consistent with Fig. A.2, the X-valley transport has a very small effect on the modal gain. With a total waveguide loss of 20 cm\(^{-1} \), the threshold current density \( (J_{th}) \) was found from Fig. A.3 to be 10 kA/cm\(^2 \), two times higher than the desired value of 5 kA/cm\(^2 \), which is close to the \( J_{th} = 4 \) kA/cm\(^2 \) of the state-of-the-art 9.4 \( \mu \)m GaAs QCL [1].
Figure A.3 Modal gain vs current density at the lattice temperature of 80 K, with and without the X-valley transport, for the initial 7.9 µm QCL. Electronic states used in the simulation were obtained from the self-consistent Schrödinger-Poisson solver, as described in Chapter 2. The dash-dot horizontal line denotes the total waveguide loss, provided by Mithun D’Souza. The intersection of the loss line with the modal gain linear curve gives rise to the threshold current density.

large $J_{\text{th}}$ value is not due to the X-valley leakage, since it has a negligible effect on the performance at 80 K, as seen in Figs. A.2-A.3. In order to investigate the source of the large $J_{\text{th}}$ value, it was necessary to explore the different components of the current, as presented in the next section.

**A.1.2 Analysis of Current Components**

Since the X-valley leakage has a negligible effect on the performance shown above, only the Γ-valley transport was included in the following simulation. Without the X-valley transport, the current through a QCL structure is proportional to the scattering rates (electron-LO and electron-electron) within the Γ-valley between two adjacent stages. Figure A.4 shows how the Γ-valley current components are defined in two adjacent stages of a QCL structure. Each current component (e.g., $J_{\text{up}}$) is proportional to the net scattering rate from all the fifteen Γ-states in the stage $\lambda$ to the
corresponding states (e.g., the upper lasing level) in the next stage ($\lambda + 1$). The total current through the structure is the sum of the five components.

![Diagram of two adjacent stages in a QCL structure](image)

Figure A.4 Schematics of two adjacent stages in a QCL structure to define the $\Gamma$-valley current components. The green denotes the $\Gamma$-continuum states, the black levels represent the upper lasing level, the lower lasing level, and the active ground level, respectively, and the red denotes the $\Gamma$-injector states. $J_{\text{cont}}$ is the net current from all the fifteen $\Gamma$-states in stage $\lambda$ to the $\Gamma$-continuum states in the next stage ($\lambda + 1$) ($J_{\text{cont}}$ is similar to $J_c$ defined in Chapter 3, where $J_c$ is the net current from the injector states to the next-stage $\Gamma$-continuum states). Similar definitions are applied to $J_{\text{up}}$, $J_{\text{low}}$, $J_{\text{gnd}}$, and $J_{\text{inj}}$. The total $\Gamma$-valley current through a QCL structure is defined as the sum of the five components.

With the current components defined and the associated changes made in the code, the simulator was rerun to obtain the values of current components. In this case, the emphasis was on determining the relative contribution of each current component, so in order to save the computation time, the electronic states were obtained from the Schrödinger solver, within the linear
potential energy drop approximation. The current components as a function of electric field are shown in Fig. A.5, from which one can see that the current through the $\Gamma$-continuum states ($J_{\text{cont}}$) is the most dominant current component at fields above 45 kV/cm, leading to a very poor injection efficiency. To achieve a high injection efficiency, the current through the upper lasing level ($J_{\text{up}}$) should be the largest component, so that most electrons are injected into the upper lasing level where they contribute to the lasing process. Unfortunately, in the initial 7.9 $\mu$m QCL structure, the $J_{\text{up}}$ component is rather small (less than 40%) with respect to the total current, whereas the $J_{\text{cont}}$ component is significant, especially at high fields (Fig. A.5). The large leakage current through the $\Gamma$-continuum states ($J_{\text{cont}}$) results in a very poor injection efficiency, which leads to a high threshold current density. The reason for the large $J_{\text{cont}}$ is that, in the initial 7.9 $\mu$m QCL, the $\Gamma$-continuum states [green curves in stage $(\lambda+1)$ in Fig. A.1] are peaked in the first active well, to the right of the injection barrier, resulting in a large wavefunction overlap with the injector states from the previous stage (thin black curves in stage $\lambda$) and consequently a strong $\Gamma$-continuum leakage.

Figure A.5 Electric field vs current density at the lattice temperature of 80 K, without the X-valley transport, for the initial 7.9 $\mu$m QCL. Electronic states used in the simulation were obtained from the Schrödinger solver, within the linear potential energy drop approximation. The current components are defined in Fig. A.4, and $J_{\text{tot}}$ is the sum of all the current components.
A.2 The Improved Design

According to the results and analysis, we suggested that decreasing the first active well thickness with an appropriate adjustment of other layers thickness would reduce the $\Gamma$-continuum leakage. With this improvement strategy in mind, Mithun D’Souza modified the structure and obtained an improved design. The resulting conduction band profile and the $\Gamma$-valley subbands in two adjacent stages are shown in Fig. A.6 for the improved 7.9 $\mu$m QCL. It can be seen that the $\Gamma$-continuum wavefunctions peaks are shifted away from the injection barrier, due to a decrease of the thickness of the active well right next to the injection barrier. The layer material, thickness, and doping concentration, used in one stage of the improved QCL starting from the injection barrier, are given in Table A.2. Note that the doping concentration has been changed to obtain a sheet density of $4.0 \times 10^{11}$ cm$^{-2}$ per stage.

Figure A.6 Fifteen $\Gamma$-valley subbands in two adjacent stages for the improved 7.9 $\mu$m QCL. In each stage, the three green curves are the $\Gamma$-continuum states, the three bold black curves are the lasing levels, and the nine thin black curves are the injector states. Electronic states were obtained from the self-consistent Schrödinger-Poisson solver, as described in Chapter 2.
<table>
<thead>
<tr>
<th>Material</th>
<th>Layer Thickness (Angstrom)</th>
<th>Doping (cm$^{-3}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Al$<em>{0.2}$Ga$</em>{0.8}$As</td>
<td>13</td>
<td>0</td>
</tr>
<tr>
<td>Al$<em>{0.75}$Ga$</em>{0.25}$As</td>
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<td>0</td>
</tr>
<tr>
<td>GaAs</td>
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<td>0</td>
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<tr>
<td>Al$<em>{0.75}$Ga$</em>{0.25}$As</td>
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<td>0</td>
</tr>
<tr>
<td>In$<em>{0.08}$Ga$</em>{0.92}$As</td>
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<td>0</td>
</tr>
<tr>
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<td>0</td>
</tr>
<tr>
<td>In$<em>{0.12}$Ga$</em>{0.88}$As</td>
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<td>0</td>
</tr>
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</tr>
<tr>
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<td>0</td>
</tr>
<tr>
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<td>0</td>
</tr>
<tr>
<td>In$<em>{0.15}$Ga$</em>{0.85}$As</td>
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<td>0</td>
</tr>
<tr>
<td>GaAs$<em>{0.4}P</em>{0.6}$</td>
<td>23</td>
<td>0</td>
</tr>
<tr>
<td>In$<em>{0.1}Ga$</em>{0.9}As</td>
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<td>0</td>
</tr>
<tr>
<td>GaAs$<em>{0.4}P</em>{0.6}$</td>
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<td>0</td>
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<tr>
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</tr>
<tr>
<td>GaAs$<em>{0.4}P</em>{0.6}$</td>
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<td>0</td>
</tr>
<tr>
<td>GaAs</td>
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</tr>
<tr>
<td>Al$<em>{0.75}$Ga$</em>{0.25}$As</td>
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<td>$4.5 \times 10^{17}$</td>
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<td>GaAs</td>
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</tr>
<tr>
<td>GaAs</td>
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<td>0</td>
</tr>
</tbody>
</table>

Table A.2 Layer material, thickness, and doping concentration in one stage (defined as active region + Bragg reflector + injector) starting from the injection barrier for the improved 7.9 $\mu$m QCL. The doping profile gives rise to a sheet density of $4.0 \times 10^{11}$ cm$^{-2}$ per stage.
The improved 7.9 µm QCL was simulated, and its electric field vs current density characteristics is shown in Fig. A.7. Compared to Fig. A.5, the leakage current $J_{\text{cont}}$ is indeed greatly reduced in the improved design, and becomes the smallest component in the total current. The current component $J_{\text{up}}$ becomes the most dominant contribution in the total current, which means an improved injection efficiency. However, similar to Fig. A.5, the sum of the current components through the lower lasing level ($J_{\text{low}}$), the active ground level ($J_{\text{gnd}}$), and the injector states ($J_{\text{inj}}$) still constitutes a non-negligible portion of the total current. Actually, these currents can never be eliminated in a QCL structure, but may be minimized in a good QCL design.

![Figure A.7 Electric field vs current density at the lattice temperature of 80 K, without the X-valley transport, for the improved 7.9 µm QCL. Electronic states used in the simulation were obtained from the self-consistent Schrödinger-Poisson solver, as described in Chapter 2. The current components are defined in Fig. A.4, and $J_{\text{tot}}$ is the sum of all the current components.](image)

The modal gain as a function of the current density is shown in Fig. A.8 at the lattice temperature of 80 K, without the X-valley transport, for the improved 7.9 µm QCL. The modal gain was calculated using the same waveguide parameters as in Sec. A.1.1, with $L_p = 59.2$ nm and $\langle z_{32} \rangle = 1.7$ nm. With a total waveguide loss of 20 cm$^{-1}$, the threshold current density was found
Figure A.8 Modal gain vs current density at the lattice temperature of 80 K, without the X-valley transport, for the improved 7.9 µm QCL. Electronic states used in the simulation were obtained from the self-consistent Schrödinger-Poisson solver, as described in Chapter 2. The dash-dot horizontal line denotes the total waveguide loss (the waveguide was designed by Mithun D’Souza). The intersection of the loss line with the modal-gain linear curve identifies the threshold current density.

to be about 7 kA/cm² (smaller than 10 kA/cm² obtained for the initial 7.9 µm QCL), due to the reduced Γ-continuum leakage and the lower doping density.

In summary, the performance of two QCL designs that both emit at 7.9 µm were simulated using the Monte Carlo simulator developed in Chapter 2. Quantification of various current components in the initial QCL demonstrated that the Γ-continuum leakage current dominates in the total current, leading to a high threshold current density. With a reduced wavefunction overlap between the injector states and the next-stage Γ-continuum states, the improved design indeed shows a much lower Γ-continuum leakage current, and also a lower threshold current density.