TUNABLE MID-INFRARED QUANTUM CASCADE LASERS

MENG BO

School of Electrical & Electronic Engineering
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School of Electrical & Electronic Engineering

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Abstract

The tremendous developments of quantum cascade lasers (QCLs) have proven themselves as powerful tools for spectroscopy, homeland security, thermal imaging, and free-space communication applications. Compact broadly tunable QCLs in both mid-infrared (3-24 μm) and terahertz (1.2-5 THz) spectrum regions are especially important for high sensitivity spectroscopy due to their intrinsic narrow linewidth, high output power, robustness, and versatile emission wavelength designability. To achieve single-mode emission from QCLs, a number of schemes have been employed to exploit the broad gain spectrum of QCLs, e.g. external cavity QCLs (EC-QCLs), distributed feedback QCLs (DFB-QCLs), DFB QCL array, and sampled grating QCLs (SG-QCLs) etc. However, the above approaches either suffer from complex fabrication processes, e.g. InP regrowth and e-beam lithography, or reduced robustness due to the mechanically moving parts. Monolithic single-mode QCLs with broad tuning range and simple fabrication process through photolithography are thus necessary for both real-life applications and laboratory researches. Therefore, the main objective of this thesis is to develop novel tunable single-mode QCLs with easy fabrication and high performances.

In this thesis, first, we have proposed and experimentally demonstrated compact tunable single-mode QCLs based on slot waveguide tunable structure at wavelength of ~10 μm. The slot-QCLs demonstrates a tuning range of 77 cm⁻¹, which corresponds to ~7.8 % of relative tuning, while maintaining ~20 dB side mode suppression ratio (SMSR) within the whole tuning range. Compared with DFB-QCLs, the broader tuning range together with significantly simplified fabrication process makes slot-QCLs better candidates for high resolution spectroscopy.

To further increase the wavelength modulation speed, we have also proposed and investigated tunable single-mode mid-infrared quantum cascade lasers based on surface-
acoustic-wave (SAW) modulation mechanism. The air-waveguide and surface plasmon waveguide structures with two-section active regions were proposed, together with Zinc Oxide (ZnO) thin film deposited on top of these devices to enhance the piezoelectricity of the materials. Coupling coefficients of ~2.5 cm\(^{-1}\) were calculated for both waveguide structures, showing the possibility of achieving tunable single-mode emission by using SAW modulation.

Furthermore, to improve the single-mode QCLs performances in terms of power and modulation bandwidth, we have studied the high modulation bandwidth injection-locked single-mode QCLs. Mid-infrared QCLs with wavelength of 4.6 \(\mu\)m and 9 \(\mu\)m were investigated for comparison. Enhanced modulation bandwidths of ~30 GHz and ~70 GHz were obtained for 4.6 \(\mu\)m and 9 \(\mu\)m, respectively, under a 5-dB optical injection ratio, showing threefold increases of modulation bandwidth for both wavelengths. These injection-locked QCLs are expected to be important components in mid-infrared free space communications.
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Chapter 1
Introduction

1.1 Motivations

With astonishing insight, the great Maxwell, for the first time ever, revealed the hidden side of the nature to the entire human being. The elegant Maxwell’s equations amazingly describe the macroscopic interactions between electrons and photons, paving the way for modern physics and technology, e.g., theory of relativity, quantum mechanism and modern telecommunications. The summit of Maxwell’s equations is the prediction of electromagnetic (EM) wave. From today’s point of view, according to wavelength, this EM spectrum can be categorized into gamma rays, X-rays, ultraviolet, visible light, infrared radiation, microwave and radio wave. The corresponding wavelength range for each category is specified in Fig. 1-1.

![EM Spectrum Diagram](image)

Fig. 1-1: Relative positions of different radiations in the EM spectrum
The infrared (IR) radiation, though was known since 1800, did not obtained much attention until the invention of semiconductor laser in the earlier of 1960s. The IR radiation can further be subcategorized into three regions: near-infrared (NIR) (0.71-2.5 μm), mid-infrared (MIR) (2.5-25 μm), and far-infrared (FIR or THz) (2.5 μm-1 mm). The NIR radiation region can be readily accessed by using laser diode consisting of direct bandgap semiconductors, owing to the genius heterojunction structure demonstrated by Zhores Alferov [1]. Now, diode lasers with several hundreds of watts continuous wave (CW) power are being provided by industrial companies in the market. Due to their high performance, diode lasers have become standard light sources for telecommunication, spectroscopy, astronomy, medicine, and agriculture applications in labs and fabs.

At the same time, mid-infrared radiation finds its important applications in the spectroscopy of toxic gas [2], free space communications (FSO) [3], and directional infrared counter measures (DIRCM) [4]. Almost forty years have passed since the first demonstration of laser-based spectroscopy. Up to now toxic gas concentrations in the parts-per-million (ppm) to parts-per-trillion (ppt) can be effectively characterized using a variety of spectroscopic approaches, e. g., long pass absorption [5], cavity ringdown spectroscopy [6], photoacoustic spectroscopy [7], quartz-enhanced photoacoustic spectroscopy [8], and Faraday rotation spectroscopy [9]. Mid-infrared radiation is also a good choice for FSO, thanks to the atmospheric transmission windows, i. e., 3-5 μm and 8-12 μm, and the reduced Mie-scattering due to the longer wavelength [10]. Equipped with directional and powerful mid-infrared emitters, the quantum cascade lasers (QCLs)-based DIRCM system is able to automatically detect and steer the coming infrared seeking missiles thus providing the shield for various aircrafts in the battlefields. Among mid-infrared QCLs, broadly tunable single-mode mid-infrared QCLs have drawn special attention due to their broad tunability [11] and narrow linewidth [12-13]. Today, broadly tunable mid-infrared QCLs have become important mid-infrared radiation sources for multi-gas and high sensitivity spectroscopies [8, 14-15].

THz radiation is the spectrum range between mid-infrared radiation and microwave; thus, it shows some of the properties of these neighboring EM spectra. Because THz can
penetrate multiple non-conducting materials, with penetration depth of several millimeters, a variety of application will benefit from using THz radiation. Typical applications include astronomy research, security surveillance, remote chemical sensing and spectroscopy, concealed military weapon detection, medical imaging, and industrial manufacturing monitoring [16-17]. However, there is a large room for exploiting the THz radiation, due to the lack of effective THz radiation sources and corresponding detectors. The advent and recent developments of THz quantum cascade lasers (THz-QCLs) have significantly facilitate the THz applications in various fields [18] and further impact the THz-based research as THz-QCLs work towards higher temperature. Like broadly tunable mid-infrared QCLs, broadly tunable THz QCLs [19] are also highly desirable single-mode THz sources as local oscillators for high sensitivity heterodyne detections [20-21].

In summary, the broadly tunable QCLs at both mid-infrared and THz regions have been proven to be important radiation sources for high sensitivity spectroscopies, industrial monitoring, and stand-off explosives detection; meanwhile they would open new prospects in these fields during the developments.

1.2 Scope and organization of this thesis

Given the high performance of mid-infrared QCLs, the objective of this thesis project is set to carry out both comprehensive experimental and theoretical investigations on the novel tuning schemes for improving the performance of broadly tunable mid-infrared single-mode QCLs

Common approaches to achieve tunable single-mode emission from mid-infrared QCLs are the external cavity QCLs (EC-QCLs) [11], distributed feedback QCLs array (DFB-QCLs array) [22] and sampled grating QCLs (SG-QCLs) [23]. However, each approach has its pros and cons. Among different tuning schemes, the EC-QCLs demonstrated the broadest tuning range reported, with maximum tuning range corresponding to 40% of the centered wavelength [24]. However, the external components make the EC-QCLs vibration sensitive and expensive. On the other hand, the DFB-QCLs array avoid the
external components by incorporating several DFB-QCLs centered at different wavelengths onto a single chip, thus significantly facilitate the system integration, yet additional components are needed to combine the outputs from the DFB-QCLs array in the far field. Recently, the SG-QCLs were employed to exploit the broad gain bandwidth of QCLs. Though the SG-QCLs demonstrated the broadest tuning range from a single device, the InP regrowth process seriously complicates the fabrication process. Thus, new tuning schemes are needed to overcome the challenges imposed by the previous methods.

In this thesis, firstly, to achieve broadly tunable single-mode emission from monolithic QCLs, we have experimentally and theoretically investigated the tunable QCLs based on slot waveguide structure [25]. Secondly, to increase the tuning speed, DFB-QCLs based on surface acoustic wave (SAW) modulation were theoretically studied [26]. Finally, the injection locked QCLs were theoretically investigated to enhance the performance of the single-mode QCLs in terms of output power and dynamic modulation bandwidths [27]. The details of each study will be presented in the following chapters. All the corresponding experimental works in different chapters were performed within Nanyang Technological University (NTU) Singapore.

The works in different chapters are organized as:

In Chapter I, firstly, the applications of mid-infrared radiation is generally introduced followed by a brief introduction of various competing mid-infrared coherent sources. The breakthroughs in the QCLs development history will be reviewed at the last of this chapter, with special emphasis on the first demonstration of QCL.

In Chapter II, the working principle of mid-infrared QCLs is presented.

In Chapter III, the device processing for electroluminescence (EL) mesa, and both pulsed and continuous wave (CW) operation QCLs are described. The corresponding measurement technique is presented following each processing method.

In Chapter IV, novel tunable QCLs based on slot waveguide structures are experimentally demonstrated and theoretically investigated.

In Chapter V, novel tunable QCLs based on travelling surface acoustic wave (SAW)
5

approach is theoretically studied.
In Chapter VI, the injection-locking tuning scheme is introduced, followed by the theoretical investigation of injection-locked QCL in the remaining of the chapter.
In Chapter VII, all the results are summarized and the future works are given.

1.3 Competing mid-infrared coherent sources

Though tunable single-mode NIR radiation sources have been employed in a number of spectroscopy methods, sensitivities of these systems are limited by the molecular absorption lines of interested gas molecules. Because the absorption can be 30-300 stronger for MID-IR than those in NIR, making MID-IR region a perfect choice for high sensitivity toxic gases measurements. Absorption lines of typical gases are shown in Fig. 1-2.
1.3.1 Lead salt diode lasers

The lead salt-based diode lasers were demonstrated a few years (1964 by J. F. Butler [28]) after the first demonstration of lasers. Since the invention, these lasers have developed to cover wavelengths from 3 to 30 μm. The carrier combination is realized through homojunction structure, with lead-based IV-VI materials such as PbS, PbSe, and PbTe acting as the barrier and the ternary or quaternary compounds such as PbₓSn₁-xTe and PbₓEu₁-xSe₁-yTe₁-y forming the active region for the laser action. Because the effective masses for conduction and valence bands of these materials are similar, it seems that the lead-based diode lasers favor high temperature operation, due to the small Auger recombination rate. However, the very small intrinsic bandgap $E_g$ greatly limits the high temperature performance of the lead-based diode lasers. Now, the reported record for pulsed operation lead diode laser is 333 K [29], and continuous wave (CW) device is 223 K [30]. The lasing wavelength of these devices can be varied by changing the constituents during growth, or changing the device working temperature after
packaging. Tuning range ~100 cm\(^{-1}\) is obtainable through temperature tuning. Fine tuning ~1-2 cm\(^{-1}\) can be achieved by varying the injection current of the device before the lasing mode jumps to adjacent longitudinal mode. Typical linewidth (full-width at half maximum (FWHM)) from Pb-salt diode lasers were measured to be 0.6-25 MHz, which varies dramatically with outer vibrations [31]. Though spectrometry employing these sources shows amazing sensitivity, the low working temperature, large beam divergence and astigmatism, and relatively low CW output power severely limit the development of these lasers and the industrial markets.

1.3.2 Antimonide-based semiconductor lasers

1.3.2.1 Type-I quantum well diode lasers

The active region of type-I quantum well diode lasers consist of GaInAsSb quantum wells (QWs) confined by AlGa(In)AsSb barrier grown on GaSb substrate. The emission wavelength ranging from ~2-3 \(\mu\)m is available from these lasers by varying the concentration of In element in the QWs. Even longer wavelength is achievable by incorporating the In element into the barriers materials. Employing the quinary AlGaInAsSb alloy as waveguide and barrier materials leads to a stronger hole confinement, thus an improved laser performance. Room temperature (RT) CW operation type-I diode laser at 3 \(\mu\)m has been demonstrated by Shterengas et al. with output power over 300 mW and peak power-conversion efficiency around 8\% [32]. Optimization of the waveguide structure can also lead to an improvement on the laser performance. Reducing the waveguide width from 1470 to 470 nm, lasers with quaternary waveguides show 200 mW CW output power at RT, demonstrated by Hosoda et al. [33]. Type-I GaSb laser with wavelength of 3.73 \(\mu\)m has been demonstrated by Vizbaras et al. [34]. The difference between type-I and type-II GaSb-based diode lasers are illustrated in Fig. 1-3.
1.3.2.2 Type-II “W” quantum well diode lasers

Because of the fact that the laser transition in Type-I material occurs at the same quantum well, i.e. GaInAsSb QWs, the wavelength from this material system is limited up to 3 μm. To access the longer wavelength, separating the electrons and holes confinement seems to be an ideal approach. With this idea, type-II diode lasers utilizing the broken gap band materials (e.g. InAs and Ga(In)Sb) are proposed by Grein et al.\cite{35}. This material system leads to partial separation of the electrons and holes and a reduced Auger coupling strength. Meyer et al. later proposed and demonstrated an improved active region design, later called “W” quantum well, to combine the advantages of excellent optical and electrical confinement, strong electrons and holes overlap, suppressed Auger recombination rate, and two dimensional dispersion for both electrons and holes, which leads to narrower gain spectrum \cite{36}. Later experiments show maximum CW operation up to 218 K and pulsed operation up to 317 K by Canedy et al. using “W” quantum wells \cite{37}.
1.3.3 Interband cascade lasers

Similar to QCLs, interband cascade lasers (ICLs) employ a cascade periodic structure and resonant tunneling to achieve population inversion between the lasing states. However, in this case, the radiative transition happens between the electron state in the conductance band and the hole state in the valence band. The novel ideal of ICLs was first proposed by Yang in 1995, soon after the first experimental demonstration of QCLs by Faist et al. in 1994 [38]. Meyer et al. later proposed and demonstrated an improved active region design with detailed calculated of the gain spectrum [39]. The schematic diagram of Meyer’s design is shown in Fig. 1-4. The proposed type-II ICL was based on InAs/Ga$_{1-x}$In$_x$Sb material system. Electrons from the left are injected into the InAs QW through intersubband resonant tunneling followed by a diagonal transition into the hole state in the Ga$_{1-x}$In$_x$Sb QW. Once the electrons have made the radiative transitions, they will tunnel into the adjacent Ga$_{1-x}$In$_x$Sb quantum well in ~100 ps. Finally, they will transport to the next injection region by elastic interband tunneling and be re-injected into the next upper laser state by graded InAlAsSb or InAs/AlSb superlattice (SL). The first experimental demonstration was realized by Lin et al. in 1996 with laser emission wavelength at 3.8 $\mu$m for temperature up to 170 K [40]. One year later, Vurgaftman et al. proposed the ICL active region design employing “W” QWs which leads to a much stronger overlap between the electrons in the InAs QWs and the holes in the Ga$_{1-x}$In$_x$Sb QW [41]. At 3.5 $\mu$m, RT CW output power up to 592 mW has been demonstrated by Canedy et al. with wallplug efficiency of 10.1% and beam quality factor of $M^2=3.7$ [41]. By the same group, nearly diffraction-limited output beam from ICLs were demonstrated with hundreds of miliwatt CW power [42]. Highest working temperature of 80 °C has been realized by Kim et al. with an output power over 1 mW at 80 °C [43].
1.3.4  **mid-infrared sources based on optical parametric generation**

To generate tunable and high power mid-infrared radiation, nonlinear materials were commonly used based on the optical parametric generation process. The two important generation methods are: difference frequency generation (DFG) and optical parametric oscillator (OPO). In both cases, three optical frequencies are involved: pump ($v_p$), signal ($v_s$), and idler frequency ($v_i$). In the case of DFG, the idler frequency is generated when focusing the pump and signal waves onto the nonlinear crystal, with generated optical frequency $v_i = v_p - v_s$, as shown in Fig. 1-5 (a). In the later method, the input pump frequency beam splits into two lower frequency beams designated as signal and idler waves. The parametric nature of the OPO leads to the energy conservation of the system, e.g. $v_p = v_s + v_i$ in Fig. 1-5 (b). To effectively generate the output signal, both cases have to satisfy the phase-matching condition, e.g. $\Delta k = k_p - k_s - k_i = 0$, where $\Delta k$ is called the phase mismatch.

Fig. 1-4: Schematic band diagram of type-II interband cascade laser proposed by Meyer et al with one and half periods.
1.3.4.1 Difference frequency generation

To effectively generate the idler wave, the three waves (pump, signal and idler) should travel in phase, e. g. $\Delta k = 0$. This phase-matching condition requirement can be met by using birefringent nonlinear crystals with pump (signal) wave traveling along the ordinary axis and signal (pump) wave traveling along the extraordinary axis. For wavelength below 5 $\mu$m, nonlinear crystals such as KTA, KTP and RTA are commonly used for DFG application. New materials such as AgGaS2, AgGaSe2, GaSe, and ZnGeP2 are also available for longer cut-off wavelength up to 12 $\mu$m. However, even with nonlinear crystal, the non-perfect phase-matching condition still exists. This problem can be partially overcome by quasi-phase-matching condition, which has been successfully applied to LiNbO3, LiTaO3, and KTiOPo4, leading to the important nonlinear component of PPLNs, PPLT and PPKTP. The orientation-patterned GaAs,
GaP, and ZnSe are materials of choice for longer wavelength [44]. Using a waveguide PPLN crystal, Kuma et al. demonstrated DFG of 4.8 µm mid-infrared radiation with two external-cavity diode lasers at 871 and 1064 nm. The output power is of 2 mW with conversion efficiency ~2%/W. The laser linewidth is determined to be ~2 MHz through Lamb-dip spectroscopy [45]. Phillips et al. reports a mid-infrared source tunable from 6.7 to 12.7 µm via DFG in orientation-patterned GaAs, with 1.3 mW average output power. The reported DFG process agrees well with the numerical model [46]. Stievater et al. demonstrated mid-infrared DFG in a suspended GaAs waveguide with thickness of 181 nm. A 0.4 W/A efficiency has been obtained for the nonlinear mixing in the 1.2 mm long waveguide [47]. Recently, 4H-SiC was found to be able to produce mid-infrared light through phase-matching DFG. Wang et al. shows a broadband spectrum spanning from 3.9 to 5.6 µm in 4H-SiC crystal with maximum output power of 0.2 mW [48].

1.3.4.2 Optical parametric oscillator

Similar to the case of DFG, an OPO also uses a nonlinear crystal, but with the nonlinear crystal enclosed into an optical cavity like the laser. Because of the fact that the parametric gain of generated wave has to compensate the total loss, the OPO exhibits a threshold behavior similar to the laser. The output signal is determined by the resonance condition. Two widely employed configurations are the singly resonant OPO where only one wave (signal or idler wave) resonates and doubly resonant OPO where both signal and idler waves resonate. Utilizing in-band pumping source of Nd:YVO4 laser and nonlinear medium of PPLN crystal, Sheng et al. reported the tunable idler output with wavelength ranging from 3.66 to 4.22 µm and maximum output power of 1.54 W at 3.66 µm under pumping power of 21.9 W. The optical conversion efficiency corresponds to 7.0% [49]. Stoeppler et al. demonstrated tunable mid-infrared radiation between 6.27 µm and 8.12 µm from a cascaded parametric arrangement employing ZnGeP2 (ZGP) OPO as the nonlinear medium [50]. Recently, by Gebhardt et al., ZGP has shown 27.9 kW mid-infrared peak power in a doubly resonant oscillator under the pumping of a
novel Tm:fiber laser [51]. Meanwhile, new mid-infrared nonlinear materials, e.g. HgGa2S4 [52] and CdSiP2 [53], are being exploited to further reduce the threshold and to extend the wavelength range of the OPOs.

1.4 Quantum cascade lasers

1.4.1 Interband versus intersubband

Different from interband diode lasers, which rely on the optical transitions between the conduction band and valence band, the optical transitions of QCLs occur between the conduction quantized states arising from the semiconductor heterostructures. Thus, only the electrons are involved in the optical process. The unipolar nature of QCLs makes them exhibit many different properties from diode lasers.

(a) Because the emission wavelength of QCLs is determined by the confinement energy difference of the conduction subbands, the wavelength can be tailored by engineering the thickness of each structure layer. This wavelength design ability releases QCLs from being a slave of the material bandgap and makes QCLs novels semiconductor lasers for a number of applications. By using the same material system, emission energy with up to ~40-50% of the conduction band offset can be realized.

(b) As demonstrated in Fig. 1-6, because the involved laser states have the same curvature in phase space (neglecting the nonparabolicity effect), QCLs exhibit an atomic-like joint density of state, thus a symmetric and narrow gain spectrum which is broadened by the interface roughness scattering and the interaction between the electrons and the LO phonons [54]. However, as we will show later, the gain spectrum of the actual QCLs can be designed to be very broad utilizing multiple lasing transitions.

(c) Due to the symmetric gain spectrum, the linewidth enhancement factor of QCLs is predicted to be close to zero because of the Kramers-Kronig relation [55] and thus a narrow laser linewidth is expected from QCLs [56].
Fig. 1-6: Comparison of (a) an interband optical transition and (b) an intersubband transition.

(d) The intersubband nature results in an ultrashort laser lifetime (determined by the electron-LO phonon scattering) and dephasing time, allowing fast intensity modulation without relaxation oscillation and making QCLs the only class-A lasers in solid-state lasers [57]. The latter provides QCLs with remarkable dynamic properties than diode lasers.

(e) Though the threshold current density of QCLs is larger compared with diode lasers, due to the ultrashort laser lifetime, the working temperature of QCLs is expected to be higher because of weaker temperature dependence of LO phonon scattering process than the Auger recombination process in diode lasers. The common characteristic temperature $T_0$ is ~200-300 K for high performance QCLs, with record of ~750 K under CW operation [58].

(f) In diode lasers, the catastrophic optical damage (COD) will arise due to surface nonradiative recombination and severely limit the maximum output from the lasers. This COD effect will be strongly prohibited in QCLs because of the unipolar nature of these devices, thus improving the robustness and lifetime of the lasers.

(g) The cascaded bandstructure offers QCLs the possibility of greater-than-unity internal quantum efficiency because of the fact that one electron is reinjected $N$ (number of cascades) times into the repeated period, shown in Fig. 1-7. Therefore, high output power can in principle be achieved with increased number of cascades and reduced
threshold current density.

Fig. 1-7: Schematic diagram of quantum cascade lasers working principle. Each period consists of an active region and an injector which injects electrons into the next period.

1.4.2 Active region designs of Mid-infrared QCLs

Since the first inception of QCLs in 1994 [59], many new designs have been investigated. One year after the first demonstration, Faist et al. introduced the so called ‘three-well vertical-transition active region’ design as shown in Fig. 1-8 (a), in which a very thin well is inserted between the injection barrier and the active region [60]. This scheme maximizes the electron injection efficiency into the upper laser state by increasing the spatial overlap of injector ground state and the upper laser state; meanwhile, reducing the injections into the lower laser state. Due to the enhanced injection efficiency, first time pulsed operation at room temperature was reported using this design, which showed much improved performance in wavelength of 3.5 µm [61], 4.6 µm [62], and 8 µm [63] etc.
Fig. 1-8: Schematic diagrams of bandstructures of (a) three-well vertical design, (b) superlattice design, (c) bound-to-continuum design, (d) two-phonon resonance design, (e) three-phonon resonance design, and (f) injectorless design, with the moduli squared of relevant wavefunctions shown at corresponding energy levels for each design.
In 1997, Scamarcio et al. proposed the so-called ‘superlattices active region design’ shown in Fig. 1-8 (b) where the laser action occurs between two minibands instead of subbands [64]. Advantages of such design are the much increased dipole matrix elements between laser states, enhanced carrier transport, and high power. To reduce the doping level in the injector, ‘chirped superlattice active region design’ was investigated by Tredicucci et al. in which the well thickness decreases in the direction of electron transport [65-66]. However, because the minibands take up large energy space in the band offset, the superlattice design is more suitable for longer wavelength, with high performance [67-68].

To overcome the electron extraction bottleneck, two novel designs ‘bound-to-continuum’ and ‘two-phonon resonance’ designs are studied by Faist group in 2001 [69-70] (Fig. 1-8 (c) and (d)). In the bound-to-continuum design, the radiative transition happens between a subband and a mimiband which will provide efficient carrier extraction from the lower laser state due to the fast intraband scattering. In the two-phonon resonance design, the two additional states are below the lower laser state, with each two states separated by one LO phonon energy. The energy ladder yielded a shortened effective lower laser state lifetime thus a more efficient carrier extraction. Thermal backfilling is suppressed in both designs due to the large energy separation between the injector ground state and the laser lower state. Because of the broad gain spectrum provided by bound-to-continuum design, it serves as the bases for most of the broadly tunable mid-infrared QCLs. The concentrated oscillation strength and high extraction rate make two-phonon resonance design suitable for high performance QCLs. In fact, it is with this design and improved waveguide thermal management, Beck et al. demonstrated the first RT CW operation QCLs at 9.1 µm [71]. The two-phonon resonance design has been proven to be the building block for achieving high performance QCLs. Now, ~50 % WPE for pulsed mode operation [72-73] and CW output power of 5.1 W, with WPE ~27 % were experimentally demonstrated through exploiting two-phonon resonance design [74].

In 2009, Wang et al. demonstrated the so called “three-phonon resonance” active design, which is in fact the improvement of the two-phonon design [75] (Fig. 1-8 (e)). Instead
of using a two-phonon ladder below the lower laser state, the proposed design uses a three-phonon energy ladder to further extract the electrons from the lower laser state. The calculated effective lower laser state lifetime reduces from 0.23 ps to 0.14 ps by adding another LO phonon below the lower state. The devices with the proposed design demonstrated high performance at 9 µm. By using the three-phonon resonance design, Maulini et al. demonstrated 19% WPE at room temperature at wavelength of ~7.1 µm [76].

To include more active regions periods in the same thickness, so as to enable higher output power and reduced current density, as shown in Fig. 1-8 (f), the injectorless active regions were investigated since 2001 [77]. Though the injectorless QCLs have demonstrated comparable pulsed mode performances to most injector-based QCLs, their CW performances are still far from the case of injector-based devices due to the severe thermal backfilling under CW mode operation [78-79]. The state-of-the-art mid-infrared QCLs with highest RT (288 K-298 K) CW output powers reported at the time of writing this thesis are summarized in Fig. 1-9. In recently years, long wavelength QCLs have undergone remarkable progresses (e. g. see [80-82]).

Fig. 1-9: Summary of the state-of-the-art mid-infrared QCLs with highest CW output powers at the time of writing this thesis.
1.4.3 Tunable single-mode QCLs

1.4.3.1 Distributed feedback quantum cascade lasers (DFB-QCLs)

Single mode quantum cascade lasers with high power, narrow linewidth, broad tunability are powerful tools for chemical, astronomical, physical and biological scientific researches. To achieve single-mode emission, different schemes have been proposed. Among various approaches, DFB-QCLs and EC-QCLs stand out due to their excellent performances. As first derived by Kogelnik and Shank using the coupled-mode theory, in DFB lasers [83], one important quantity describing the effectiveness of DFB grating is the coupling coefficient $\kappa$, which describes the coupling strength between the forward and backward traveling waves in the laser resonant cavity. The coupling coefficient $\kappa$ can be expressed as Eq.(1.1),

$$\kappa = \frac{\pi}{\lambda_b} n_1 + i \frac{\alpha}{2}$$  \hspace{1cm} (1.1)

In which $n_1$ is the periodical modulation amplitude of the effective index of the cavity mode, and $\alpha_1$ is the corresponding periodical loss modulation amplitude. Given the expression of the coupling coefficient, to achieve effective coupling between different modes, two methods which termed as loss-coupled and index-coupled have been demonstrated in QCLs. After the first demonstration of quantum cascade laser, Faist et al. realized the first DFB-QCL through loss-coupling scheme [84]. Top metal grating etched inside the plasmon-confining layer with depth of 250 $\mu$m was used to provide the optical feedback. Tuning range of ~60 nm was achieved by changing the temperature from 100 to ~300 K. From the same group, Gmachl et al. reported the first index-coupled DFB-QCL at ~8.5 $\mu$m in 1998 [85]. The DFB grating was fabricated into the GaInAs layer directly on the active region and regrown with 2.62 $\mu$m InP serving as the cladding layer. Continuous single-mode tuning range of 140 nm and side mode suppression ratio (SMSR) $\geq$ 30 dB was observed. Most of today’s production of DFB-QCLs are using this method due to its lower loss and higher coupling efficiency. Taking into account the interplay between the guided waveguide modes and the grating surface plasmonic mode
allows researchers to predict the grating coupling coefficient and optical loss more accurately [86].

The small tuning range caused by the change of temperature or current density greatly limits DFB-QCLs in the real field application. Integrating different grating periods on the same wafer which consists of bound-to-continuum active region have been shown to be an effective way to provide a much wider tunable range for DFB-QCLs. Wittmann et al. reported a room temperature, CW operated DFB-QCL with tunable range of 100 cm⁻¹ (7.7 - 8.3 μm) at RT and maximum working temperature up to 60 °C [87]. The broader tunable range was realized by integrating 25 different periods on the same gain medium optimized for emission wavelength of 7.9 μm. The same idea was also demonstrated by Lee et al. who etched an array of 32 different grating on the same gain medium using a two bound-to-continuum active region design centering at wavelength of 9 μm [88]. The demonstrated tuning range covers 220 cm⁻¹ around the center wavelength, accounting for 20% of the center frequency. It is shown that stable single-mode operation can be achieved either by depositing the antireflection coating on the laser facets or by increasing the coupling coefficient to reduce the laser facet effect.

To increase the peak output power from the DFB array QCLs, Rauter et al. recently demonstrated an array of master-oscillator configuration QCLs with enhanced peak output power up to 3.9 W which later was increased up to 10 W [89-90].

The far field characteristic of QCLs is limited by the strong diffraction due to the small apertures of laser chips. To overcome this problem and meanwhile get high purity of the spectrum, Bai et al. demonstrated the first photonic crystal distributed feedback quantum cascade laser (PCDFB-QCL) at 4.75 μm in 2007 [91], using 100 μm wide ridges and the same safer as the above work. The photonic crystal pattern was etched into the 100 GaInAs layer on top of the etch stop layer, with periods of the first order Λ₁ = 2.349 μm and Λ₂ = 0.783 μm for the two primary axes, resulting a full width at half maximum (FWHM) of 2.4° for the far field distribution. Two years later (2009), the same group reported the improved design of the PCDFB-QCL with 12 W output power [92]. Different from the first design, a 100 nm InP cushion layer was evaporated between the active region core and the grating. Three different periods were fabricated to testify the
designed parameters. Most recently, they demonstrated room temperature 4.36 μm PCDFB-QCLs. Single spatial, spectral modes, 34 W peak output power were also reported [93].

1.4.3.2 External cavity quantum cascade lasers (EC-QCLs)

Although DFB-QCL are good choices for some spectroscopy applications, owing to their compact volume, stable output spectrum, and continuous tuning ability, their relative small tuning range (1–2% around the center wavelength) strongly limits their application in parallel detection of chemicals with broad absorption spectrum, e.g. heavy molecules and gas mixtures, and the study of physical reaction dynamics. Compared with DFB-QCLs, EC-QCL have a much broader tuning range, though with a more sophisticated tuning mechanism. Typical EC-QCLs setup is shown in Fig. 1-10.

![Schematic diagram of the EC-QCL configuration.](image)

The first combination of quantum cascade lasers with external cavity was demonstrated by Luo et al. in 2001 [94]. The laser chip consisting of a three-quantum-well active resign was mounted in a variable-temperature liquid nitrogen dewar without antireflection coating. The Littrow scheme and anti-reflection coated window were used in the experiment setup. Tuning range of ~65 nm and ~88 nm was achieved at 80 K for
gain chips emitting at 4.5 and 5.1 \( \mu \text{m} \), respectively. The tuning range for both of the lasers dropped down to \(~23\) nm as the temperature increased to 203 K. The same group reported the improved performance of EC-QCLs in the next year (2002) [95]. Laser chips operating at wavelength of 5.1 \( \mu \text{m} \) combined with both Littrow and Littman-Metcalf configurations were examined. With depositing anti-reflection coating on one end of the chip, tuning range of 140 nm (54 cm\(^{-1}\)) and 127 nm (49 cm\(^{-1}\)) was achieved at temperature of 80 K and 243 K, respectively. The first room temperature operating EC-QCL was reported by Totschnig et al. in the same year [96]. Without the help of antireflection coating, they achieve a tuning range of 76 nm (7 cm\(^{-1}\)) using a 10.4 \( \mu \text{m} \) QCL working in pulsed mode.

The first development jump of EC-QCLs was demonstrated by Maulini et al. from Faist’s group in 2004 [97]. Using a bound-to-continuum active region design, spontaneous emission spectrum with full width at half maximum of 297 cm\(^{-1}\) was achieved in pulsed mode at room temperature. The ZnS coated front facet resulted in a 4% residual reflectivity, improving the coupling efficiency between the gain chip and the grating. The EC-QCL could be tuned over 150 cm\(^{-1}\) (1.45 \( \mu \text{m} \)), corresponding to 15% of the center wavelength. Though relative poor side mode suppression ratio (SMSR) was obtained, this was mainly due to the presence of the F-P modes of the chip at the beginning of the pulse.

In 2005, the same author demonstrated the first TEC cooled CW operation EC-QCL at wavelength of 5.15 \( \mu \text{m} \) based on a bound-to-continuum active region design [98]. The device shows a single mode tuning range of more than 140 cm\(^{-1}\) with power over 10 mW over 100 cm\(^{-1}\). By using a heterogeneous active region design based on two bound-to-continuum designs, Maulini et al. realized tunable EC-QCL covering wavelength from 8.2 to 10.4 \( \mu \text{m} \), corresponding to 265 cm\(^{-1}\) [99]. Mohan et al. demonstrated the first room temperature operation CW EC-QCL with tuning range of 120 cm\(^{-1}\) from 7.96 to 8.84 \( \mu \text{m} \) and 1.5 mW average output power [100]. Using a heterogeneous gain chip consisting of two bound-to-continuum designs, Wittmann et al. reported CW mode tuning of 172 cm\(^{-1}\) with over 20 mW output power at RT [101]. By using only one active region based on dual-upper-state design, Dougakiuchi et al. demonstrated tuning ranges
of 321 cm$^{-1}$ under pulsed mode operation and 248 cm$^{-1}$ under CW operation [102].

Using a symmetric active region arrangement of five different wavelengths, Hugi et al. realized a flat increased gain spectrum in the range of $\sim 7.5 - \sim 11.5$ μm [24]. The whole structure was grown by MBE, with 74 stages, and fabricated into buried heterostructure fashion. Mounded in conventional Littrow external cavity configuration, the gain chip coated with bilayer dielectric AR coating could be tuned in access of 432 cm$^{-1}$, from 7.6 μm-11.4 μm (39 % of the center wavelength), with 1 W maximum peak output power in pulsed mode, and 15 mW average output power.

### 1.4.3.3 Vernier-effect-based QCLs

Though remarkable tuning ranges have been demonstrated using EC-QCLs, the mechanical parts of the EC-QCLs make them vibration sensitive and expensive. One of the approaches to overcome these limitations of DFB-array QCLs and EC-QCLs is to utilize Vernier-effect tuning mechanism, based on which the coupled-cavity QCLs [103-104] and the SG-QCLs [23, 105] were proposed. Using coupled-cavity QCLs, Fuchs et al. demonstrated a quasi-continuous single-mode emission range of 242 nm between 8.394 and 8.785 μm [104]. Recently, around 500 nm continuous range was demonstrated at wavelength $\sim 4.6$ μm using SG-QCLs [105].
Chapter 2

Theory of quantum cascade lasers

In this chapter I will present the theoretical background necessary for understanding the optical process in mid-infrared QCLs. The chapter starts with the discussion of the electronic states in semiconductor heterostructures which are the building blocks for QCLs, followed by the radiative and non-radiative transitions between subbands confined in the quantum wells, and introduction of standard rate equations model for modeling the basic property of QCLs.

2.1 Electronic states in semiconductor heterostructures

The active regions of QCLs are planar heterostructures formed by alternating semiconductor lattice matched or strained materials (e.g. InGaAs/InAlAs). The conduction band edge profile along the growth direction $E_c(z)$ of this heterostructure is designed to generate confined quantum states for optical radiative transitions. Though multiple approaches (e.g. first principle and density function) can provide accurate description of the band structure of material, envelop function approximation is more popular for simulating semiconductor heterostructure electronic states due to its high accuracy and efficiency [106].

Under this formalism, the actual wavefunction consists of a slowly varying envelop function and a fast varying Bloch function. The wavefunction can be expressed as

$$\psi(r) = \sum_n F_n(r)u_{n,k=0}(r)$$  \hspace{1cm} (2.1)

Where $F_n(r)$ is the envelop function, $u_{n,k=0}(r)$ is the Bloch function, and $n$ is the subband index. Because the lattice constant of the considered material system is quite close, especially for the lattice matched materials, we assume a constant Bloch function for the material system. Given the translation invariance in the two dimensional plane of the heterostructure layer, the envelop function can be written as
where $F_n(r) = \frac{1}{\sqrt{S}} e^{i(k_x x + k_y y)} \chi_n(z) = \frac{1}{\sqrt{S}} e^{i\kappa_y r} \chi_n(z)$

(2.2)

where $S$ is the area of the heterostructure layer, $k_x(k_x, k_y)$ is the in-plane wavevector for the wavefunction, and $\chi_n(z)$ is the $n$th order envelop function. To calculate the electronic states in the heterostructure, most cases use eight band model to take into account the heavy-hole, light-hole, and the split-off valence bands. For efficient modeling, an effective two-band model is used for this thesis which reads [107]

$$\left[ \frac{\hbar^2}{2} \frac{d}{dz} \frac{1}{m^*(E, z)} \frac{d}{dz} + V(z) \right] \chi_{c}(z) = E \chi_{c}(z)$$

(2.3)

where, taking into account the effect of the valence band, the effective mass is

$$m^*(E, z) = m^*(z) \left\{ 1 + \frac{E-V(z)}{E_{s, eff}} \right\}$$

(2.4)

where the effective band gap is related to the non-parabolicity coefficient $\gamma$ by [108]

$$E_{s, eff} = \frac{\hbar^2}{2 \gamma m^*(z)}$$

(2.5)

Boundary conditions are introduced to solve Eq.(2.3) [106]

$$\chi_{c}^{A}(z) = \chi_{c}^{B}(z)$$

(2.6)

$$\frac{1}{m_{A}^*(E, z)} \frac{d \chi_{c}^{A}(z)}{dz} = \frac{1}{m_{B}^*(E, z)} \frac{d \chi_{c}^{B}(z)}{dz}$$

(2.7)

which is introduced by Bastard. Neglecting the in-plane momentum, i.e. at the center of the Brillouin zone, the complete envelop function under effective two-band model reads

$$\psi = \chi_{c}(z) u_{c, k=0}(r) + \chi_{v}(z) u_{v, k=0}(r)$$

(2.8)

where $u_{c, k=0}(r)$ and $u_{v, k=0}(r)$ are the Bloch functions for the conduction and valence bands, respectively. In order to normalize the conduction band wavefunctions, the effect of the valence band has to be taken into account. The normalization of conduction band wavefunctions including the effect of nonparabolicity is given by [108]
\[
\langle \chi_c(z) \rangle 1 + \frac{E - V(z)}{E - V(z) + E_{s, \text{off}}(z)} |\chi_c(z)\rangle = 1
\] (2.9)

For the calculation of the optical dipole matrix element which is the key parameter determining the coupling strength between the involved quantum states, the effect of the valence band leads to [108]

\[
z_{ij} = \frac{\hbar}{2(E_i - E_j)} \langle \chi_{c,i} | p_z \frac{1}{m^*(E_i, z)} + \frac{1}{m^*(E_j, z)} p_z | \chi_{c,j} \rangle
\] (2.10)

where \( p_z = -i\hbar (\partial / \partial z) \) is the momentum operator in the growth direction.

To avoid the space-charge formation within the QCLs structure and reduce the impurity scattering, the injector is doped with sheet density of \( \sim 1 \text{e}^{11} \text{cm}^2 \), leading to the generation of Hartree potential \( V_H(z) \), which has to be considered for accurate QCLs bandstructure modeling. The \( V_H(z) \) and the doping can be related by the Possion equation:

\[
\frac{d}{dz} \left[ \epsilon(z) \frac{dV_H(z)}{dz} \right] = -\rho(z)
\] (2.11)

where \( \epsilon(z) \) is the dielectric constant profile of the material system and \( \rho(z) \) is the sheet carrier density distribution which is expressed as:

\[
\rho(z) = q_0 \left[ N_{D}^*(z) - n(z) \right]
\] (2.12)

where \( q_0 \) is the electron charge, \( N_{D}^*(z) \) is the ionized dopant profile, and \( n(z) \) is the conduction band electron distribution, given by

\[
n(z) = \sum_i n_i |\chi_i(z)|^2
\] (2.13)

where \( n_i \) is the sheet electron density in the \( i^{th} \) subband in the structure. One efficient and good approximation is to assume a thermal distribution of electron in one period of the active region, with common Fermi energy, which can be evaluated using the charge neutrality in one period of active region, i.e.:

\[
\sum_i n_i = \sum_i \int D_i(E) f(E)dE \quad (\text{sheet density of electrons})
\]

\[
\int N_{D}^*(z)dz = n_d \quad (\text{sheet density of dopants})
\] (2.14)
Where the density of states is 

\[ D_i(E) = m^* (E) / (\pi \hbar^2) \cdot \theta(E - E_i) \]

due to the two dimensional confinement, and 

\[ f(E) = (1 + \exp((E - \mu) / kT))^{-1} \]

is the Fermi-Dirac distribution given a common Fermi level \( \mu \) measured from the ground state of each period. The complete potential of the active region is the sum of the material conduction band edge, external potential \( \Delta V(z) \) resulted from the electric bias and the Hartree potential:

\[ V(z) = V_z(z) + \Delta V(z) + V_\mu(z) \]  

(2.15)

Accurate modeling requires iteratively solving Eqs. (2.3) and (2.11) until a steady state is reached.

### 2.2 Intersubband radiative transitions and optical gain

Governed by the theory of quantum electrodynamics, the optical transitions rate between two subbands \( (E_i, k_i) \) and \( (E_f, k_f) \) within the QCLs structure is determined by Fermi’s golden rule [109]:

\[ W_{if} = \frac{2\pi}{\hbar} \left| \langle \psi_f | H'(r) | \psi_i \rangle \right|^2 \delta(E_f - E_i \pm \hbar \omega_f) \]  

(2.16)

where \( | \psi_{i(f)} \rangle \) is the product state of the conduction band confined quantum states, \( \omega_f = (E_f - E_i) / \hbar \) is the angular frequency, and the photon states and \( H' \) is the interaction Hamiltonian expressed as

\[ H' = -\frac{e}{m} A \cdot p \]  

(2.17)

where \( A \) is the Lorentz-gauge vector potential related to the polarized electromagnetic (EM) wave and \( p \) is the angular momentum operator. The \( A \) can be quantized using raising and lowering operators \( a^\dagger \) and \( a \), like the case of the harmonic oscillator as

\[ A = \sqrt{\frac{\hbar}{2\varepsilon_0 V}} \hat{e}_\sigma \left[ a^\dagger e^{-iqr} + a e^{iqr} \right] \]  

(2.18)

In the above formula, \( V \) is the volume of the field cavity, \( \varepsilon \) is the material permittivity, \( r \) is the position, \( q \) is the photon wavevector, and \( \hat{e}_\sigma \) is the field polarization vector. The initial and final electron states can be expressed using Eq. (2.2) in real space. Inserting Eqs. (2.2), (2.17) and (2.18) into (2.16) leads to the following expressions for
spontaneous and stimulated transition rates:

\[ W_{sp} = \frac{\pi e^2\omega}{eV} |\hat{\vec{e}}_\sigma \cdot \hat{\vec{Z}}| z_{gf}^2 \delta(E_f - E_i - h\omega_g) \]  

(2.19)

\[ W_{st} = \frac{\pi e^2\omega}{eV} |\hat{\vec{e}}_\sigma \cdot \hat{\vec{Z}}| z_{gf}^2 \delta(E_f - E_i \pm h\omega_g) n_{ph,\sigma} \]  

(2.20)

where \( z_{gf} \) is the dipole matrix element along the growth direction, and \( n_{ph,\sigma} \) is the mode photon number.

For calculation of spontaneous rate, all the photon modes and polarizations have to be included. In the momentum \( q \) space and for cavity size much larger than the wavelength, the mode number in a differential volume is

\[ \rho(q)d^3q = V\frac{q^2dq\sin\theta d\phi d\nu}{8\pi^3} \]  

(2.21)

For spontaneous emission rate between subbands, one needs to sum over all modes and polarizations. The expression can be obtained as

\[ W_{sp} = \frac{e^2\omega}{3\pi\epsilon_0hc^3} |z_{gf}|^2 = \frac{e^2\omega}{6\pi m^*\epsilon_0c^3} f_{gf}^2 \]  

(2.22)

where \( f_{gf} = 2m^*(E_f - E_i) |z_{gf}|^2 / \hbar^2 \) is the scaled oscillator strength and it obeys the famous sum rule \( \sum_{f \neq i} f_{gf} = 1 \) and \( \epsilon_0 \) is the vacuum permittivity. The unscaled oscillator strength \( f_{gf,\text{unscaled}} = m_0 / m^* f_{gf} \) is also useful in directly comparing the transition strength between different material systems due to the sum rule \( \sum_{f \neq i} f_{gf} = m_0 / m^* \) [108]. Thus, it is preferred to choose a small band gap material so as to increase the oscillator strength [110-111].

For the intersubband transition, the stimulated emission rate can be related to the spontaneous emission rate by [109]

\[ W_{st} = \frac{3\lambda_{gf}^2I_v}{8\pi\hbar\nu n^2} L(\nu) \]  

(2.23)

where \( I_v \) is the mode intensity at frequency of \( \nu \) and \( L(\nu) \) is the normalized Lorentian lineshape function taking into account the energy broadening due to scattering effects and is given by
\[ L(v) = \frac{(\Delta v/2\pi)}{(v-v_0)^2 + (\Delta v/2)^2} \]  
\[(2.24)\]

where \(\Delta v\) is the full width at half maximum (FWHM) around the center frequency \(v_0\). For a single cavity mode, the optical gain is defined as the relative increase of mode intensity per unit length as it propagates through the gain medium: \(dI_x/dx = g(v)I_x\), where \(x\) is defined as the propagation direction. Given that \(g(v) = \hbar \omega (N_f - N_i)W_s/I_v\), where \(N_f\) and \(N_i\) are the three-dimensional electron densities in the final and initial states, respectively, the optical gain is determined as

\[ g(v) = \frac{\Delta Ne^2 v_0 |z_f|^2}{\hbar \epsilon_c \epsilon_0 L(v)} \]  
\[(2.25)\]

where \(\Delta N = N_f - N_i\) is the population inversion between the interaction states. Inserting Eq. (2.24) into (2.25), the maximum gain at center frequency is

\[ g(v_0) = \frac{2e^2 v_0 |z_f|^2}{\hbar c \epsilon_0 \Delta v} \Delta N \]  
\[(2.26)\]

Using the above expression, the optical gain per period within QCLs structure can thus be expressed as

\[ g(v_0) = \frac{2e^2 v_0 |z_f|^2}{\hbar c \epsilon_0 \Delta v L_p} \Delta N_{2D} \]  
\[(2.27)\]

where \(L_p\) is the period length of the active region and \(\Delta N_{2D}\) is the sheet doping density. The gain expression is equivalent to [60]

\[ g(v_0) = \frac{2\pi e^2 |z_f|^2}{\epsilon_0 n_0 \lambda_0 \gamma_f} \Delta N_{2D} \]  
\[(2.28)\]

where wavelength \(\lambda_0 = h c/(E_f - E_i)\), and \(\gamma_f\) is the half width at half maximum of energy broadening. The modal gain which takes into account of the mode volume is given by \(g_M(v_0) = g(v_0)\Gamma\) where \(\Gamma\) is the mode overlap factor of the optical eigenmode within the cavity. The useful gain cross section \(g_c(v_0)\) and differential gain \(g_c'(v_0)\) are related by the model gain through \(g_d(v_0)J = g_c(v_0)\Delta N_{2D} = g_M(v_0)\), where \(J\) (kA/cm²) is the current density injected into the active region.
2.3 Intersubband relaxation time

The population inversion within QCLs relies on the careful design of the subband lifetimes which are dominated by various nonradiative intersubband transitions: acoustic and optical phonon scattering, interface roughness scattering, impurity scattering and electron-electron scattering etc. For subband energy separation greater than the material optical phonon energy $\hbar \omega_{LO}$, the dominant scattering mechanism is the electron-LO phonon scattering [112-113]. The schematic of intra and intersubband LO phonon emission is shown in Fig. 2-1. For our current device calculations, we only consider this scattering mechanism. We follow the approach first proposed by Ferreira and Bastard for dispersion-less bulk phonons [112]. Here we only consider $k_z = 0$ for the upper laser state, due to the low doping levels. At temperature $T = 0$, the scattering rate between initial state $\chi_i$ and final state $\chi_f$ is equal to the rate of spontaneous emission of a LO phonon:

![Fig. 2-1: (a) Intrasubband LO phonon scattering. (b) Intersubband LO phonon scattering. The initial wave-vector is $k$, the final wave-vector is $k'$, and the exchanged vector is $q$. The energy difference is one LO phonon energy $\hbar \omega_{LO}$.](image)

$$\frac{1}{\tau_{em}} = \frac{m^* e^2 \omega_{LO}}{2 \hbar^2 \varepsilon_F q_{ij}} \int dz' \int dz \chi_i(z) \chi_f(z) e^{-q_{ij} z' - z} \chi_i(z') \chi_f(z')$$

(2.29)
where $q_{gf} = \sqrt{2m'(E_i - E_f - \hbar\omega_{LO})/\hbar^2}$ is the norm of momentum exchange and $\varepsilon_P^{-1} = \varepsilon_w^{-1} - \varepsilon_s^{-1}$, where $\varepsilon_w^{-1}$ and $\varepsilon_s^{-1}$ are the high frequency and static dielectric constants, respectively. Formula (2.29) implies a lifetime of the order of a few picoseconds and thus an ultrafast transport time through the QCLs structure [114]. This ultrafast lifetime explains the lack of relaxation oscillation when QCL was under high speed modulation and makes QCLs the only class A solid state lasers.

For temperature other than zero Kelvin, the LO phonon absorption is possible. Thus, both the LO phonon absorption and emission have to be included into the lifetime calculation, leading to the total scattering rate $\tau_{gf}^{-1}$ expressed as

$$\tau_{gf}^{-1} = (1+n_{BE})\tau_{em}^{-1} + n_{BE}\tau_{abs}^{-1}$$

(2.30)

where $\tau_{abs}^{-1}$ is the LO phonon absorption rate computed using Eq. (2.29), but with $q_{gf} = \sqrt{2m'(E_i - E_f + \hbar\omega_{LO})/\hbar^2}$. The phonon distribution is also considered by including the Bose-Einstein factor given by [112]

$$n_{BE} = \frac{1}{\exp(\hbar\omega_{LO}/kT)-1}$$

(2.31)

which leads to a weak temperature dependent laser state lifetimes, and thus explains in essence the weak temperature dependence of the threshold current density of QCLs. For real devices, within one period, multiple transitions can occur from each subband. Therefore, all the final states have to be considered when calculating the lifetime of a specific initial i.e. Fig. 2-2 shows the LO phonon scattering time of a typical bound-to-continuum active region at 10 μm [97].

$$\frac{1}{\tau_i} = \sum_j \frac{1}{\tau_{gf}}$$

(2.32)
Fig. 2-2: Upper laser state lifetime of bound-to-continuum design at 10 μm [97] for temperature from 0 to 300 K.

2.4 Intersubband transition linewidth

To optimize the laser performance, the linewidth broadening between the lasing states has to be minimized because it is inversely proportional to the optical gain. Back in 1996, Campman et al. suggested that the interface roughness is the dominant scattering mechanism affecting the intersubband transition linewidth, excluding the effect of phonon and alloy scatterings [115]. The role of interface roughness scattering is further confirmed by Unama et al. through comparison of theory and experiments [116-117]. In this thesis, we follow the formula given by by Unama et al. Here, this model assumes that the roughness $h_\parallel (r)$ and $h_\parallel (r')$ at positions $r$ and $r'$ have a Gaussian autocorrelation function

$$\langle h(r)h(r') \rangle = \Delta^2 \exp \left(-\frac{|r-r'|^2}{\Lambda^2} \right)$$

(2.33)

with a mean height of $\Delta$ and a correlation length of $\Lambda$. In the case of single quantum well, the intrasubband and intersubband interface roughness scatterings are given by [116]
\[
\gamma_{\text{IFR}}^{\text{IFR}} = \frac{m^* \Delta^2 \Lambda^2}{\hbar^2} (F_{ii} - F_{ff})^2 \int_0^\infty d\theta e^{-q^2 \Delta^2 / 4} 
\]  
(2.34)

\[
\gamma_{\text{IFR}}^{\text{inter}} = \frac{m^* \Delta^2 \Lambda^2}{\hbar^2} F_{ii}^2 \int_0^\infty d\theta e^{-\bar{q}^2 \Delta^2 / 4} 
\]  
(2.35)

where \(F_{ii} = \sqrt{\left(\partial E_i / \partial L\right) \left(\partial E_i / \partial L\right)}\), \(L\) is the well width, \(E_{i(f)}\) is the confined energy of the \(i(f)\) subband, \(\theta\) is the scattering angle, and \(q\) and \(\bar{q}\) are the two-dimensional intra and inter scattering vectors, given by

\[
q^2 = \frac{4m^*}{\hbar^2} E(1 - \cos \theta) 
\]  
(2.36)

\[
\bar{q}^2 = \frac{4m^*}{\hbar^2} \left[ E + \frac{E_{\text{lo}}}{2} - \sqrt{E(E + E_{\text{lo}}) \cos \theta} \right] 
\]  
(2.37)

In QCLs structure, because the wavefunctions extend over multiple quantum wells, e.g. facing many interfaces between quantum wells and barriers, all the contributions from each interface at \(z_k\) has to be considered. As shown by Tsujina et al., the individual contribution from interface \(k\) can be calculated by the expression \(F_{ii}^k = \Delta E_c |\chi_i(z_k)|^2\), where \(\Delta E_c\) is the band offset [118]. Adding all the interfaces leads to intrasubband interface roughness scattering as

\[
\gamma_{\text{IFR}}^{\text{IFR}} = \frac{m^* \Delta^2 \Lambda^2}{2\hbar^2} \Delta E_c \sum_k \left| \chi_i(z_k) \right|^2 - \left| \chi_f(z_k) \right|^2 \int_0^\infty d\theta e^{-q^2 \Delta^2 / 4} 
\]  
(2.38)

The overall intersubband transition linewidth \(2\gamma_{\text{ff}}\) can be approximated by [54]

\[
2\gamma_{\text{ff}} \approx \gamma_{\text{IFR}}^{\text{IFR}} + \gamma_{\text{IFR}}^{\text{OPT}} 
\]  
(2.39)

where the optical phonon scattering broadening \(\gamma_{\text{IFR}}^{\text{OPT}} = \hbar / \tau_{\text{IFR}}^{\text{OPT}} \approx \hbar (1/\tau_{\text{IFR}}^{\text{ud}} + 1/\tau_{\text{IFR}}^{\text{df}})\) [119], with \(\tau_{\text{IFR}}^{\text{OPT}}\) and \(\tau_{\text{IFR}}^{\text{ud}}\) (\(\tau_{\text{IFR}}^{\text{df}}\)) being the intrasubband LO phonon scattering time, and intrasubband LO phonon scattering in the initial (final) state, respectively. Though intersubband interface roughness has little effect on the linewidth broadening, its significance is found in the laser states lifetimes as demonstrated in recent experimental and theoretical studies [120-121]. A new explanation of interface roughness scattering is presented by Khurgin, who explains the interface roughness as an inhomogeneous broadening caused by localization rather than traditional believed scattering mechanism.
The new explanation gives a new similar formula as Eq. (2.38), but with a factor of 1.6 [122].

2.5 Rate equation model

The most straightforward way to model the electron transport in QCLs structures is by using the rate equation model which provides the macroscopic physical parameters such as the threshold current and slope efficiency. The rate equation model involves a set of time differential equations describing the time evolution of the carrier populations (cm$^{-2}$ per period) in different energy states and the photon flux density (cm$^{-1}$s$^{-1}$ per period). Fig. 2-3 illustrates the energy states and lifetimes used in the rate equation model which is valid for most of the QCLs structures. The effect of state 1 is included into the lifetime calculation of state 2. The set of coupled equations are expressed as [123]

$$\frac{dn_3}{dt} = \eta \frac{J}{e} \frac{n_3}{\tau_3} - Sg_e (n_3 - n_2)$$  \hspace{1cm} (2.40)

$$\frac{dn_2}{dt} = (1 - \eta) \frac{J}{e} \frac{n_3}{\tau_{32}} + Sg_e (n_3 - n_2) - \frac{n_2 - n_2^{therm}}{\tau_2}$$  \hspace{1cm} (2.41)

Fig. 2-3: Schematic diagram of bandstructure and lifetimes involved in the rate equation model.
\[
\frac{dS}{dt} = \frac{c}{n} \left[ (g_c(n_3 - n_2) - \alpha_{tot})S + \beta \frac{n_3}{\tau_{sp}} \right]
\]  

(2.42)

Where \( \eta \) is the injection efficiency to the upper state, \( J \) is the current density, \( \tau_3 \) and \( \tau_2 \) are the lifetimes of laser states 3 and 2, \( \tau_{sp} \) is the nonradiative relaxation time between states 3 and 2, \( \tau_{tot} \) is the spontaneous lifetime, \( \beta \) is the fraction of spontaneous emission emitted into the lasing mode, \( n_2^{\text{therm}} = n_g \exp(-\Delta/kT) \) is the thermal population caused by thermal backfilling, with \( n_g \), \( \Delta \), and \( T \) represent the injector ground state population, voltage defect (energy difference between the lower laser state and the coming injector’s ground state) [124], and electronic temperature, respectively. By setting \( S = 0 \) in Eq.(2.40), an expression describing the relationship between upper state population and injection current is shown to be

\[
n_3 = \eta J \tau_3 / e
\]

(2.43)

Combining Eqs. (2.41) and (2.43), the population inversion \( \Delta n \) is derived

\[
\Delta n = \frac{J \tau_1}{e} \left( \eta \left( 1 - \frac{\tau_2}{\tau_{32}} \right) - \left( 1 - \eta \right) \frac{\tau_2}{\tau_3} \right) - n_2^{\text{therm}}
\]

(2.44)

For mid-infrared QCLs, due to the large energy spacing, the \( \eta \) is assumed to be close to unity, thus

\[
\Delta n = \frac{J \tau_1}{e} \left( 1 - \frac{\tau_2}{\tau_{32}} \right) - n_2^{\text{therm}} = \frac{J \tau_{\text{eff}}}{e} - n_2^{\text{therm}}
\]

(2.45)

where the effective upper state lifetime \( \tau_{\text{eff}} = \tau_3(1 - \tau_2 / \tau_{32}) \) is defined to quantitatively relate the population inversion to the injection current. The thermal effect is manifested in \( n_2^{\text{therm}} \) which increases with temperature. Therefore, higher temperature leads to a decrease of the population inversion thus a deteriorated laser performance. The threshold current density is determined by the condition

\[
g_c \Delta n = \alpha_{tot}
\]

(2.46)

Thus, the threshold current density \( J_{th} \) is determined as

\[
J_{th} = \frac{e}{\tau_{\text{eff}} g_c} \left( \alpha_{tot} + n_2^{\text{therm}} g_c \right)
\]

(2.47)
where $\alpha_{\text{tot}} = \alpha_m + \alpha_w + \alpha_{\text{res}}$, with $\alpha_m$, $\alpha_w$ and $\alpha_{\text{res}}$ being the mirror loss, waveguide loss, and absorption loss by resonant intersubband transition, respectively.

Because the F-P lasers are formed by two cleaved facet, each facet will cause partial transmission from the cavity. This transmission is described by the mirror loss $\alpha_m = \alpha_{m,1} + \alpha_{m,2} = -\ln(R)R_2/2L$, where $L$ is the cavity length and $R_1(R_2)$ is the front(back) facet power reflectivity. The second loss source is due to the free carrier absorption with the laser cavity and the metallic contact layers. We attributed this waveguide loss as $\alpha_w$. The third absorption path which is often ignored is the resonant intersubband transition, which is usually more pronounced at longer wavelengths due to the small energy spacing in the minibands. However, both $\alpha_w$ and $\alpha_{\text{res}}$ can be minimized by quantum design; meanwhile, $\alpha_m$ can be optimized by carefully designed optical coatings to achieve high output power.

The slope efficiency of the optical power from single facet can be computed as

$$\frac{dP}{dI} = \frac{N_p \hbar \omega \alpha_{m,1}}{e} \frac{dS}{dJ} = \frac{N_p \hbar \omega \alpha_{m,1}}{\alpha_{\text{tot}}} \frac{\tau_{\text{eff}}}{\tau_{\text{eff}} + \tau_2} = \frac{N_p \hbar \omega}{e} \eta_{\text{ext}} \eta_{\text{int}}$$

which clearly shows that the slope efficiency is proportional to the period number $N_p$, photon energy $\hbar \omega$, and the internal ($\eta_{\text{int}} = \tau_{\text{eff}} / (\tau_{\text{eff}} + \tau_2)$) and external ($\eta_{\text{ext}} = \alpha_{m,1} / \alpha_{\text{tot}}$) quantum efficiencies.

### 2.6 Resonant tunneling in QCLs

One of the main physical mechanisms governing the carrier transport in QCLs structures is the resonant tunneling through semiconductor heterostructure. For the device optimization, it is therefore very important to understand the relevant parameters which have significant impacts on the resonant tunneling. Though rate equation model can provide the population evolution in different energy states, it fails to provide the phase information between different states. The quantum mechanical analysis by the tight-binding approximation and the density matrix formalism is necessary to describe the complex tunneling process. The most important tunneling channel is the coupling
between the injector ground state and the upper laser state in the coming period. The schematic of the resonant tunneling between these two states is shown in Fig. 2-4. The detailed theoretical analysis is presented by Kazarinov and Suris, who give the formula for resonant tunneling current density as [125]

\[ J_{\text{res}} = qN_s \frac{2|\Omega|^2 \tau_\parallel}{1 + \Delta^2 \tau_\parallel^2 + 4|\Omega|^2 \tau_\parallel \tau_3} \]  

(2.49)

where \(2\hbar|\Omega|\) is the anticrossing gap between states \(|g\rangle\) and \(|3\rangle\), \(N_s\) is the sheet carrier density, \(\hbar\Delta\) is the energy detuning from resonance, and \(\tau_\parallel\) is the in-plane dephasing time which is responsible for the loss of phase between states \(|g\rangle\) and \(|3\rangle\), with a typical value of 50-100 fs. Under tight-binding approximation, the wavepacket of carriers oscillates between states \(|g\rangle\) and \(|3\rangle\), with Rabi oscillation frequency of \(\Omega_{\text{Rabi}} = \Delta/\hbar\) and dephasing time of \(\tau_3\) which is mainly determined by intrasubband scatterings.

Two resonant tunneling current regimes can be identified by varying relevant parameters

A. Weak coupling regime: \(4|\Omega|^2 \tau_\parallel \tau_3 \ll 1\). We obtain the maximum current density as

\[ J_{\text{max}} = 2eN_s |\Omega|^2 \tau_\parallel \]  

(2.50)

In this regime, as the energy broadening \(\tau_3^{-1}\) is much larger than the anticrossing gap \((4|\Omega|^2 \tau_\parallel)^{-1}\) between states \(|g\rangle\) and \(|3\rangle\), the maximum current is mainly limited by the scattering process, and most carriers will be restricted within the injectors.

B. Strong coupling regime: \(4|\Omega|^2 \tau_\parallel \tau_3 \gg 1\). This leads to the maximum current density as

\[ J_{\text{max}} = \frac{eN_s}{2\tau_3} \]  

(2.51)

In this case, the current density is limited by the upper laser state lifetime, leading to an equal distribution of electrons between states \(|g\rangle\) and \(|3\rangle\).
Fig. 2-4: Schematic diagram of the resonant tuning between injector ground state and coming laser upper state. The blue arrow indicates the possible resonant tunneling path.

2.7 Optical resonator and waveguide loss

Besides the quantum bandstructure engineering, a detailed understanding of the electromagnetic behavior is also necessary for device performance optimization, since better manipulation of electromagnetic behavior will lead to an enhanced model gain and a decreased waveguide loss. Though the slab model is often used to simulate the optical mode distribution along the growth direction, it is inefficient in modeling the two-dimensional (2D) mode distribution in the lateral direction. Today, most of the mid-infrared QCLs rely on the index guided waveguide structure (Fig. 2-5) where the optical confinement in the growth direction $\vec{z}$ is achieved by sandwiching the active region between lower refractive index InP cladding layers and in the lateral direction $\vec{x}$ by dry etching. Since both dimensions show similar length scale to the wavelength, to accurately modeling the device, it is important to simulate the 2D optical mode distribution.
Fig. 2-5: Schematic of index-guided mid-infrared waveguide structure. The active region is sandwiched between the upper and bottom InP cladding layers.

Given the translational symmetry of the waveguide structure and the TM nature due to the selection rule, it is convenient to perform the analysis using the magnetic ($\vec{H}$) field which is expressed as

$$H(x, y, z, t) = H(x, z)e^{j(\omega t - ky)}$$

(2.52)

where $k$ is the propagation constant and $\omega$ is the angular frequency. Derived from the Helmholtz equation, the eigenvalue equation for the magnetic field $\vec{H}$ is

$$\nabla \times (n^2 \nabla \times \vec{H}) - k^2 \vec{H} = 0$$

(2.53)

where $n$ is the refractive index of each layer. After obtaining the $\vec{H}$, the electric field $\vec{E}$ can be obtained using the Maxwell equations. Throughout the thesis, we used commercial 2D solver COMSOL to obtain the eigenvalues of Eq. (2.53). The refractive index of each waveguide layer is calculated by the Drude model. According to the Drude model [126], the dielectric constant $\varepsilon(\omega)$ as function of the optical frequency $\omega$ is defined as:

$$\varepsilon(\omega) = \varepsilon_\infty \left[1 - \frac{\omega_p^2 \tau^2}{1 + (\omega \tau)^2} + i \frac{\omega_p^2 \tau}{\omega (1 + (\omega \tau)^2)}\right]$$

(2.54)

where $\tau$ is the scattering time in the media, $\varepsilon_\infty$ is the high frequency dielectric constant, and $\omega_p^2 = N_e e^2 / m^* \varepsilon_\infty$ is the plasma frequency of the media.
The eigenvalue Eq. (2.53) is given by \( \lambda_{\text{eig}} = -j\beta \), leading to the mode effective refractive index \( n_{\text{eff}} = \beta/k \). Thus, the waveguide loss \( \alpha_w \) can be written as

\[
\alpha_w = -2k \Im(n_{\text{eff}}) \quad (2.55)
\]

For \( \omega \gg \omega_p \) which is the case for most mid-infrared QCLs, the waveguide loss can be approximated as

\[
\alpha_w = -2k \Im(n_{\text{eff}}) = \frac{\omega_p^3}{\omega^2 c \tau} \sqrt{\frac{\varepsilon}{\varepsilon_0}} = \frac{N \varepsilon^2 \lambda^2}{4\pi^2 m^* c^3 \tau \varepsilon_0} \sqrt{\frac{\varepsilon}{\varepsilon_{\text{core}}}} \frac{N \lambda^2}{\tau} \quad (2.56)
\]

where \( \lambda \) is the free space wavelength. Apparently, the waveguide loss increases linearly with the doping level and quadratically with the wavelength for moderate doped waveguides. Therefore, for THz QCLs, the doping level is much lower than that in the mid-infrared QCLs in order to reduce the waveguide loss. The mode confinement factor used in the modal gain calculation can be found by

\[
\Gamma = \frac{\int_{\text{active}} \vec{z} \cdot \Re(\vec{E}_i \times \vec{H}_i) d^2 \vec{r}_i}{\int_{\text{waveguide}} \vec{z} \cdot \Re(\vec{E}_i \times \vec{H}_i) d^2 \vec{r}} \quad (2.57)
\]

where \( \vec{E}_i \) and \( \vec{H}_i \) are the electric and magnetic fields components in the lateral direction.

Fig. 2-6 shows the COMSOL result of a 30 \( \mu \)m ridge structure QCL.

Fig. 2-6: Fundamental mode (TM\(_{01}\)) distribution of 30 \( \mu \)m wide ridge structure. Arrows indicate the direction of the electric field.
Chapter 3

Devices processing and characterization

Throughout this thesis, the QCLs wafers were grown by molecular beam epitaxy (MBE), given the highly demanding active region consisting of hundreds of layers with thicknesses from a few angstroms to a few nanometers. In the first section, the fabrication process for electroluminescence (EL) mesa and EL measurement are given, which can be used to characterize the growth quality and the gain bandwidth of the active region design. In the following section, the fabrication and characterization of pulsed operation QCLs is discussed. Finally, using improved thermal management design, the corresponding continuous wave (CW) QCLs fabrication and testing are presented.

3.1 Electroluminescence measurement

To fast verify if the emitting wavelength is the same as the designed value or find out the possible reasons for poor laser performances, the EL measurement is often performed with much reduced feedback from the laser cavity at different heatsink temperatures.

3.1.1 Mesa preparation

To reduce the cavity feedback effect, the circular mesa structure is often applied by wet etching which results in tilted side walls. The fabricated circular mesa is then cleaved to enhance the EL extraction. A schematic drawing of such circular mesa is shown in Fig. 3-1. The processing consists of the following steps: a) mesa patterning using photoresist (S1818), b) wet etching using acid mixture (volume ratio, HBr:HNO3:H2O=1:1:10), c) top contact patterning with contact lithography, d) top contact deposition (Ti/Au (5/200 nm)) with electron beam (e-beam) evaporator or sputter, e) liftoff by Acetone or resist
developer, f) die lapping down, g) back contact deposition (Ti/Au (5/200 nm)) by e-beam evaporator, and h) mesa cleaving. Fig. (i) shows the top view of the fabricated half-circular mesa. Fig. 3-2 shows the microscope images of the fabricated mesas.

Fig. 3-1: Flow diagram of QCL EL mesa fabrication process. The final mesa is cut in the half to enhance the EL extraction.

Fig. 3-2: Microscope images of the processed EL mesas. (a) Top view and (b) side view showing the active region.
3.1.2 Electroluminescence measurement

Because the EL radiation from intersubband transition is extremely inefficient and the blackbody radiation background at 300 K is centered at ~9 μm, highly sensitive detection technique is required to extract the signal from the strongly noisy background. In our experiment, to measure the EL spectrum, we performed step-scan mode measurement in the Fourier-transform spectrometer (FTIR) equipped with external liquid Nitrogen (LN)-cooled MCT detector. The detailed experiment schematic is shown in Fig. 3-3. The sample was loaded in the continuous liquid nitrogen (LN)-cooled cryostat where the temperature was automatically controlled by high resolution temperature controller. The mesa was driven by high power pulser which triggers all the other equipment. The pre-amplified MCT detector signal is fed into the input port of lock-in amplifier which is synchronized by the pulser. The lock-in amplifier output signal is collected by the FTIR operation software. The EL spectra of our broad band QCLs design (~10 μm) at different temperatures at shown in Fig. 3-4.

![Fig. 3-3: Schematic illustration of experiment setup for EL measurement. The FTIR is running in step scan mode.](image_url)
3.2 Pulsed QCLs fabrication and characterization

To initiate laser operation, the EL radiation has to be amplified multiple times by the gain medium. This can be achieved by incorporating the gain medium into a Fabry-Perot (F-P) cavity. In this section, the most straightforward fabrication method of F-P QCLs for pulsed operation is given.

3.2.1 Pulsed mode QCLs fabrication

The processing can be summarized into the following steps: a) ridge patterning, b) drying etching of the ridge, c) insulation layer deposition, d) top window patterning, e) top window opening, f) top contact evaporation, g) lifting-of, h) polishing, and i) back contact evaporation. The schematic diagram of fabrication flow is given in Fig. 3-5.

The ridges are patterned by conventional photolithography method (in our case, we used photoresist, SiN or SiO$_2$ as hard mask). A mixture of Cl$_2$/Ar dry etching gas recipe is used in the inductively coupled plasma reactor (ICP-RIE) to obtain ~10 µm deep ridges with vertical and smooth sidewalls. The insulation layer with thickness of around 300 nm was deposited by plasma enhanced chemical vapor deposition (PECVD). The top-
window is opened by reactive ion etching (RIE), after which the top contact of Ti/Au (10nm/200nm) is evaporated by E-Beam evaporator. The 500 μm thick substrate is then polished down, using Al₂O₃ powder, to around 200 μm thick. Afterwards, the clean room processing recipe is finished by depositing the metal contact of Ti/Au (5/200) on the back side of the wafer. The processed wafer will be cleaved, using cleaving machine, into laser bars, and then indium soldered onto the copper submounts for better heat dissipation. The bars will be wire-bonded to the gold pad for further testing using the wire-bonding machine. The fabricated F-P ridge lasers varying between 16 to 32 μm are shown in Fig. 3-6.

![Flow diagram of the pulsed mode mid-infrared F-P ridge QCLs fabrication process.](image)

Fig. 3-5: Flow diagram of the pulsed mode mid-infrared F-P ridge QCLs fabrication process.
3.2.2 Pulsed mode QCLs electrical and optical characterizations

Fig. 3-7 shows our characterization setup used for the standard power-current-voltage (LIV) curve measurement for heat sink temperatures from cryogenic to room temperature. The main electronic components of the characterization setup consist of the pulse generator, oscilloscope, thermopile detector, and continuous flow cryostat.

Fig. 3-7: Schematic diagram of the L-I-V curve measurement setup for the pulsed mode QCLs. The data acquisition is controlled by the GPIB instrument bus line.
While the pulse generator is generating voltage pulse drive to the QCLs, the electrical signals (voltage and the current) are read from the oscilloscope, which is synchronized by the trigger signal of the pulse generator. The optical power detected by the thermopile detector will be read out by the DAQ board sending the signal to the processing software afterwards. All the data collecting and the equipment controlling are accomplished by the computer-assisted data acquisition software written by Labview, through the GPIB instrument bus line. The main components for the optical characterization measurement setup are similar with the electrical setup, except that the power meter is replaced by the Fourier transform infrared spectrometer (FTIR). In essence, the FTIR is a Mach–Zehnder (M-Z) type interferometer, by analyzing the time domain interferogram with Fourier transform, the frequency domain information of the detected signal can be obtained. The setup is shown in Fig. 3-8. The obtained LIV curve and the spectrum from a previous design at 10 μm [97] are shown in Fig. 3-9. The obtained lasing wavelength is in good agreement with that in [97] and our simulations.

![Diagram](image)

**Fig. 3-8:** Schematic diagram of spectrum characterization for pulsed mode QCLs. The data acquisition is controlled by the GPIB instrument bus line.
3.3 Continuous wave (CW) QCLs fabrication and characterization

3.3.1 CW QCLs processing

Under CW operation, the QCLs will suffer from severe self-heating resulting from the huge amount of injected electrical power (~10 W–20 W for typical CW QCLs). One simplest way to reduce the electrical power injection is to reduce the width of the device. Typical width value for CW QCLs varies from 6 to 16 μm depending on the device structures. Like the case of diode lasers, gold electroplating is applied to enhance the thermal dissipation from the active region. The gold electroplating has the advantages of being compatible with the QCLs processing and facilitating the device package, e.g. preventing solder from overflow during epilayer down bonding [127]. However, the sidewall effect becomes prominent as the device width reduces due to sidewall roughness caused scattering and Si₃N₄ or SiO₂ absorption. To minimize the effect, anisotropic wet etching process is preferred to ensure smooth sidewalls [128]. Typical fabrication flow of CW fabrication process is shown in Fig. 3-10. The fabricated devices are presented in Fig. 3-11.

Fig. 3-9: (a) Typical LIV curves for QCL at ~10 μm with and without high reflection (HR) coating. (b) The lasing spectrum near roll-over point of the dynamic range.
Fig. 3-10: Flow diagram of the CW mid-infrared QCLs fabrication process.

Fig. 3-11: Fabricated CW ridge lasers. (a) Microscope image of the fabricated array. (b) SEM image of the output facet.
3.3.2 CW mode QCLs electrical and optical characterizations

For our current devices, due to the relatively small electrical injection power, the QCLs chip can be loaded into the cryostat for different temperature stabilization. The devices were driven by high voltage DC power supply, while the output power was collected by thermopile detector. Typical performance for a CW laser at 4.7 μm \[129\] is shown in Fig. 3-12. The obtained spectrum matches very well with that in \[129\] and our simulations.

Fig. 3-12:(a) CW LIV curve for 12 μm wide device with length of 2 mm at temperature of 160 K. (b) Lasing spectrum around roll-over point of the dynamic range at the same temperature.
Chapter 4

Tunable single-mode quantum cascade lasers based on slot waveguide structures

4.1 Introduction of slot waveguide QCLs

As we mentioned previously, QCLs are unipolar semiconductor lasers, making the optical transitions between the quantized states in the conduction band of semiconductor heterostructure. Through quantum bandstructure designs, QCLs covering from ~3 μm to ~230 μm wavelengths have been experimentally reported [130-132]. Because many gases have their “fingerprint regions” in the mid-infrared range, QCLs are the ideal mid-infrared radiation sources for various sensing and spectroscopy applications [2, 133-134]. For practical applications such as spectroscopy, single-mode QCLs are more advantageous owing to the much reduced laser linewidth [12].

Even though DFB-QCLs have shown attractive performance, the single-mode tuning range of a single DFB QCL is about 10 to 20 wavenumbers by temperature or DC current tunings. To overcome this limitation of DFB-QCLs, an array of DFB-QCLs with a broad tuning range has been reported [22, 135], while this method poses fabrication challenges in ensuring each ridge laser working properly and extra efforts are needed to spatially combine the beams from the array of DFB-QCLs for better collimation [136-137]. Despite the large tuning range of EC-QCLs, the mechanical parts within the EC-QCLs cavity make them vibration sensitive and comparatively expensive. One effective approach to overcome these limitations is to utilize Vernier-effect tuning mechanism, based on which the coupled-cavity QCLs and the SG-QCLs were proposed [103-104, 138-139]. Nevertheless, both approaches have their intrinsic drawbacks which severely limit their real field applications. Recently, an interesting surface acoustic wave (SAW) approach was theoretically proposed and investigated for achieving tunable single-mode
emission from QCLs [26, 140]. The SAW approach is expected to exhibit fast tuning speed for broad tuning range.

In this chapter, we present tunable single-mode two-section QCLs that utilized etched slot structures and a separately electrical pumping scheme to achieve a broad single-mode tuning range. The tuning mechanism can be explained as follows: slots in both front and back sections form two reflectors with the spacing of the reflection spectral peaks determined by the slot spacing. Lasing action occurs when the reflection peaks of the two sections overlap. A slight refractive index change of one section leads to the overlap of another two reflection peaks. As a result, tunable single-mode operation in a wide spectral range can be achieved [141].

4.2 Slot-QCLs fabrication and characterization

For the present work, a lattice matched 35-periods In$_{0.53}$Ga$_{0.47}$As/In$_{0.52}$Al$_{0.48}$As active region based on the four well bound-to-continuum design was used with a high doping [97]. The waveguide structure is similar to that in [75]. The device processing started with the fabrication of the arrays of slots. A layer of 300-nm-thick Si$_3$N$_4$ was deposited as the hard mask using plasma-enhanced chemical vapor deposition (PECVD). The slot patterns were then defined, by using optical lithography and inductively coupled plasma reactive ion etching (ICP-RIE) reactor with a Cl$_2$/Ar plasma, ~2.2 μm etched into the top cladding layer. Then ridge laser devices, 16 μm wide and 10 μm deep, were defined through ICP-RIE dry etching with a Cl$_2$/Ar plasma recipe. Laser ridges were then insulated with a 400-nm-thick Si$_3$N$_4$ layer, and Ti/Au metallization was evaporated as the top contact with e-beam evaporation. After that, the sample was thinned down to around 200 μm thick and the Ti/Au metallization bottom contact was evaporated. The front and back sections of the slot-QCLs were electrically separated by a 100-μm gap in the Ti/Au top contact layer during the lift-off process. The electrical resistance between the two sections of the device was measured to be of ~ 300 Ω, enough for good electrical separation between the two sections. Fig. 4-1 shows the microscopy image of the slot-QCL, with front section length $L_F = 974$ μm, the back section length
$L_b = 1087 \, \mu m$, and the middle insulation section length $L_M = 100 \, \mu m$. The zoom-in views show the width of the slot and the slot spacings ($L_f = 80 \, \mu m$, and $L_b = 96 \, \mu m$, where $L_f$ and $L_b$ represent the slot spacings of the front and back sections, respectively) in the two sections. On each section, 11 slots were fabricated to provide a narrow linewidth of the reflection peak while keeping a short device length. For spacings $L_f = 80 \, \mu m$, and $L_b = 96 \, \mu m$, the spectrum reflection peak spacings are $\sim 18.1 \, \text{cm}^{-1}$ and $\sim 15.1 \, \text{cm}^{-1}$, respectively. For spectral characterization, the as-cleaved devices were mounted epilide up onto the cold finger of a liquid N$_2$ flow cryostat. Spectral characterization was conducted using Fourier transform spectrometer (FTIR) at a highest resolution of 0.2 cm$^{-1}$ with a calibrated room temperature (RT) HgCdTe detector.

Fig. 4-1: (a) Optical microscopy image of a typical two-section slot-QCL. The total lengths are 974 $\mu m$ and 1087 $\mu m$ for the front and the back sections, respectively. The two sections are separated by a 100 $\mu m$ gap in the top metal contact layer. The etched slots are 5.5 $\mu m$ wide and 2.2 $\mu m$ deep. (b) Zoom-in view of the front section. (c) Zoom-in view of the back section.
The added slots introduce negligible losses to the laser cavity compared to the F-P ridge lasers due to the small distortion to the laser waveguide structure. To verify this, we measured the threshold current density of a device (1 mm×16 μm) with only the front section patterned with slots. The measured value of 6.2 kA/cm² is very close to that (~5.9 kA/cm²) of the unpatterned device with the same size. For the slot-QCL, the threshold current density of the front section is measured to be ~ 10 kA/cm² at 300 K when the back section is unbiased. The increased threshold current density is attributed to the strong free carrier absorption in the back section due to the high doping and some current spreading between the two sections. The waveguide loss measured by using the 1/L method results in ~23 cm⁻¹. To characterize the slot-QCLs, the front section of the device was driven above the threshold by a pulsed current $I_f = 1.2I_{th}$, where $I_{th}$ is the front section threshold current density when the back section is unbiased. The current pulse duration was 100 ns (short enough to avoid the thermal chirping effect) with a pulse repetition rate of 10 kHz. The back section was driven by using a DC current $I_b$ from 10 to 200 mA. A relatively wide tunable single-mode operation with high SMSRs was obtained by simultaneously varying the $I_b$ as well as the heat sink temperature $T$. Fig. 4-2 (a) shows the evolution of the single-mode peak positions by varying $I_b$ and $T$. The inset figure shows the current tuning behavior at a fixed temperature. For clarity, Fig. 4-2(b) shows selected single-mode emission peaks of the fabricated slot-QCL over the entire tuning range. For comparison, the spectrum from a traditional F-P ridge laser (2 mm × 16 μm) which is processed from the same QCL wafer is also shown (Fig. 4-2 (c)) which was measured at 300 K. The F-P laser shows a clear multi-mode operation just above the threshold current density at a center wavenumber of 968 cm⁻¹.
Fig. 4-2: (a) Single-mode peak evolution with temperature. The back section DC currents (in unit of mA) are also shown above their emission peaks. The inset shows the current tuning behavior of the device at T = 297 K. (b) Selected single-mode spectra of slot-QCLs. Measurements were taken at 100 ns and 10 kHz under pulse operation for the front section. The DC current into the back section varied from 10 mA to 200 mA and the substrate temperature changes from 80 K to 300 K. (c) Lasing spectrum of a traditional F-P laser (2 mm x 16 μm) biased at 1.2 times of the threshold. The measured temperature is 300 K.

Fig. 4-2 (b) shows that a total single-mode tuning range of 77 cm⁻¹ (corresponding to 785 nm), which is of 7.8 % relative tuning range, was realized for slot-QCLs. Continuous tuning range is about 24 cm⁻¹, which accounts for 31 % of the whole tuning range. A large continuous tuning range (mode-hop-free tuning range) is observed around 970 cm⁻¹, which is due to the flat gain profile and high gain at the centered wavelength as shown in Fig. 4-2 (c). Within the tuning range, most lasing peaks exhibit SMSRs of >=20 dB. Single-mode peak powers are in the range of ~30 to ~100 mW, which are sufficient for most of the spectroscopy applications. From Fig. 4-2 (a) and the inset figure, the temperature tuning coefficients of \( dn_{eff}/dT \) and \( dn_{eff}/dI_b \) are calculated to be ~
2.17 × 10^{-4} \text{ K}^{-1} \text{ and } \sim 4 \times 10^{-5} \text{ mA}^{-1}, \text{ respectively, where } n_{\text{eff}} \text{ is the group effective refractive index. The extracted values of } dn_{\text{eff}}/dT \text{ and } dn_{\text{eff}}/dI_b \text{ are in agreement with those reported using similar QCLs at similar wavelengths [103-104].}

### 4.3 Slot-QCLs theoretical analysis

To illustrate the Vernier-effect-based tuning mechanism of slot-QCLs, we use experimental results of tuning between two single-mode emission spectra at \sim 1010 \text{ cm}^{-1} \text{ and } \sim 1030 \text{ cm}^{-1} \text{ for analysis (Fig. 4-3(a)). The emission spectrum at } \sim 1010 \text{ cm}^{-1} \text{ was recorded at } I_f = 1.2 I_{th}, I_b = 50 \text{ mA, and } T = 158 \text{ K, while the spectrum at } \sim 1030 \text{ cm}^{-1} \text{ was characterized at } I_f = 1.2 I_{th}, I_b = 30 \text{ mA, and } T = 78 \text{ K. The lasing wavelength is primarily determined by the reflection spectra of the front and back sections. To calculate the field reflectivity from the front and back sections, we used the following approximate expressions taking into account the laser facet effect as given by}

\begin{align}
  r_F &= r_s \left[1 - t_s^{2N} \exp(gNL_f) \exp(-2 jkn_{\text{eff}} NL_f) \right] \\
  &+ r_j t_s^{2N} \exp\left[g(L_f(N-1) + L_{pf})\right] \exp\left[-j2((N-1)kn_{\text{eff}} L_f + kn_{\text{eff}} L_{pf})\right] \\
  \tag{4.1}
\end{align}

\begin{align}
  r_B &= r_s \left[1 - t_s^{2N} \exp(\alpha_b NL_b) \exp(-2 jkn_{\text{eff}} NL_b) \right] \\
  &+ r_j t_s^{2N} \exp\left[\alpha_b(L_b(N-1) + L_{pb})\right] \exp\left[-j2((N-1)kn_{\text{eff}} L_b + kn_{\text{eff}} L_{pb})\right] \\
  \tag{4.2}
\end{align}

where \( r_s \) and \( t_s \) are the field reflection and transmission coefficients of a single slot, \( r_F \) (\( r_B \)) is the total field reflectivity of the front (back) section, \( g \) is the net modal gain of front section, \( \alpha_b \) is the loss of the back section, \( N \) is the number of slots, \( k = 2\pi/\lambda \) is the wavenumber in vacuum, where \( \lambda \) is the free space wavelength, \( r_j \) is the laser facet field reflection coefficient, and \( L_{pf}(L_{pb}) \) is the spacing between the laser facet and the adjacent slot in the front (back) section.

In our model, we treat the middle and the back sections as one passive partial reflecting mirror of the whole laser cavity as both sections are biased well below the threshold (no net modal gain). The field reflectivity of this partial reflecting mirror can then be written
as $r_M = r_B \exp(-\alpha_m L_M) \exp(-2 j k n_{eff} L_M)$, where $\alpha_m \sim 23$ cm$^{-1}$ is the waveguide loss of the middle section and the back section reflection $r_B$ can be directly calculated using Eq. (4.2). However, calculation of the front section reflection $r_F$ is not so straightforward.

To calculate $r_F$, as the front section is biased above the threshold, we need to determine the net modal gain $g$ and the effective length of the front section $L_{eff\;F}$ (which is also the effective length of the whole laser cavity). The $g$ can be written as $g = \ln(1/|r_F||r_M|)/L_{eff\;F}$, where $L_{eff\;F} = \tanh(\kappa/L_F)/2\kappa$, and $\kappa = |r_F|/L_F$ is the reflection per unit length of the front section [142]. With the expressions of $g$ and $r_F$ as shown in Eq. (4.1), a self-consistent approach can then be used to calculate the net modal gain $g$ and $r_F$ of the front section.

We used finite-difference time-domain (FDTD) package Lumerical software to determine the $r$s and $t$s, by monitoring the optical power of the TM eigenmode at positions just before and after the slot in 3D simulations in which the laser ridge width effects are considered. In the 3D model, the corresponding dielectric constants of different materials in the waveguide structure were calculated using the semi-classical Drude model. The calculated $r$s and $t$s of the slot with a width of 5.5 µm and a depth of 2.2 µm are determined to be 0.04 and 0.98. The loss introduced by the slot is ~2.5 cm$^{-1}$.

The $r_F$ is set to be 0.54, which is equivalent to the facet power reflectivity of 0.3. The $L_{eff\;F}$ and $L_{eff\;B}$ are measured to be ~160 µm and ~96 µm for the front and back sections, respectively. Using Eqs. (4.1) and (4.2), the calculated $r_F$ and $r_B$ at different characterization conditions are shown in Fig. 4-3 (b) – (e), where $g$ ($\alpha_b$) has been included in the front (back) calculation. It is noted that the reflectivity of the front section in the calculation is greater than unity, which is due to the net modal gain effect in the front section. The measured temperature tuning coefficients $d_{neff}/dT = 2.17 \times 10^{-4}$ K$^{-1}$ and $dn_{eff}/dI_b = 4 \times 10^{-5}$ mA$^{-1}$ were utilized in the simulations. It shows that when the temperature changes from 78K to 158K, a clear redshift of the reflectivitycomb is observed, resulting in the reflectivity peak shifted from 1030 cm$^{-1}$ (pink solid line in Fig. 4-3 (e)) to 1010 cm$^{-1}$ (pink solid line in Fig. 4-3 (c)), matching well with Fig. 4-3(a).
Fig. 4-3: (a) Single-mode spectra at wavenumbers of ~1010 cm\(^{-1}\) (black line) and ~1030 cm\(^{-1}\) (red line).
(b) Reflection spectra from the front section (red dashed line) and the back section (blue solid line) at \(I_f = 1.2 \, I_{th}\), \(I_b = 50\) mA, and \(T = 158\) K. (c) Total reflectivity at \(I_f = 1.2 \, I_{th}\), \(I_b = 50\) mA, and \(T = 158\) K. (d) Reflection spectra from both the front (red dashed line) and back (blue solid line) sections at \(I_f = 1.2 \, I_{th}\), \(I_b = 30\) mA, and \(T = 78\) K. (e) Total reflectivity at \(I_f = 1.2 \, I_{th}\), \(I_b = 30\) mA, \(T = 78\) K, and \(r_f = 0.54\).
The facet effects on the power reflectivity of one section, e.g. the front section, were also investigated by varying $L_{pf}$ and $r_f$. Using Eq. (4.1), the corresponding calculated results are shown in Fig. 4-4 (a) and (b), where $g = 0$, $r_s = 0.04$, $t_s = 0.97$, $N = 11$, and $L_f = 80 \ \mu\text{m}$. The reflectivity shows strong dependence on $L_{pf}$ and $r_f$, with reduced SMSRs. The physics behind is the interference between the light reflected from the laser facet and the slot array. The constructive interference at the designed wavelengths will only occur when $L_{pf} = nL_f$, where $n$ is an integer, as shown in Fig. 4-4 (a) with $L_{pf} = 80 \ \mu\text{m}$. Reducing the facet reflectivity will significantly reduce the effect of laser facet (Fig. 4-4 (b)) and thus lead to an enhanced SMSR and the mode selectivity. Thus, depositing an antireflection (AR) coating on the laser facet would be an effective approach to reduce the laser facet effect.

To examine the generation efficiency of Joule heating by the injected current, we estimated the expected temperature variation in the back section. The temperature change in the back section can be written as $\Delta T = V_bI_b/W_{act}L_bG_{th}$, where $V_b$ is the DC voltage applied to back section, $W_{act}$ is the width of the active region, and $G_{th}$ is the active region thermal conductance. For values $V_b = 1.3 \ \text{V}$, $I_b = 40 \ \text{mA}$, $W_{act} = 16 \ \mu\text{m}$, $L_B = 1.09 \ \text{mm}$, and $G_{th} = 45 \ \text{W/ K cm}$ [104], the $\Delta T$ is calculated to be 6.56 K, in agreement with the experimental result of 5.97 K. The difference can be attributed to the current leakage between the two sections.

### 4.4 Discussion and conclusions

Compared with other types of coupled-cavity structures [104], besides the relatively larger spanning range, our device also possesses a broader continuous tuning range. Although there are still several continuous tuning gaps in the spectra (Fig. 4-2 (b)), this drawback can be overcome if we can vary the local refractive indices of the two sections simultaneously [105], for instance, by adding another DC current $I_{f, dc}$ to the front section on top of the pulsed current applied. For comparison, we calculated the normalized total reflectivity of slot-QCLs with $L_f = 80 \ \mu\text{m}$, and $L_h = 96 \ \mu\text{m}$ under one-DC-current and two-DC-current pumping schemes as shown in Fig. 4-5 (a) and (b), respectively.
Fig. 4.4: (a) Total power reflectivity for $L_{pf} = 60 \mu$m, 80 $\mu$m, and 90 $\mu$m, respectively, with the $g = 0$, $r_s = 0.04$, $t_s = 0.97$, $N = 11$, and $L_f = 80 \mu$m. (b) Total power reflectivity for $r_f = 0.1$, 0.32 and 0.54, respectively, with the $g = 0$, $r_s = 0.04$, $t_s = 0.97$, $N = 11$, $L_f = 80 \mu$m, and $L_{pf} = 90 \mu$m.

In Fig. 4-5 (a), the group effective refractive index $n_{eff,f}$ is fixed at 3.415 (the central wavenumber is chosen as 970 cm$^{-1}$) while that of the back section $n_{eff,b}$ changes from 3.427 to 3.439, only one reflectivity maximum is observed indicating discontinuity of the tuning spectra. In Fig. 4-5 (b), by changing $n_{eff,f}$ from 3.409 to 3.421 and scanning $n_{eff,b}$ from 3.427 to 3.439, maximum reflectivity can be obtained continuously within the entire scanning range, enabling much wider continuous tuning spectra. In addition, as seen from Fig. 4-2 (b), a larger continuous tuning range can also be achieved through active region design with flat gain profiles. Furthermore, by optimizing the slot configurations in both sections, i.e. slot number and slot spacing, it is possible to achieve relatively broad continuous tuning slot-QCLs at RT.
In conclusion, we have experimentally demonstrated tunable single-mode two-section slot-QCLs which have shown a relatively broad tuning range of 77 cm\(^{-1}\) (785 nm), corresponding to 7.8 % relative tuning range, with a 31 % continuous tuning range and an average SMSR of ~20 dB within the whole tuning range. The total tuning range is close to that of ~90 cm\(^{-1}\) with a SMSR of ~20 dB by using EC-QCLs with the same active region as our devices. The relatively broad single-mode tuning range combined with the easy device fabrication realized by only using conventional contact photolithography makes slot-QCLs appealing miniaturized mid-infrared sources for sensing and spectroscopy applications. Through numerical simulations, we showed that a much larger continuous tuning range can be achieved by separately pumping the front and the back sections through DC biases. Meanwhile, to achieve a broad tuning range at RT, the slot configurations can be optimized to reduce the spacing of the reflection peak while still maintaining high spectrum SMSRs. Due to the relatively high doping level, it is difficult to achieve CW operation in our current devices. However, it is possible to achieve CW operated slot-QCLs by using high performance QCL wafers, e.g. at ~4.6 μm. Further investigations are underway to realize RT broadly tunable single-mode CW slot-QCLs.
Fig. 4-5: (a) Calculated normalized total reflectivity of the slot-QCLs when varying $I_b$ only. The group effective refractive index of the front section $n_{eff,f}$ is fixed at 3.415, and that of the back section $n_{eff,b}$ is scanning from 3.427 to 3.439. (b) Normalized total reflectivity of the slot-QCLs when varying both $I_{dc}$ and $I_b$. The group effective refractive index of the front section $n_{eff,f}$ is in the range of 3.409 to 3.421 and that of the back section $n_{eff,b}$ is from 3.427 to 3.439.
Chapter 5
Tunable single-mode quantum cascade lasers via surface-acoustic-wave modulation

5.1 Introduction of surface acoustic wave modulated QCLs

Modern spectroscopies require laser sources with not only broad tuning range but also high speed tuning speed for noise reduction and fast dynamic activity monitoring. As discussed previously, the DFB-QCLs can be tuned by changing the effective refractive index of the active region through varying the operating temperature or the injection current. However, both approaches reply on the thermal-induced refractive index change which is a slow process in the range of milliseconds. On the other hand, though EC-QCLs have achieved a very broad tuning range of 432 cm\(^{-1}\) [24], the additional mechanical parts (e.g. the mirror) greatly limit their tuning speed due to the nature of mechanical tuning. Hence, new technology is need to provide broad tuning range with fast tuning speed.

A possible solution is the acoustic wave technology, which can be classified into bulk acoustic wave (BAW)-based and surface acoustic wave (SAW)-based, provides an attracting approach to tune the mechanical, electrical and optical properties of optoelectronic devices [143-144], having potential advantages of fast tuning speed, broad tuning range, and flexibility in the design. Nowadays, acoustic wave technology has attracted growing interest in controlling the electrical and optical properties of materials, such as semiconductor quantum wells [145], micro photonic structures [146], and even liquids [147]. Acoustic approach was proposed to achieve single-mode tunable emission from QCLs shortly after their first demonstration, utilizing the piezo-electric
property of the laser material [148]. Due to the unipolar nature of QCLs, the acoustic modulation makes practical sense for these novel semiconductor lasers, where an acoustic wave propagating along the optical axis generates periodic modulation of the carrier density. Because the optical gain is linear in carrier density, this periodic modulation of carrier density does not affect the average gain but does provide a distributed feedback for the optical wave. This is to be contrasted with the situation in bipolar lasers, where the longitudinal piezoelectric field would spatially separate electrons and holes, thus degrading the average material gain. This approach was carefully examined in [140] where sheer (bulk) acoustic wave was proposed to achieve single-mode operation of QCLs with good side-mode suppression ratio through gain modulation. The physical mechanism behind is the modulation of the electron density induced by the BAW, forming a distributed feedback grating on the active region of QCLs, which can be used for achieving single-mode laser action [140]. Compared with BAW, SAW has a better performance in terms of input electrical power, tunability, single-mode operation, and long-term stability as indicated in [140], but requires carefully designed waveguide structures as well as a comprehensive theoretical model for the design of the device. In the past, SAW-modulated DFB diode lasers were studied theoretically by a few groups [149-150], after the first proposal by Kogelnik and Shank [151]. The first experimental demonstration of SAW-modulated DFB laser was realized by Yamanishi, et. al. [152-153], in which acoustic mode conversation from BAW to SAW was achieved to compensate the large propagation loss of high frequency SAW. In this chapter, we report the theoretical investigation of DFB-QCLs modulated by traveling SAW. We first study the underlying physics of the travelling SAW-modulated DFB-QCLs, then derive the analytical expression of the coupling coefficient of the DFB gratings formed by the SAW on QCLs. In order to increase the coupling coefficient, air-waveguide and surface plasmon waveguide structures are proposed which provide high modulation because the active regions of such structures are close to the top surface of the device. In addition, to increase the coupling strength induced by the SAW, a highly piezoelectric Zinc Oxide (ZnO) thin film is proposed, applied beneath the SAW transducers. A similar design has been proposed in [140], where the transducers were
patterned on sidewalls of the devices. However, their approach is more technically difficult than our designs.

Through numerical simulations, we found that by designing the waveguide and active region thicknesses to match the SAW intensity distribution in the waveguide growth direction, moderate coupling coefficient of $\approx 2.5 \text{ cm}^{-1}$ can be achieved for both air-waveguide and surface-plasmon waveguide structures. Our previous experiments already demonstrated the possibility of achieving an enhanced SAW modulation on III-V semiconductors assisted by a ZnO thin film at high SAW frequencies (a few GHz) [154]. Such high frequency SAWs can be used for light modulation at wavelength of a few micrometers, which is in the mid-infrared range [154]. The SAW modulated DFB-QCLs may provide an alternative solution to the tunable mid-infrared light sources for spectroscopy, free-space communication, and integrated optoelectronics applications. We note that J. D. Cooper et. al. [155] has recently proposed the use of BAWs converted from SAWs for modulation of QCLs, which is different from the schemes proposed here.

5.2 Theoretical analysis of SAW modulated DFB QCLs

Normally, SAWs can be effectively generated by patterning interdigital transducers (IDTs) directly on the material surface. Fig. 5-1 shows the schematic structure of the proposed SAW-modulated DFB-QCLs, where the active region is sandwiched by the lower and upper passive regions (cladding layers) and the SAW is generated by IDTs and then propagates along the laser ridge. Also shown is the ZnO thin film deposited on top of the ridge to enhance the piezoelectric properties of the waveguide structure. Using standard submicron fabrication process, e.g. e-beam lithography [154], we can fabricate the IDTs on the top surface of QCLs. The physical picture of traveling SAW- modulated QCLs is as follows: when a sinusoidal SAW is travelling along the laser ridge, a sinusoidal modulation of the carrier density $\hat{\rho}$ is generated due to the periodic electric potential modulation $\hat{\phi}$ caused by the propagating SAWs. Because the optical gain/loss $g/\alpha$ of QCL is directly proportional to the carrier density $\rho$, a periodic optical gain/loss modulation $\hat{g}/\hat{\alpha}$ is thus achieved within the cavity. This gain/loss modulation in turn
leads a dielectric constant modulation $\Delta \varepsilon$. Due to the fact that the refractive index $n_{\text{eff}} = \sqrt{\varepsilon}$, a sinusoidal refractive modulation is expected to be induced by the travelling SAW, thus forming a DFB grating with period determined by the wavelength of SAW.

Fig. 5-1: Schematic diagram of the surface acoustic wave (SAW) modulated quantum cascade laser (QCL). The travelling SAW is generated by applying an RF signal on the integrated interdigital transducer (IDT) and propagates along the x’[110] crystallographic direction. The inset shows the general waveguide structure and the conduction band (CB) electron density modulation of QCLs caused by the travelling SAW. A Zinc Oxide (ZnO) thin film is deposited on the top of the waveguide structure to increase the coupling efficiency.

This electric potential originates from the periodic strain/stress induced by the SAWs. Because the optical gain of QCL is directly proportional to the carrier density, a periodic optical gain modulation is thus achieved simultaneously. This leads to a distributed feedback effect on the optical mode in the laser cavity. Thus, the proposed SAW-modulated DFB-QCL is a type of gain/loss-coupled DFB laser. Apart from having the properties of conventional DFB QCLs, SAW DFB-QCL also shows unique
characteristics in terms of the tunability. Different from the conventional DFB-QCLs, for which the wavelength is basically fixed by the grating embedded in the waveguide structure (unless it is tuned through temperature or injected current), the Bragg wavelength of travelling SAW-modulated DFB-QCLs can be tailored simply by changing the propagating wavelength (the RF modulation frequency) of the SAWs. As shown in [154], a broad range of SAW can be generated even with a fixed IDT using different RF frequency signals, showing the possibility of achieving broadly tunable single mode SAW-modulated DFB-QCLs.

5.2.1 QCL modulated by traveling SAW DFB gratings

To analyze the strength of the interaction between the optical lasing mode and the travelling SAW within the QCL laser cavity, we start the analysis using coupled-mode theory. The exponential forms of the forward-propagating optical wave, the backward-propagating optical wave, and the forward propagating acoustic wave can be expressed as,

\[ A_f(x',t) = A_f(x') \exp[i(\beta_f x' - \omega_f t)] \exp[-gx'/2] \]  
\[ A_b(x',t) = A_b(x') \exp[-i(\beta_b x' + \omega_b t)] \exp[-gx'/2] \]  
\[ A_{ac}(x',t) = \cos(kx' - \Omega t) \exp[-\alpha_{ac}x'/2] \]

where \( A_f(b), \beta_f(b), \omega_f(b), \) and \( g, \) correspond to the amplitude, propagation constant, angular frequency of the forward- (backward-) propagating optical field, and the optical gain coefficient, respectively; \( \alpha_{ac}, k, \) and \( \Omega \) represent the loss coefficient, propagation constant and angular frequency of the acoustic wave, respectively. The corresponding modulated dielectric constant, directly related to the coupling coefficient, is

\[ \Delta \varepsilon(x', y, z, t) = \Delta \varepsilon(y, z) \cos(kx' - \Omega t) \exp[-\alpha_{ac}x'/2] \]  

According to coupled-mode theory [109], the amplitudes of the forward-optical wave \( A_f \) and the backward-optical wave \( A_b \) satisfy the following coupled-mode equations:

\[ \frac{dA_f(x')}{dz} = iK_{fb} \exp(-\alpha_{ac}x'/2) A_b \exp[-i2\Delta x'] \times \exp[i(\omega_f - \omega_b - \Omega) t] \]  

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\[
\frac{dA_b(x')}{dz} = -iK_{bf} \exp(-\alpha_{ac} x'/2) A_f \exp[i2\Delta x'] \times \exp[-i(\omega_f - \omega_b - \Omega)t] \tag{5.6}
\]

where \(K_{pf}\) and \(K_{bf}\) are the coupling coefficients, and the phase mismatch is expressed as:

\[
2\Delta = \beta_f + \beta_b - K - ig .
\]

Due to the Doppler effect, we have \(\omega_f - \omega_b = \Omega\). Then Eq. (3a) and Eq. (3b) can be simplified as:

\[
\frac{dA_f(x')}{dz} = iK_0 \exp(-\alpha_{ac} x'/2) A_b \exp(-i2\Delta x') \tag{5.7}
\]

\[
\frac{dA_b(x')}{dz} = -iK_0 \exp(-\alpha_{ac} x'/2) A_f \exp(i2\Delta x') \tag{5.8}
\]

where we have defined, \(K_{pf} \approx K_{bf} \approx K_0\). The exact expression of \(K_0\) is shown in the following section. Due to the acoustic wave propagation loss, the SAW intensity attenuates as it propagates along the laser ridge; thus, the coupling coefficient decreases as well (see Fig. 5-2). Also shown in this figure is the effective DFB cavity length \(L_e\), defined as the distance where the coupling coefficient decreases from \(K_0\) to \(K_0/e\), where \(K_0\) is the coupling coefficient at the IDT location. We define the region within \(L_e\) as the effective DFB region. Because of the low piezoelectric properties of the QCLs materials (commonly InP or GaAs-based materials), the SAW will attenuate rapidly on the surface of QCLs due to the relatively large SAW loss. However, by adding highly piezoelectric materials on top of QCLs (see Fig. 5-1) one can increase the piezoelectric effect, thus the SAW modulation efficiency. Theoretical analysis [156] at \(~1.6\) GHz corresponding to SAW wavelength \(~1.6\) \(\mu\)m shows that the loss of SAW on the ZnO material is \(~3\) dB/mm, leading to an effective DFB cavity length \(L_e = 2/\alpha_{ac}\) of \(~2.9\) mm at \(1.6\) GHz. Normally the state-of-the-art ridge-waveguide QCLs have a cavity length \(L\) of 4 – 5 mm [76, 132]. This leads to a strong attenuation of \(~97\%) of SAW amplitude at the laser facet. The strong attenuation is actually advantageous due to the following two reasons:

First, acoustic wave reflection from the laser facet (located at \(X' = X'_0\) in Fig. 5-2) is minimized. Any moderate reflection of SAW from the laser facet will reduce the electron density modulation; therefore suppress the DFB grating formation. Second, if significant SAW reflection occurs at the laser facet, standing SAW will be formed within the laser cavity; thus, only the acoustic wave wavelength corresponding to the cavity

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resonant frequency will be supported. In this case, achieving continuous tuning of SAW wavelength (thus the tuning of light emission) is impossible.

Fig. 5-2: Schematic of the coupling coefficient distribution along the propagation direction of SAW. \( L_e \) is the effective DFB cavity length of the SAW-modulated QCLs defined as the distance where the coupling coefficient decreases from \( K_0 \) to \( K_0/e \).

### 5.2.2 Coupling coefficient induced by SAW DFB grating

To calculate the coupling coefficient induced by the SAW gratings, we look at simplified structure where SAW is propagating along an air/semiconductor interface, as shown in Fig. 5-2. To achieve the highest SAW modulation efficiency possible, the acoustic wave is assumed to propagate in the [110] crystallographic direction as it can produce the largest transverse displacement in the (001) plane of the QCLs wafers [157]. We use \( u_z \) and \( u'_z \) to represent the transverse displacement and longitudinal displacement in the [001] and [110] crystallographic directions, respectively.

The fundamental relation for the displacement and the electric field in a piezoelectric material is expressed as \([D] = \varepsilon[\varepsilon][E] + [\varepsilon][S][158]\), where \( D \), \( \varepsilon \), \( E \), and \( S \) represent the electric displacement, material permittivity, electric field, piezoelectric coefficient, and strain of the material, respectively. In our case, the expanded expression of the above formula is shown as,
\[
\begin{bmatrix}
D_x \\
D_y \\
D_z
\end{bmatrix} =
\begin{bmatrix}
\varepsilon_{11} & 0 & 0 \\
0 & \varepsilon_{11} & 0 \\
0 & 0 & \varepsilon_{11}
\end{bmatrix}
\begin{bmatrix}
E_x \\
E_y \\
E_z
\end{bmatrix} +
\begin{bmatrix}
0 & 0 & \varepsilon_{14} \\
0 & 0 & 0 \\
0 & 0 & 0
\end{bmatrix}
\begin{bmatrix}
S_{11} \\
S_{22} \\
S_{33}
\end{bmatrix}
\]

where \( S_{11} = \partial u_x / \partial x \), \( S_{22} = \partial u_y / \partial y \), \( S_{33} = \partial u_z / \partial z \), \( S_{13} = (\partial u_x / \partial x + \partial u_z / \partial z) / 2 \), \( S_{12} = (\partial u_y / \partial x + \partial u_z / \partial y) / 2 \), and \( S_{23} = (\partial u_y / \partial y + \partial u_z / \partial z) / 2 \), in which \( u_x \), \( u_y \), and \( u_z \) represent the displacements along the [100], [010], and [001] crystallographic directions, respectively. The corresponding crystallographic directions are shown in Fig. 5-1; \( D_{x, y, z} \) and \( E_{x, y, z} \) are the electric displacements and the electric fields along the [100], [010], and [001] crystallographic directions, respectively; \( \varepsilon_{11} \) and \( \varepsilon_{14} \) are the corresponding dielectric constant and the piezoelectric constant for the material used. It is noted that, in the coordinate system we use here, the SAW is propagating long the [110] crystallographic direction, so the displacement in the propagation direction is \( u'_x = \sqrt{2} u_x \). We will use \( u'_x \) hereafter in the calculations. According to the basic electromagnetic relations \( \text{div} D = \rho \), and \( E = -\text{grad} \phi \), where \( \phi \) stands for the electric potential, we have

\[
\varepsilon_{11} \left( q_{ac}^2 - \frac{d^2}{dz^2} \right) \frac{d\phi}{dz} + \frac{\varepsilon_{14}}{2} \left( \pm 2i q_{ac} \frac{du'_x}{dz} - q_{ac}^2 u'_x \right) = \tilde{\rho}
\]

where \( q_{ac} \) is the acoustic wave propagation constant. Because in the active regions of QCLs, the electron transport is governed by resonant tunneling; thus, the conductivity of the active region is different from that of the cladding layer. In the following, we will use passive region to stand for the cladding layer. Using small signal analysis, the AC components of the current density in the \( x' \) and \( z \) directions can be expressed as,

\[
\tilde{j}_{x'} = \sigma \tilde{E}_{x'} - i D q_{ac} \tilde{\rho}
\]

\[
\text{Passive: } \tilde{j}_z = \sigma \tilde{E}_z + \nu_{\varepsilon} \tilde{\rho} - D_{z} \frac{d\tilde{\rho}}{dz}
\]

\[
\text{Active: } \tilde{j}_z = \sigma \varepsilon \tilde{E}_z + \nu_{\varepsilon} \tilde{\rho} - D_{z} \frac{d\tilde{\rho}}{dz}
\]
where $D_z$, $\sigma_z$ represent diffusion coefficients and conductivities for the passive and active regions, respectively, and $v_{tc}$ is the transverse DC electron velocity for both regions. In QCLs, the current density induced by resonant tunneling between the active region and the injector region is [159],

$$ j_{st} = eN_s \frac{2\Omega(E)^2 \tau_{deph}(E)}{1 + (\Delta E_{1z}/\hbar)^2 \tau_{deph}^2(E) + 4\Omega(E)^2 \tau_s(E)\tau_{deph}(E)} $$  \hspace{1cm} (5.14)

where $N_s$, $\tau_0(E)$, and $\Omega(E)$, correspond to sheet doping density, upper laser state lifetime, and coupling strength between the injector ground state and upper laser state of the following period, respectively; $\tau_{deph}(E)$, and $\Delta E_{1z}(E)$ are the dephasing time and energy detuning from resonance; $E$ represents the electric field provided by the power supply to the device. Based on the expression of the resonant tunneling current density, the effective conductivity of the active region can be derived as,

$$ \sigma_z = \frac{dj_{st}}{dE} = eN_s \left[ \frac{df(E)}{dE} \right] $$  \hspace{1cm} (5.15)

where $f(E) = 2\Omega(E)^2 \tau_{deph}(E)/(1 + (\Delta E_{1z}/\hbar)^2 \tau_{deph}^2(E) + 4\Omega(E)^2 \tau_s(E)\tau_{deph}(E))$. Due to the fact that the current in the active region is governed by resonant tunneling within the active region, it is safe to assume that the transverse diffusion coefficient of the active region $D_z \approx 0$. Substituting Eqs. (5.11), (5.12), and (5.13) into the current continuity equations, $\nabla \cdot j + \partial \rho / \partial t = 0$, we have

$$ \text{Passive: } \varepsilon_{11} \left( q_{ac}^2 - \frac{d}{dz^2} \right) \phi = \left[ \pm i \omega_{ac} \tau_M - \left( q_{ac}^2 - \frac{d}{dz^2} \right) \lambda_D^2 - \tau_M v_{tc} \frac{d}{dz} \right] \rho $$  \hspace{1cm} (5.16)

$$ \text{Active: } \varepsilon_{11} \left( q_{ac}^2 - \frac{\sigma_z}{\sigma} \frac{d}{dz^2} \right) \phi = \left[ \pm i \omega_{ac} \tau_M - q_{ac}^2 \lambda_D^2 - \tau_M v_{tc} \frac{d}{dz} \right] \rho $$  \hspace{1cm} (5.17)

where $\tau_M = \varepsilon_{11}/\sigma$ is the dielectric relaxation time [160], and $\lambda_D = (D \tau_M)^{1/2} = (\varepsilon_{11} k_B T / e^2 N_0)^{1/2}$ is the Debye screening length [161], where $k_B$, $T$, $e$, $N_0$ are the Boltzmann constant, temperature, electron charge, and electron density, respectively. Combining Eqs. (5.10), (5.16) and (5.17), we have the equations for the carrier density variations in the passive and active region as Eqs. (5.18) and (5.19).
\[ \text{Passive: } 1 + i \omega_0 \tau_M + \left( q_{ac}^2 - \frac{d^2}{dz^2} \right) \rho = \frac{e_{1u}}{2} \left( \pm 2i q_{ac} \frac{du_x}{dz} - q_{ac}^2 u_z \right) \] (5.18)

\[ \text{Active: } 1 - \left( \pm i \omega_0 \tau_M - q_{ac}^2 \frac{d^2}{dz^2} + \tau_M \frac{d}{dz} \right) \left( 1 - \frac{1}{q_{ac}^2} \frac{d}{dz} \right) \rho = \frac{e_{1u}}{2} \left( \pm 2i q_{ac} \frac{du_x}{dz} - q_{ac}^2 u_z \right) \] (5.19)

It is shown that \( \sigma_z \ll \sigma \), given that \( \sigma \approx 10 \text{ mho/m} \), according to Eq. (5.15), with current density \( j \approx 1 \text{kA/cm}^2 \) (common for normal QCLs designs), and \( \sigma \approx 10^3 \text{ mho/m} \) for \( N_0 \approx 10^{17} \text{cm}^3 \). For typical QCLs, \( N_0 \approx 10^{17} \text{cm}^3 \) and \( \mu \approx 10^3 \text{cm}^2/V \text{s} \), \( \tau_M \approx 7 \times 10^{-14} \text{s} \); from \( \omega_{ac} \approx 2\pi \times 1.6 \times 10^9 \text{s} \), \( \lambda_D \approx 1.3 \times 10^{-6} \text{cm} \) (for 300 K), and \( \nu_{ec} = j_0/eN_0 \approx 6 \times 10^4 \text{cm/s} \), it shows that \( \omega_{ac} \tau_M \approx 1, \quad (q_{ac} \lambda_D)^2 \approx 3 \times 10^{-3} \ll 1, \) and \( \tau_M \nu_{ec} q_{ac} \approx 1.7 \times 10^{-4} \ll 1 \). Thus, we have the simplified expression for the carrier density variation,

\[ \tilde{\rho} \approx \frac{e_{1u}}{2} \left( \pm 2i q_{ac} \frac{du_x'}{dz} - q_{ac}^2 u_z \right) \] (5.20)

which applies to both the passive and active regions in QCLs. Using the exponential forms of \( u_x' \) and \( u_z \), \( \tilde{\rho} \) can be expressed as,

\[ \tilde{\rho} \approx \left( \frac{e_{1u}}{2} \right) \left( \pm 2i q_{ac} \exp[\pm i(\theta_x - \theta_z)] \frac{du_x'}{dz} - q_{ac}^2 u_x \right) \exp[\pm i(q_{ac}x - \omega_{ac}t + \theta_z)] \] (5.21)

The loss and gain modulations in the passive and active regions are represented by,

\[ \text{Passive: } \tilde{\alpha} = -\left( \frac{\tilde{\rho}}{eN_0} \right) \alpha_0 \] (5.22)

\[ \text{Active: } \tilde{g} = -\left( \frac{\tilde{\rho}}{eN_0} \right) g_0 \] (5.23)

where \( \alpha_0 \) (\( g_0 \)) is the local absorption (gain) coefficient. The corresponding variations of specific dielectric constant \( \Delta \epsilon \) are [162],

\[ \text{Passive: } \Delta \epsilon = i \left( \frac{c_0 n_{\text{neff}}}{\omega_{opt}} \right) \tilde{\alpha} \] (5.24)

\[ \text{Active: } \Delta \epsilon = -i \left( \frac{c_0 n_{\text{neff}}}{\omega_{opt}} \right) \tilde{g} \] (5.25)
where $c_0$ is the vacuum speed of light, $n_{neff}$ is the effective refractive index of the optical guided mode, and $\omega_{opt}$ is the angular frequency of the guided mode. According to coupled mode theory, the modulation of the dielectric constant leads to the following expression for the coupling coefficient $K_0$ [163],

$$K_0 = \left( \frac{k_0}{2n_{neff}} \right) \int \int \Delta \varepsilon(y,z) \left| E_{opt}(y,z) \right|^2 dydz \frac{\int \int \left| E_{opt}(y,z) \right|^2 dydz}{\int \int \left| E_{opt}(y,z) \right|^2 dydz}$$

(5.26)

where $k_0=2\pi/\lambda$, is the wavenumber of the guided lasing mode. Taking Eqs. (5.21) to (5.25) into (5.26), using the $\exp[i(q_{ac}x-\omega_{ac}t)]$ term in Eq. (5.21), the coupling coefficient is represented as Eq.(5.27). In the expression for coupling coefficient of the DFB grating, we want to highlight three points. Firstly, the gain modulation in the expression is only for the $E_z$ component of the optical mode, leaving $E_y$ unmodulated by the SAW. Thus, the distribution of $E_z$ will strongly influence the coupling coefficient of the SAW grating. Secondly, $u'_{ac}(z)$, $u_{ac}(z)$, $\theta_{ac}-\theta_0$ and $E_{opt}(y,z)$ will be calculated numerically by computer simulations, such as using COMSOL in our case. Thirdly, since the absorption coefficient in the passive region $\alpha_0$ is proportional to the carrier density $N_0$, as shown in [164] $\alpha_0 = N_0 c^2 \tau / \varepsilon_0 \varepsilon_{opt} m^* c$, where $\tau$, $\varepsilon_0$, $c$, and $m^*$ correspond to scattering time in semiconductor materials, vacuum permittivity, vacuum speed of light, and effective mass of the electron in the material, respectively; thus, $\alpha_0/N_0$ is virtually independent of $N_0$ in the first term of the expression, meaning that the doping in the passive region has no effect on the coupling coefficient between the optical wave and the SAW-generated DFB grating. Thus, we can reduce the doping level in the passive region to reduce the free carrier absorption.

$$K_0 = \left( \frac{i}{4} \right) \int \int_{passive} \frac{\varepsilon_{14}}{\varepsilon_{N_0}} \alpha_0 \left\{ q_{ac}^2 u_z(z) - 2i q_{ac} \exp[i(\theta_x - \theta_0)] \frac{du'_{ac}(z)}{dz} \right\} \left| E_{opt}(y,z) \right|^2 dydz$$

$$\int \int_{all} \left| E_{opt}(y,z) \right|^2 dydz$$

$$- \left( \frac{i}{4} \right) \int \int_{active} \frac{\varepsilon_{14}}{\varepsilon_{N_0}} g_0 \left\{ q_{ac}^2 u_z(z) - 2i q_{ac} \exp[i(\theta_x - \theta_0)] \frac{du'_{ac}(z)}{dz} \right\} \left| E_{opt}(y,z) \right|^2 dydz$$

$$\int \int_{all} \left| E_{opt}(y,z) \right|^2 dydz$$

(5.27)
5.2.3 New waveguide concepts calculation of coupling coefficients

Depositing highly piezoelectric material on top of QCLs can help increase the SAW modulation efficiency. Simultaneously, one can also design suitable waveguide structures to increase the coupling coefficient. In this section, we propose two possible waveguide structure designs to achieve high coupling coefficients between the optical field and the SAW-generated DFB grating, namely air-waveguide and surface plasmon waveguide structures.

If we simply use the traditional air-waveguide structure, piezo-modulation cancellation effect will happen (see Fig. 5-3 (a)). The physical mechanism is as follows: due to the oscillation effect of SAW along the depth of the waveguide, there is a $\pi$ phase shift of the SAW between part I and II (Fig. 5-3(a)). It is also the same case between part II and III. This means that the electron density is increased by the SAW modulation in part I but decreases in part II. Thus, according to Eqs. (5.23) and (5.25), the sign of the local dielectric modulations in part I and part II is different. This leads to an overall dielectric constant modulation $|\Delta \varepsilon| = |\Delta \varepsilon_I| + |\Delta \varepsilon_{II}| + |\Delta \varepsilon_{III}|$, where $|\Delta \varepsilon_I|$, $|\Delta \varepsilon_{II}|$, and $|\Delta \varepsilon_{III}|$ represent the norm of the dielectric constant modulations in part I, II and III, respectively. A clear cancellation effect of SAW modulation is shown. However, this cancellation effect can be removed if we use waveguide structures with two-section active regions separated by a lightly doped InGaAs layer as a passive layer (see Fig. 5-3 (b)). In this configuration, according to Eqs. (5.24) and (5.25), the local dielectric constant modulations in different parts of the waveguide structure will have the same sign. Thus, the total dielectric constant modulation becomes $|\Delta \varepsilon| = |\Delta \varepsilon_I| + |\Delta \varepsilon_{II}| + |\Delta \varepsilon_{III}|$, larger than that in the traditional air-waveguide structure. From Eq. (5.27), an increase of $\Delta \varepsilon$ will enhance the coupling coefficient $K_0$. To reduce the free carrier absorption in the passive waveguide structure in Fig. 5-3 (b), the passive InGaAs layer is lightly doped.
Fig. 5-3: Comparison of dielectric constant modulation between the traditional air-waveguide structure and the air-waveguide structure with two-section active regions. (a) The dielectric constant modulation in the traditional air-waveguide structure caused by the $u_z$ component of the SAW. The resulted dielectric constant modulation is $\Delta \varepsilon = |\Delta \varepsilon_1| - |\Delta \varepsilon_2| + |\Delta \varepsilon_3|$. (b) The dielectric constant modulation in the air-waveguide structure with two-section active regions. The resulted dielectric constant modulation is $|\Delta \varepsilon| = |\Delta \varepsilon_1| + |\Delta \varepsilon_2| + |\Delta \varepsilon_3|$.

### 5.2.3.1 Air-waveguide structure

Fig. 5-4 shows the three dimensional (3D) schematic diagram of the proposed air-waveguide structure. Two dimensional numerical simulations of the waveguide transverse optical mode and the SAW were performed by using a commercial finite-elements solver, Comsol Multiphysics. The cross-sectional view of the fundamental optical mode intensity distribution and the $u_z$ of the SAW are shown in Fig. 5-5 (a) and Fig. 5-5 (b). Part of the generated SAW is diffracted into substrate as shown in Fig. 5-5 (b). The electric field distribution of the fundamental transverse optical mode is shown in Fig. 5-6 (a). Fig. 5-6 (b) shows the amplitudes of the $u_z$ and $u_x'$ of SAW as a function of the depth of the laser structure, with corresponding parameters $v_{\text{phase}} = 2560 \text{ m/s}$, $\lambda_{ac} \sim 1.6 \mu m$, $f = 1.6 \text{ GHz}$, $N_{\text{finger}} = 100$, and $V = 4\text{V}$, which are close to that in [154]. The voltage is chosen to generate large enough SAW amplitude. Though thick ZnO film will
induce larger SAW displacement in the film, it will reduce SAW displacement inside the device. The thickness of the ZnO thin film is optimized to be 200 nm generating the largest modulation in the waveguide structure. The thicknesses of the active region I, the InGaAs layer, and the active region II is ~0.8 μm, ~1.3 μm, and ~1 μm, respectively.

Fig. 5-4: 3D schematic diagram of the air-waveguide structure.
Fig. 5.5: Optical and acoustic wave simulations of the air-waveguide structure. (a) Cross-sectional view of the intensity distribution of fundamental transverse optical mode inside the air-waveguide structure. (b) Amplitude distribution of the SAW $u_c$ component in the air-waveguide structure. Inset shows the waveguide structures. Zoom-in view of the SAW is also shown.

Fig. 5.6: Optical wave and acoustic wave distributions as a function of the depth of the QCL in the air-waveguide structure. (a) Normalized electric field of the fundamental transverse optical mode of the air-waveguide structure. (b) Amplitude distributions of $u_c$ and $u'$, along the depth of the waveguide structure.
For the air-waveguide structure, the resulted optical confinement factors for the two active regions are ~ 8% and ~ 40%, respectively. Given $NL = 2\mu m$, $g_0 \sim 50 \text{cm}^{-1}$, $\alpha_0 \sim 2.1 \text{cm}^{-1}$, $\varepsilon_{14} = 0.11 \text{C/m}^2$, $\lambda_{ac} = 1.6 \text{μm}$, $\theta_x - \theta_z = -\pi/2$, $N_0 = 1.57 \times 10^{16} \text{cm}^{-3}$, $W = 30 \mu m$, where $N$, $L$, $W$, $\lambda_{ac}$, $\theta_x - \theta_z$, are the number of period, period length, width of the active region, SAW wavelength, phase different between $u_z$ and $u'_x$, respectively, a coupling coefficient $K_0 \sim 2.5 \text{cm}^{-1}$ is calculated, according to Eq. (5.27). This results in $K_0L_e \sim 0.75$, close the critical value of $K_0L_e = 1$ for DFB lasers [165].

It is noted that even though the SAW exhibits damped oscillation from the ZnO thin film (e.g. within region I), this non-uniform SAW distribution doesn’t restrict the operation of the proposed DFB QCL structure. This is because the obtained coupling coefficient $K_0 \sim 2.5 \text{cm}^{-1}$ is calculated from Eq. (5.27) with the consideration of this damping effect.

### 5.2.3.2 Surface Plasmon Waveguide structure

An alternative approach to enhance the coupling between the SAW and the optical mode is to use the surface plasmonic waveguide structure commonly used in terahertz QCLs [16]. In such waveguide structure, the optical mode is more confined to the top metal surface due to the existence of the surface plasmon mode. The proposed 3D waveguide structure is shown in Fig. 5-7. Fig. 5-8 shows the mode electric field distribution of the fundamental transverse optical mode and the amplitude of $u_z$ in the surface plasmon waveguide structure, at $f = 1.56 \text{GHz}$. Most of the parameters such as the cavity size, piezoelectric constant, and refractive index are the same as those of the air-waveguide. A larger gain coefficient [166] is required in the calculation to compensate the larger loss caused by the additional top metal contact layer. The thicknesses for active region I, InGaAs layer, active region II is ~1μm, ~0.9 μm, ~1μm, respectively.
Fig. 5-7: 3D schematic diagram of the proposed surface plasmon waveguide structure.

Fig. 5-8: Optical wave and acoustic wave distributions as a function of the depth of the QCL in the surface plasmon waveguide structure. (a) Normalized electric field distribution of the fundamental transverse mode of surface plasmon waveguide structure. (b) Amplitude distribution of $u_z$ along the depth of the waveguide structure.
Further numerical calculations from Eq. (5.27) show that the $u'$ contribution to the coupling coefficient is about one order of magnitude lower than that of the $u_z$, so only $u_z$ is shown for the surface plasmon waveguide structure simulation. For the surface plasmon waveguide structure, $\sim 51\%$ and $\sim 13\%$ confinement factors are obtained for active region I and II, respectively, with a much higher waveguide loss, $\sim 59 \text{ cm}^{-1}$. Due to the high loss, a higher doping density in the active region is required to support lasing. In this case, under $g_0 \sim 80 \text{ cm}^{-1}$, $\alpha_0 \sim 2.1 \text{ cm}^{-1}$, $e_{14} = 0.11 \text{ C/m}^2$, $\lambda_{ac} = 1.6 \mu\text{m}$, $N_0 = 5 \times 10^{16} \text{ cm}^3$, $W = 30 \mu\text{m}$, the resulted coupling coefficient $K_0$ is around $2.6 \text{ cm}^{-1}$, leading to $K_0L_e \sim 0.75$, comparable to the proposed air-waveguide structure.

5.3 Discussion and conclusions

The numerical simulations in the previous sections show that, moderate coupling strengths in the range of $K_0L_e \sim 0.75$ can be achieved with SAW modulation. These coupling strength values could yield single mode laser operation with moderate side mode suppression ratio (SMSR) [166]. It needs to be mentioned that the quality of the ZnO thin film is strongly affected by the deposition method and substrate surface quality; thus, optimizing the growth quality in experiments is crucial in order to achieve the theoretically predicted coupling strength. Compared with other piezoelectric materials, ZnO is adopted as it can be deposited by various relatively easy fabrication processes and it has a high piezoelectricity.

Interestingly, the surface plasmon waveguide has a similar coupling coefficient to the air-waveguide structure. However, the additional top metal contact layer greatly increases the free carrier absorption loss of the optical mode within the cavity. This will result in a larger threshold, a lower slope efficiency and a smaller dynamic range. QCLs based on air-waveguide structures on the other hand have demonstrated relatively good performance [167-168]. In addition, all our simulations assume a perfect ZnO thin film layer. In reality, depositing high quality ZnO thin film on the Au film (surface plasmon waveguides) is more difficult than on the semiconductor (air-waveguides), as the surface
roughness of the Au film further deteriorates the quality of the ZnO thin film. Therefore, it seems that the air-waveguide structure is a better choice for achieving SAW-modulated QCLs. The purpose of this paper is to examine the possibility of DFB mid-infrared QCLs formed by travelling SAW; thus, detailed investigation of the acoustic power is necessary for real experiments. The effectiveness of the SAW grating is directly related to the modulation depth $C$ and the power density $P$ carried by the SAW. The modulation depth $C$ and the acoustic wave power density $P$ for the SAW are defined as $C = K_0/(g_{th}/2)$ and $P = \rho_{mat} \omega_{ac}^2 V_{ac} u_{ac}^2 / 2$, respectively, as shown in [140], where $g_{th}$, $\rho_{mat}$, $V_{ac}$, and $u_{ac}$ stand for the threshold gain, density of the material, acoustic wave velocity, and shear displacement of the material, respectively. The modulation depth $C$ corresponds to $2\xi$ in the bulk shear wave case [140], where $\xi$ represents the relative gain modulation.

For a simple estimate, we look at the case with $C = 0.1$, $P \sim 10\text{ kW/cm}^2$, and $g_{th}L_e \sim 3$. Around 15 mW acoustic electrical power is needed to achieve a displacement $u_{ac} \sim 0.2$ nm for a ridge with a depth of 5 $\mu$m and a width of 30 $\mu$m. For $L_e \sim 3$ mm, which is common for the current device fabrication, the threshold gain for QCLs is $\sim 11 \text{ cm}^{-1}$, which is achievable for conventional mid-infrared QCLs [169]. However, the SAW modulation mechanism will encounter problems when it comes to the THz range. In the THz regime, (e.g. $\lambda_{opt} \sim 100\mu$m, $\lambda_{ac} \sim 15\mu$m), because of the much longer wavelength and $q_{ac}$-dependence of $K_0$ (See Eq. (5.27)), much higher power is needed to obtain a close coupling coefficient $K_0$ close to that of mid-infrared QCLs. Applying $u_z \sim 30$ nm, and shear strain $\sim 1.2\times10^{-2}$, we can obtain acoustic power density $P \sim 2\text{ MW/cm}^2$, much larger than the electrical drive power density. Such high acoustic power density can actually damage the device even before lasing.

It needs to be mentioned that the required acoustic displacement can be achieved by fabricating the IDT directly on top of the waveguide through E-Beam lithography method, as shown in [154]. It seems that other methods, such as direct patterning of the piezoacoustic transducers on the laser facet to generate bulk acoustic waves, can also be applied to modulate the gain of the active region [140]. However, the small size of the laser facet of the mid-infrared ridge QCLs makes this process difficult to realize.
Injection of SAW directly into the active region by using an etch-down waveguide structure (i.e. conversion from SAW to BAW) may provide an alternative method to modulate the QCL, just as shown in [155], while the effectiveness of this approach may be greatly reduced owing to, first, the incident SAW will suffer strong reflection from the end facet due to the large acoustic refractive index contrast; second, a large portion of the incident SAW will be diffracted into the substrate, because of the small laser aperture for SAW coupling.

In conclusion, we have proposed a scheme to achieve tunable, single mode operation of QCLs through SAW modulations and theoretically studied the feasibility. To enhance the interaction between the optical lasing mode and surface acoustic waves, air-waveguide and surface plasmon waveguide structures with two-section active regions, combined with highly piezoelectric (ZnO) thin film are proposed. The results show that coupling coefficients of ~2.5 cm$^{-1}$ can be obtained for both structures, using typical QCLs active region parameters; thus, demonstrating the potential of the proposed schemes for future applications. However, achieving high quality thin film growth is crucial to achieve the theoretically predicted coupling strength; meanwhile fabrication the IDTs directly on top of the active region of the QCLs ridge waveguide also imposes challenges, due to the small size of the ridge width and the electrical isolation between the IDT electrodes and the top metal contact.
Chapter 6
Theoretical investigation of injection-locked high modulation bandwidth mid-infrared quantum cascade lasers

6.1 Introduction of Injected-locked QCLs

Semiconductor laser sources with high modulation bandwidths are always desirable for high speed data transmission systems. However, the modulation bandwidth of direct modulated semiconductor laser is largely limited by relaxation resonance frequency determined by the interactions of the carriers and photons in the laser cavity. Optical injection locking scheme has been shown theoretically [170] and experimentally [171] to be an effective approach to enhance the relaxation resonance frequency of semiconductor lasers, making it a useful method to increase the modulation bandwidth of lasers. With this technique, vertical-cavity surface-emitting lasers (VCSELs) with a 80 GHz intrinsic 3-dB bandwidth [172], quantum-dot (QD) DFB lasers with a 16.3 GHz bandwidth [173], and distributed reflector (DR) lasers with wire-like active regions [174] with a 15 GHz bandwidth have been demonstrated recently.

Quantum cascade lasers (QCLs) are the most promising semiconductor-based mid-infrared and terahertz sources. As they have intrinsic ultrafast intersubband transitions and versatile-designed wavelengths, they are very attractive for high-speed free-space optical (FSO) communication systems [175-176] (two atmospheric transmission windows are in the regimes (3-5 μm) and (8-14 μm), respectively.) and Light Detection and Ranging (LIDAR) applications. Recent advancement of high-performance continuous-wave room-temperature operated QCLs [177-178] has further brought interests of applying QCLs for FSO applications [3]. With direct modulation of 8.1 μm
QCLs, enhanced link stability has been demonstrated in FSO communication system in fog weather due to the less Mie scattering at relatively longer wavelengths, showing that QCL is an ideal source for FSO communication in atmospheric conditions with restricted visibility [3]. Earlier theoretical studies show that the modulation bandwidth of QCLs can reach 100 GHz or even Terahertz regimes [179-181]. However, this is misleading as the value of photon lifetime was inaccurately estimated for practical QCLs. Also in [182], the rate equations for QCLs were not correctly established, as the cascade characteristics of QCLs were not properly considered. Direct modulation of QCLs actually leads to a much lower modulation bandwidth ~10 GHz for some mid-infrared QCLs [183] and ~13 GHz for typical terahertz QCLs [184]. Further increase of the modulation bandwidth of QCLs through other approaches is highly desirable.

In this chapter, we theoretically and systematically investigate the injection-locking of QCLs [27], for the first time to our knowledge, at emitting wavelengths of 4.6 μm [132] and 9 μm [75] corresponding to the two atmospheric transmission windows, respectively. We first examine the direct intensity modulation characteristics of QCLs, using a three-level rate equations model. The results show that no resonant frequency appears in the frequency modulation response, and the maximum modulation bandwidths of round 7 GHz for QCLs at 4.6 μm and 20 GHz for QCLs at 9 μm are obtained. The theoretical analysis is in good agreement with the experimental observations [183]. In addition, we apply the optical injection locking scheme to increase the frequency modulation bandwidth of QCL. Rate equation analysis shows that the obtained modulation bandwidth of injection locked QCLs is about three times higher than that of the directly modulated QCLs for both lasers at 4.6 μm and 9 μm under a 5 dB injection ratio, with modulation bandwidths up to ~30 GHz and ~70 GHz, respectively. With a 10 dB injection ratio, a modulation bandwidth of over 200 GHz can be achieved with the injection locking scheme. A unique feature of injection locked QCLs is that there is no unstable locking range, as opposed to other semiconductor lasers [185]. We attribute this effect to the ultra-short lifetime of the upper laser state.
6.2 Theory of injection-locked QCLs

6.2.1 Direct intensity modulation of QCLs

In this study, we use a three-level rate equation model to describe the dynamic behavior of injection-locked QCLs. It is noted that neglecting the carrier number in the lower laser state is invalid in QCLs, especially for room temperature operation, also to accurately describe the dynamic behavior of QCLs, a self-consistent scattering model [186] is required, which however is too complicated for the present study. We denote the instantaneous carrier numbers in the lower and upper laser state by \( N_2 \) and \( N_3 \), respectively, and the photon number by \( P \). Here the cavity is assumed to have only one longitudinal mode. We noticed that analysis of direct intensity modulation of QCLs with rate equation model has also been investigated in [182]; however, the gain coefficients for carrier rate equations are miscalculated due to the neglect of the cascade characteristics of QCLs. Therefore, the obtained modulation bandwidth in [182] is much larger than that demonstrated in experiments [183]. This conclusion is also supported by the analysis in [187]. The resulted rate equations for QCLs should be:

\[
\frac{dN_3}{dt} = J - \frac{N_3}{\tau_3} - \frac{G}{N_p} (N_3 - N_2) P \\
\frac{dN_2}{dt} = \frac{N_3}{\tau_3} + \frac{G}{N_p} (N_3 - N_2) P - \frac{N_2}{\tau_2} \\
\frac{dP}{dt} = G(N_3 - N_2) P - \frac{P}{\tau_P}
\]

where \( J \) denotes the current injected into the active region divided by electronic charge \( e \), \( G \) is the optical gain coefficient of the entire active region, \( N_p \) is the number of stages, \( \tau_2 \) and \( \tau_3 \) represent the lifetime of lower and upper laser levels, respectively, and \( \tau_P \) is the lifetime of photon in the cavity, expressed as \( \tau_P = \nu_g (\alpha_w + \alpha_m) \) where \( \alpha_w \), \( \alpha_m \), and \( \nu_g \) are the waveguide loss, mirror loss, and group velocity, respectively. The mirror loss can be calculated using \( \alpha_m = \ln(R_1R_2)/(2L) \), where \( R_1 \) and \( R_2 \) are the power reflectivity of the facets 1 and 2, respectively. The group velocity \( \nu_g \) is given by \( \nu_g = c/n_{eff} \), where \( c \) and \( n_{eff} \)
are the speed of light in vacuum and modal effective refractive index, respectively. In our analysis, $G/N_P$ instead of $G$ as in [182] is correctly used in Eqs. (6.1) and (6.2) to represent the optical gain coefficient of one single active region period in QCLs. Denote $G_0 = G/N_P$, we have $G_0 = \Gamma \nu \sigma_{32}/V$, where $\Gamma$ is the optical mode confinement factor for a single period, and $V$ stands for the volume for a period, given by $W L P$, where $W$ and $L$ are the width and length of the active region, and $L_P$ denotes the period length, respectively. The stimulated emission cross section $\sigma_{32}$ is,

$$\sigma_{32} = \frac{4\pi e^2 z_{32}^2}{\varepsilon_0 \eta_{\text{eff}} \lambda_0 (2\gamma_{32})}$$  \hspace{1cm} (6.4)$$

where $e$ is the electronic charge, $z_{32}$ is the dipole matrix element between the upper and the lower lasing levels, $\varepsilon_0$ is the vacuum dielectric constant, $\lambda_0$ is the free-space emission wavelength, and $(2\gamma_{32})$ stands for the full width at half maximum (FWHM) of the optical transition spectrum.

### 6.2.1.1 Steady state analysis

We set the time derivatives of Eqs. (6.1)-(6.3) to zero, and denote the steady state values of $J$, $N_2$, $N_3$, and $P$ as $J_0$, $N_{20}$, $N_{30}$ and $P_0$, then the steady state rate equations can be expressed as:

$$J_0 = \frac{N_{30}}{\tau_3} - G_0 \left( N_{30} - N_{20} \right) P_0 = 0$$  \hspace{1cm} (6.5)$$

$$\frac{N_{30}}{\tau_3} + G_0 \left( N_{30} - N_{20} \right) P_0 - \frac{N_{20}}{\tau_2} = 0$$  \hspace{1cm} (6.6)$$

$$G \left( N_{30} - N_{20} \right) P_0 - \frac{P_0}{\tau_p} = 0$$  \hspace{1cm} (6.7)$$

The steady state population of lower lasing level, $N_{20}$ is given as

$$N_{20} = J_0 \tau_2$$  \hspace{1cm} (6.8)$$

with Eqs. (6.5) and (6.6), $P_0$ can be obtained in the following equation,

$$J_0 \left(1-\eta\right) \frac{N_{30} - N_{20}}{\tau_3} - G_0 \left( N_{30} - N_{20} \right) P_0 = 0$$  \hspace{1cm} (6.9)$$
where $\eta = \tau_2 / \tau_3$. Also, we note that at threshold,

$$ (N_{30} - N_{20}) = \frac{1}{N_p G_0 \tau_p} $$

(6.10)

where $J_0$ is the threshold current ($s^{-1}$), given by $J_0 = 1/G \tau_2 \tau_p (1 - \eta)$. Substituting Eq. (6.8) into Eq. (6.9), one gets the steady state photon number $P_0$,

$$ P_0 = \frac{1}{G_0 \tau_3} \left( \frac{J_0}{J_0} - 1 \right) $$

(6.11)

with Eqs. (6.8) and (6.10), the upper laser level population $N_{30}$ can be written as,

$$ N_{30} = \frac{1}{N_p G_0 \tau_p} + J_0 \tau_2 $$

(6.12)

### 6.2.1.2 Small-signal modulation analysis

The small-signal analysis is necessary in order to investigate the modulation response of QCLs. Assuming small variations $\Delta N_2$, $\Delta N_3$, $\Delta P$ and $\Delta J$ around the steady state values, and substituting $N_2 = N_{20} + \Delta N_2$, $N_3 = N_{30} + \Delta N_3$, $P = P_0 + \Delta P$ and $J = J_0 + \Delta J$ into Eqs. (6.1) – (6.3), we get the rate equations for the small deviations as follows:

$$ \frac{d \Delta N_3}{dt} = \Delta J - \frac{\Delta N_3}{\tau_3} - G_0 (\Delta N_3 - \Delta N_2) P_0 - G_0 (N_{30} - N_{20}) \Delta P $$

(6.13)

$$ \frac{d \Delta N_2}{dt} = \frac{\Delta N_1}{\tau_3} + G_0 (\Delta N_3 - \Delta N_2) P_0 + G_0 (N_{30} - N_{20}) \Delta P - \frac{\Delta N_2}{\tau_2} $$

(6.14)

$$ \frac{d \Delta P}{dt} = G (\Delta N_3 - \Delta N_2) P_0 + G (N_{30} - N_{20}) \Delta P - \frac{\Delta P}{\tau_p} $$

(6.15)

Taking the Laplace transform of Eqs. (6.13) – (6.15), and placing them into a matrix form, we have the following transformed matrix,

$$ \begin{bmatrix} s + f_{11} & f_{12} & f_{13} \\ f_{21} & s + f_{22} & f_{23} \\ f_{31} & f_{32} & s + f_{33} \end{bmatrix} \begin{bmatrix} \Delta N_3 \\ \Delta N_2 \\ \Delta P \end{bmatrix} = \begin{bmatrix} \Delta J \\ 0 \\ 0 \end{bmatrix} $$

(6.16)
where the matrix terms are expressed as

\[
\begin{align*}
    f_{11} &= \frac{1}{\tau_3} + G_0 P_0 \\
    f_{21} &= -\left(\frac{1}{\tau_3} + G_0 P_0\right) \\
    f_{31} &= -G P_0 \\
    f_{12} &= -G_0 P_0 \\
    f_{22} &= \frac{1}{\tau_2} + G_0 P_0 \\
    f_{32} &= G P_0 \\
    f_{13} &= \frac{1}{\tau_P N_P} \\
    f_{23} &= -\frac{1}{\tau_P N_P} \\
    f_{33} &= 0
\end{align*}
\] (6.17)

The normalized modulation response is

\[
H(s) = \frac{\Delta P}{\Delta J (1-\eta) \tau_P N_P} = \frac{s}{A + 1} + \frac{B}{s^3 + Cs^2 + Ds + 1}
\] (6.18)

where

\[
A = \left(\frac{1}{\eta} - 1\right) \frac{1}{\tau_3} \\
B = \frac{\tau_P \tau_2 N_P}{GP_0} \\
C = B \left(2G_0 P_0 + \frac{1}{\tau_3} \left(1 + \frac{1}{\eta}\right)\right) \\
D = B \left(\frac{1}{\tau_2} G_0 P_0 + \frac{1}{\tau_3 \tau_2} + 2GP_0 \frac{1}{\tau_P N_P}\right)
\] (6.19)

### 6.2.2 Injection locking of QCLs

#### 6.2.2.1 Steady state analysis

The injection locking scheme involves two lasers: the slave laser (SL) is optically locked by the master laser (ML) [171]. The differential equation describing electric field inside the slave laser under injection locking was first proposed by Lang and Kobayashi, given as [188]

\[
\frac{d}{dt} E_{SL}(t) - \left\{ j\omega(N) + \frac{1}{2} \left[ N_P G_0 \left(\frac{N}{\tau_P}\right) - \frac{1}{\tau_P}\right]\right\} E_{SL}(t) = f_d E_{ML}(t)
\] (6.20)

where \(f_d\) is the coupling rate between the master laser and the slave laser, approximately expressed as: \(f_d = v_g (1-R)/(2LR^{1/2})\) where \(R\) stands for the power reflectivity of the
injected cavity facet. We denote the electromagnetic fields in the slave laser and the master laser as,

\[ E_{SL}(t) = E_0(t)e^{i(\omega_0 t + \phi(t))} \]  

\[ E_{ML}(t) = E_1(t)e^{i(\omega_1 t + \phi(t))} \]  

where \( E_1(t) \) is taken as a constant, \( \phi(t) \) and \( \phi_1(t) \) are the phases of the two electromagnetic fields, \( (\phi(t) \) is usually set as zero for computing convenience); \( \omega_0 \) and \( \omega_1 \) are the angular frequencies of the slave laser and the master laser, respectively. Substituting Eqs. (6.21) and (6.22) into (6.20), the differential equation can be split into field magnitude and phase rate equations.

\[ \frac{d}{dt} E_0(t) = \frac{1}{2} N G_0 \Delta N(t) E_0(t) + f_d E_1 \cos \phi(t) \]  

\[ \frac{d}{dt} \phi(t) = \frac{1}{2} \alpha N G_0 \Delta N(t) - \frac{f_d E_1}{E_0} \sin \phi(t) - \Delta \omega_{mj} \]  

In the above equations, \( \Delta N \) is the carrier number change due to light injection from the master laser, given by \( N_3 - N_2 \). Different from rate equations for diode lasers, the carrier number in the above equations is the carrier number difference between the upper and lower laser levels, i.e. \( N=N_3-N_2 \). From Eqs. (6.13) and (6.14), and taking into account the carrier number in the lower laser state, the differential equations governing the carrier dynamics are given as,

\[ \frac{dN(t)}{dt} = J - (N + N_2) \frac{2}{\tau_3} - 2E_0^2 G_0 N + \frac{N_2}{\tau_2} \]  

\[ \frac{dN_2(t)}{dt} = (N + N_2) \frac{1}{\tau_3} + E_0^2 G_0 N - \frac{N_2}{\tau_2} \]  

where \( E_0(t) \), \( \phi(t) \), \( N(t) \), and \( N_2(t) \) represent the slave laser’s field magnitude, phase, carrier number difference between the transition states, and carrier number in the lower laser state, respectively. The field magnitude \( E_0(t) \) has been normalized, so that \( |E_0(t)|^2=P(t) \), where \( P(t) \) is regarded as the total photon number for a single longitudinal mode inside the cavity. \( \phi(t) \) is defined as the phase difference between the master and
the slave lasers, i.e. \( \phi(t) = \phi_{SL}(t) - \phi_{ML}(t) \), \( G_0 \), \( N_P \), \( \alpha \), \( J \), \( \tau_2 \), and \( \tau_3 \) are the slave laser’s gain coefficient for one stage, the number of period, linewidth enhancement factor, the injection current, the lower laser state lifetime, and the upper laser state lifetime, respectively. While \( f_d \), \( E_1 \) and \( \Delta \omega_{nj} \) represent the coupling rate, injected electrical field magnitude, and frequency detuning, respectively. The latter is expressed as \( \Delta \omega_{nj} = \omega_n - \omega_0 \). For free-running laser, \( N = N_{th}, N_2 = N_{2th}, E = E_{fr} \), with Eq. (6.26), the carrier number in the lower laser state for free-running slave laser can be obtained,

\[
N_{2th} = \left( E_{fr}^2 \gamma_p + \frac{N_{th}}{\tau_3} \right) \frac{\tau_2 \tau_3}{\tau_3 - \tau_2}
\]  

(6.27)

where we define \( \gamma_p \) as,

\[
\gamma_p = G_0 N_{th}
\]  

(6.28)

Remembering that cavity photon decay rate \( \gamma_{po} \) is given by \( GN_{th} \), so that \( \gamma_p = \gamma_{po}/N_P \). With Eq. (6.25) and the expression of \( \gamma_p \), the threshold current, defined as \( I/e \) (where \( I \) is current, and \( e \) is the electronic charge), and the free-running field magnitude of slave laser are found to be,

\[
J_{th} = N_{th} \frac{1}{\tau_3 - \tau_2}
\]  

(6.29)

\[
E_{fr}^2 = \frac{J - N_{th}}{\gamma_p \frac{\tau_3}{\tau_3 - \tau_2}} \frac{1}{\tau_3 - \tau_2}
\]  

(6.30)

Under the injection-locking region, the carrier number difference \( N \) in the slave laser should decrease, due to the enhanced stimulated emission in the gain medium. The steady state values for \( E \), \( \Delta N \), \( \phi \), and \( N_2 \) under injection locking are denoted as \( E_0 \), \( \Delta N_0 \), \( \phi_0 \) and \( N_{20} \) respectively. The corresponding expressions of \( \Delta N_0 \), \( \phi_0 \) and \( N_{20} \) are given below,

\[
\Delta N_0 = - \frac{2f_d E_i}{N_p G_0 E_0} \cos \phi_0
\]  

(6.31)

\[
\phi_0 = \sin^{-1} \left( - \frac{\Delta \omega_{nj} E_0}{f_d E_i \sqrt{1 + \alpha^2}} \right) \tan^{-1} \alpha
\]  

(6.32)
\[ N_{20} = \left[ E_0^2 (\gamma_p + G_0 \Delta N_0) + \frac{\Delta N_0}{\tau_3} + \frac{N_{th}}{\tau_3} \right] \frac{\tau_2}{\tau_3 - \tau_2} \]  

(6.33)

Applying Eqs. (6.31) - (6.33) into Eqs. (6.25), and setting the time derivative to zero, one gets a cubic equation for the steady state value \( E_0 \). Defining two new parameters,

\[ T_1 = \frac{2\tau_2 - \tau_3}{\tau_3 - \tau_2} \]  

(6.34)

\[ T_2 = \frac{2 + T_1}{\tau_3} \]  

(6.35)

the equation is shown as,

\[ E_0^3 \left( 2G_0N_p\gamma_p + G_0N_p\gamma_pT_1 \right) - E_0^2 \left( 2G_0f_dE_1 \cos \phi_0 \left( 2 + T_1 \right) \right) \]

\[ -E_0 \left( G_0JN_p - N_{th}G_0N_pT_2 \right) - 2f_dE_1 \cos \phi_0 T_2 = 0 \]  

(6.36)

The steady state value of \( E_0 \) can be solved by various numerical methods, given that the phase difference \( \phi_0 \) varies approximately from \(-\pi/2\) to \(\cot^{-1}\alpha\) in the injection locking range, and \( \Delta N_0 \) should be negative.

### 6.2.2.2 Small-signal modulation analysis

Assuming a small variances \( \Delta E \), \( \Delta \phi \), \( \Delta N \) and \( \Delta N_2 \) around the corresponding steady state value, and substituting \( E = E_0 + \Delta E \), \( \phi = \phi_0 + \Delta \phi \), \( N = N_0 + \Delta N \), \( N_2 = N_{20} + \Delta N_2 \) into Eqs. (6.23) – (6.26), one gets the rate equations for the small signal modulation:

\[ \frac{d}{dt} \Delta E = \frac{1}{2} G_0 \frac{\Delta N_2}{\tau_3} \Delta E - f_d E_1 \sin \phi_0 \Delta \phi + \frac{1}{2} N_p G_0 E_0 \Delta N \]  

(6.37)

\[ \frac{d}{dt} \Delta \phi = f_d \frac{E_1}{E_0} \sin \phi_0 \Delta E - f_d \frac{E_1}{E_0} \cos \phi_0 \Delta \phi + \frac{\alpha}{2} N_p G_0 \Delta N \]  

(6.38)

\[ \frac{d}{dt} \Delta N = \Delta J - \left( 4E_0 \gamma_p + 4E_0 G_0 \Delta N_0 \right) \Delta E - \left( \frac{2}{\tau_3} + 2E_0^2 G_0 \right) \Delta N \]

\[ + \left( \frac{1}{\tau_2} - \frac{2}{\tau_3} \right) \Delta N_2 \]  

(6.39)
\[
\frac{d}{dt} \Delta N_2 = \left(2E_0 \gamma' p + 2G_0 \Delta N_0 E_0 \right) \Delta E + \left( \frac{1}{\tau_3} + E_0^2 G_0 \right) \Delta N + \left( \frac{1}{\tau_3} - \frac{1}{\tau_2} \right) \Delta N_2 \quad (6.40)
\]

Converting the rate equations into the matrix form and taking Laplace transform, we have the following matrix:

\[
\begin{bmatrix}
Q_{11} + s & Q_{12} & Q_{13} & Q_{14} \\
Q_{21} & Q_{22} + s & Q_{23} & Q_{24} \\
Q_{31} & Q_{32} & Q_{33} + s & Q_{34} \\
Q_{41} & Q_{42} & Q_{43} & Q_{44} + s
\end{bmatrix}
\begin{bmatrix}
\Delta E \\
\Delta \phi \\
\Delta N \\\n\Delta J
\end{bmatrix} =
\begin{bmatrix}
0 \\
0 \\
0
\end{bmatrix}
\quad (6.41)
\]

In which the matrix elements are

\[
\begin{align*}
Q_{11} &= -\frac{1}{2} G_0 \Delta N_0 N_p \\
Q_{21} &= -f_c E_2 \sin \phi_0 \\
Q_{31} &= 4E_0 \gamma' p + 4E_0 G_0 \Delta N_0 \\
Q_{41} &= -(2E_0 \gamma' p + 2G_0 \Delta N_0 E_0) \\
Q_{12} &= f_c E_2 \sin \phi_0 \\
Q_{22} &= f_c E_2 \cos \phi_0 \\
Q_{32} &= 0 \\
Q_{42} &= 0 \\
Q_{13} &= \frac{1}{2} G_0 N_p \\
Q_{23} &= -\frac{1}{2} G_0 N_p \\
Q_{33} &= \frac{1}{\tau_3} + 2E_0^2 G_0 \\
Q_{43} &= 0 \\
Q_{14} &= 0 \\
Q_{24} &= 0 \\
Q_{34} &= \frac{1}{\tau_2} - \frac{1}{\tau_3} \\
Q_{44} &= \frac{1}{\tau_2} - \frac{1}{\tau_3}
\end{align*}
\]

The direct modulation frequency response is given by,

\[
H(s) = G \frac{s^2 + Es + F}{s^4 + As^3 + Bs^2 + Cs + D}
\quad (6.43)
\]

Where

\[
\begin{align*}
A &= Q_{11} + Q_{22} + Q_{33} + Q_{44} \\
B &= Q_{33} Q_{44} + Q_{34} Q_{33} + Q_{14} Q_{44} + Q_{22} Q_{33} + Q_{23} Q_{44} + Q_{11} Q_{22} - Q_{21} Q_{12} - Q_{31} Q_{13} - Q_{41} Q_{14} \\
C &= Q_{11} Q_{33} Q_{44} + Q_{22} Q_{33} Q_{44} + Q_{23} Q_{11} Q_{22} + Q_{44} Q_{11} Q_{22} - Q_{33} Q_{21} Q_{12} - Q_{44} Q_{21} Q_{12} - Q_{31} Q_{22} - Q_{43} Q_{22} Q_{14} + Q_{12} Q_{13} Q_{23} + Q_{13} Q_{44} Q_{11} - Q_{22} Q_{33} Q_{44} - Q_{41} Q_{34} Q_{11} \\
D &= Q_{11} Q_{22} Q_{33} Q_{44} - Q_{12} Q_{21} Q_{33} Q_{44} + Q_{12} Q_{21} Q_{43} Q_{34} - Q_{13} Q_{31} Q_{22} Q_{44} + Q_{12} Q_{31} Q_{23} Q_{44} - Q_{12} Q_{32} Q_{13} Q_{44} + Q_{13} Q_{23} Q_{34} - Q_{13} Q_{23} Q_{34} Q_{34} + Q_{41} Q_{32} Q_{23} - Q_{43} Q_{31} Q_{23} - Q_{44} Q_{32} Q_{23} \\
E &= -(Q_{13} Q_{23} Q_{44} - Q_{13} Q_{22} Q_{14})/Q_{13} \\
F &= -(Q_{13} Q_{23} Q_{44} - Q_{13} Q_{22} Q_{14})/Q_{13} \\
G &= -Q_{13}
\end{align*}
\]

(6.44)
6.3 Results and discussion

6.3.1 Direct modulations

First, we examine the direct modulation bandwidths of QCLs at different wavelengths in the mid-IR regime. Table 6-1 shows the parameters used in the simulations, of the state-of-the-art mid-infrared QCLs emitting at around 4.6 μm [132] and 9 μm [75], which correspond to the two atmospheric windows in the mid-infrared spectrum range. A simple way to analyze the modulation behavior of QCLs can be done by analyzing the zeros and poles of the normalized frequency response $H(s)$. Table 6-2 and Table 6-3 list the corresponding zeros and poles for the QCLs at 4.6 μm and 9 μm under different injection currents. Three poles can be obtained from the denominator of the frequency response, which are expressed as $p_1$, $p_2$ and $p_3$ respectively. One zero will be obtained from the numerator. Also shown is the calculated 3-dB bandwidth of the modulation response. We also notice that the phenomena that no resonance frequency appears and the calculated direct modulation bandwidth are in good agreement with the experimental observations [183], if the experimental parameters [183] are used in the calculations.

Table 6-1: Characteristic parameters of the state-of-the-art QCLs at 4.6 μm [132] and 9 μm [75] for direct modulation.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Symbol</th>
<th>Value (unit)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Wavelength</td>
<td>$\lambda$</td>
<td>4.6 (μm)/ 9 (μm)</td>
</tr>
<tr>
<td>Width</td>
<td>$W$</td>
<td>11.6 (μm)/12 (μm)</td>
</tr>
<tr>
<td>Length</td>
<td>$L_p$</td>
<td>5 (mm)/ 3 (mm)</td>
</tr>
<tr>
<td>Length of period</td>
<td>$L$</td>
<td>50 (nm)/ 67 (nm)</td>
</tr>
<tr>
<td>Modal effective refractive index</td>
<td>$n_{eff}$</td>
<td>3.27/ 3.27</td>
</tr>
<tr>
<td>Waveguide loss</td>
<td>$\alpha_W$</td>
<td>2.6 (cm$^{-1}$)/ 8.77(cm$^{-1}$)</td>
</tr>
<tr>
<td>Total mirror loss</td>
<td>$\alpha_m$</td>
<td>1.34(cm$^{-1}$)/ 2.23 (cm$^{-1}$)</td>
</tr>
<tr>
<td>Photon lifetime</td>
<td>$\tau_p$</td>
<td>27.6 (ps)/9.91 (ps)</td>
</tr>
<tr>
<td>Upper state lifetime</td>
<td>$\tau_1$</td>
<td>1.77 (ps)/0.66 (ps)</td>
</tr>
<tr>
<td>Lower state lifetime</td>
<td>$\tau_2$</td>
<td>0.26 (ps)/0.14 (ps)</td>
</tr>
<tr>
<td>Optical gain coefficient</td>
<td>$G$</td>
<td>2.93×10^4(s$^{-1}$)/1.2×10^5(s$^{-1}$)</td>
</tr>
<tr>
<td>Number of period</td>
<td>$N_p$</td>
<td>40/40</td>
</tr>
</tbody>
</table>
Table 6-2: Poles, zeros, and $f_{3\text{dB}}$ for QCLs at 4.6 $\mu$m.

<table>
<thead>
<tr>
<th>$I_0$</th>
<th>$p_1$(GHz)</th>
<th>$p_2$(GHz)</th>
<th>$p_3$(GHz)</th>
<th>$z$(GHz)</th>
<th>$f_{3\text{dB}}$(GHz)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.5 $I_{th}$</td>
<td>-2.0</td>
<td>-122.7</td>
<td>-697.5</td>
<td>-552.4</td>
<td>3.4</td>
</tr>
<tr>
<td>2 $I_{th}$</td>
<td>-2.9</td>
<td>-150.0</td>
<td>-759.1</td>
<td>-552.4</td>
<td>5.0</td>
</tr>
<tr>
<td>3 $I_{th}$</td>
<td>-3.9</td>
<td>-190.7</td>
<td>-897.3</td>
<td>-552.4</td>
<td>6.8</td>
</tr>
<tr>
<td>4 $I_{th}$</td>
<td>-4.4</td>
<td>-217.8</td>
<td>-1049.5</td>
<td>-552.4</td>
<td>7.6</td>
</tr>
</tbody>
</table>

Table 6-3: Poles, zeros, and $f_{3\text{dB}}$ for QCLs at 9 $\mu$m.

<table>
<thead>
<tr>
<th>$I_0$</th>
<th>$p_1$(GHz)</th>
<th>$p_2$(GHz)</th>
<th>$p_3$(GHz)</th>
<th>$z$(GHz)</th>
<th>$f_{3\text{dB}}$(GHz)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.5 $I_{th}$</td>
<td>-5.4</td>
<td>-312.7</td>
<td>-1312.5</td>
<td>-907.2</td>
<td>9.4</td>
</tr>
<tr>
<td>2 $I_{th}$</td>
<td>-8.1</td>
<td>-364.5</td>
<td>-1499.1</td>
<td>-907.2</td>
<td>14.0</td>
</tr>
<tr>
<td>3 $I_{th}$</td>
<td>-10.8</td>
<td>-428.8</td>
<td>-1914.4</td>
<td>-907.2</td>
<td>18.6</td>
</tr>
<tr>
<td>4 $I_{th}$</td>
<td>-12.2</td>
<td>-464.7</td>
<td>-2359.4</td>
<td>-907.2</td>
<td>21.0</td>
</tr>
</tbody>
</table>

Fig. 6-1: Normalized modulation response of mid-infrared QCLs. (a) QCLs emitting at 4.6 $\mu$m with parameters shown in Table 6-1. (b) QCLs emitting at 9 $\mu$m with parameters shown in Table 6-1. The 3-dB bandwidth is indicated by the black dashed line.
Fig. 6-1(a) and (b) show the normalized frequency responses of QCLs at 4.6 µm and 9 µm, for \( I = 1.5I_{th}, 2I_{th}, 3I_{th}, \) and \( 4I_{th}, \) respectively, corresponding to the injection currents from just above threshold to roll-over, respectively. Obtaining the poles and zeros of the frequency response, we can transfer it into the following form,

\[
H(s) = \frac{s}{\left(\frac{s}{|p_1|} + 1\right) \left(\frac{s}{|p_2|} + 1\right) \left(\frac{s}{|p_3|} + 1\right)}
\]  

(6.45)

This is the normal form of the Bode plot. Because the first pole is three to four magnitudes smaller than the other poles and zeros, it will mainly determine the modulation bandwidth, so \( H(s) \approx 1/(s/|p_1|+1) \) in the Bode plot. For cubic equations, we have \( p_1p_2+p_2p_3+p_1p_3 = D/B \), and \( p_1p_2p_3=-1/B \), leading to \( 1/p_1+1/p_2+1/p_3 = -1/D \). For \( p_1 \) is much smaller than \( p_2 \) and \( p_3 \), we get \( |1/p_1| \approx |D| \). Substituting \( |1/p_1| \) with \( |D| \) in the expression of \( H(s) \), and keeping in mind that \( D \) is positive in the whole locking region; one can approximate the frequency response as,

\[
H(s) = \frac{1}{sD + 1}
\]  

(6.46)

where \( D \) is given by \( (\tau_p+2\tau_2)+\tau_4(G_0P_0\tau_3) \). The definition of 3-dB bandwidth is defined as \( |H(s)| \) at half of its zero value, i.e. \( |H(s)|_{3dB} = 1/2 \) in the case of normalized frequency response. An approximated expression for 3-dB bandwidth is given as,

\[
f_{3dB} \approx \frac{\sqrt{3}}{2\pi D} = \frac{\sqrt{3}}{2\pi (\tau_p + 2\tau_2) + \frac{\tau_p}{G_0P_0\tau_3}}
\]  

(6.47)

Combining the \( D \) and \( f_{3dB} \), we can easily see that a larger value of \( \tau_p \) leads to a decreased modulation bandwidth. This can also be seen from the simulated frequency responses of QCLs at 4.6 µm and 9 µm. Due to larger optical loss, e.g. free carrier absorption (proportional to \( \lambda^2 \)) and intersubband absorption in the waveguide, the photon lifetime is much shorter for longer emission wavelengths, leading to an increased modulation bandwidth. However, a larger \( \tau_p \) means a decreased optical loss, leading to a higher optical output. Thus, the tradeoff between the modulation bandwidth and the output optical power has to be taken into account when designing QCLs for high speed
modulation applications. Another feature of QCLs is the absence of resonance peak as normally shown in conventional diode lasers. The physics behind it lies in the ultrafast upper state lifetime compared with the photon lifetime, making QCLs an overdamped system, showing no resonance peak in the frequency modulation response.

### 6.3.2 Injection locking modulations

To further enhance the modulation bandwidth of QCLs in the mid-IR regime, we can employ injection locking scheme for QCLs, the theoretical analysis of which is shown in section 6.2.2. All the additional parameters used in injection locking calculation are listed in Table 6-4, for QCLs emitting at 4.6 µm and 9 µm, respectively. The rest of the parameters which are common to direct modulated QCLs are shown in Table 6-1. Though the linewidth enhancement factor of QCLs is expected to be zero [59], the experiments showed different values (0.02 ± 0.2 in [189], -0.44 to 2.29 in [190], and -1.8 to -1.7 in [191]). Without loss of generality, we set the linewidth enhancement factor as unity in the calculations.

**Table 6-4: Characteristic parameters of the state-of-the-art QCL at 4.6 µm [132] and 9 µm [75] for injection locking modulation.**

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Symbol</th>
<th>Value(4.6 µm/9 µm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Linewidth enhancement factor</td>
<td>α</td>
<td>1/1</td>
</tr>
<tr>
<td>Threshold carrier number difference</td>
<td>N</td>
<td>1.19×10^6(#)/1×10^6(#)</td>
</tr>
<tr>
<td>Threshold current</td>
<td>Jth</td>
<td>7.9×10^{17}(s^{-1})/1.92×10^{18}(s^{-1})</td>
</tr>
<tr>
<td>Optical gain coefficient</td>
<td>G</td>
<td>3×10^4(s^{-1})/1×10^5(s^{-1})</td>
</tr>
<tr>
<td>Coupling rate</td>
<td>fd</td>
<td>11.7(ns^{-1})/19.5(ns^{-1})</td>
</tr>
</tbody>
</table>

Under injection locking scheme, four equations are involved in the calculations. This results in a four-order denominator and a two-order numerator in the frequency response. The typical locking maps for both of QCLs at 4.6 µm and 9 µm are illustrated in Fig. 6-2 (a) and (b), respectively, with $J = 4J_{th}$. In our simulations, the boundaries of phase
in the injection locking range are approximately \( \cot^{-1} \alpha \) to \(-\pi/2\), from negative to positive detuning edge. This is derived from rate equations for the field magnitude \( E \) and phase difference \( \phi \), where the noise terms and spontaneous terms are neglected. The calculated locking maps of QCLs are similar to those of diode lasers, where the locking range increases linearly with the increase of the amplitude of the injected optical field, as demonstrated in [192].

![Fig. 6-2: Locking maps for QCLs emitting at (a) 4.6 \( \mu \)m and (b) 9 \( \mu \)m. The parameters used in the simulations are shown in Table 6-1 and Table 6-4.](image)

To examine the frequency response of the injection locked QCLs, we calculate the response curves of QCLs at 9 \( \mu \)m with an injection ratio of 5 dB across the locking range and a frequency spacing of 1 GHz. The corresponding waterfall plot is shown in Fig. 6-3. Table 6-5 lists the accompanying poles, zeros and 3 dB bandwidth of the responses. The frequency response exhibits resonance-like behaviors close to both edges of the locking range (e.g. the green and the black curves, respectively, in Fig. 6-3). However, the reasons behind these two are different. A further explanation can be sought by examining the poles of the frequency response. As illustrated in Table 6-5, when the frequency comes close to the positive frequency detuning edge (black curve in Fig. 6-3), two complex conjugate poles appear. In conventional semiconductor lasers, the imaginary part of the complex conjugate poles gives the resonance frequency of the response, while the real part defines the damping term, so that a peak appears in the
frequency response. This is also the case for QCLs. At the negative detuning edge (dark curve in Fig. 6-3), because all the poles are real (and negative), the peak in the frequency response has to be analyzed by the Bode Plots. According to the Bode Plots theory, zeros will increase the magnitude of the frequency response from its critical frequency and beyond 10 dB per decade, while poles decrease the magnitude at the same rate. For QCLs, the first zero \( z_1 \) is smaller than any of the poles, making the peak in the frequency response apparent, even though all the poles are real.

Fig. 6-3: Normalized frequency response curves versus frequency detuning at a fixed injection ratio \( R=5 \) dB, for the 9 \( \mu \)m QCL.

Table 6-5: Poles, Zeros and 3-dB bandwidth \( f_{3dB} \) for the QCL in Fig. 6-3.

<table>
<thead>
<tr>
<th>( f ) (GHz)</th>
<th>( p_1 ) (GHz)</th>
<th>( p_2 ) (GHz)</th>
<th>( p_3 ) (GHz)</th>
<th>( p_4 ) (GHz)</th>
<th>( z_1 ) (GHz)</th>
<th>( z_2 ) (GHz)</th>
<th>( f_{3dB} ) (GHz)</th>
</tr>
</thead>
<tbody>
<tr>
<td>-5</td>
<td>-1.6</td>
<td>-9.9</td>
<td>-387.2</td>
<td>-1606.2</td>
<td>-0.7</td>
<td>-895.7</td>
<td>43.6</td>
</tr>
<tr>
<td>-4</td>
<td>-2.5</td>
<td>-9.7</td>
<td>-398.7</td>
<td>-1676.0</td>
<td>-2.3</td>
<td>-895.7</td>
<td>18.4</td>
</tr>
<tr>
<td>-3</td>
<td>-3.4</td>
<td>-8.8</td>
<td>-400.8</td>
<td>-1690.0</td>
<td>-3.4</td>
<td>-895.7</td>
<td>15.2</td>
</tr>
<tr>
<td>-2</td>
<td>-5.1</td>
<td>-6.9</td>
<td>-396.9</td>
<td>-1664.3</td>
<td>-4.2</td>
<td>-895.7</td>
<td>15.0</td>
</tr>
<tr>
<td>-1</td>
<td>-5.8+2.4i</td>
<td>-5.8+2.4i</td>
<td>-389.4</td>
<td>-1618.9</td>
<td>-4.7</td>
<td>-895.7</td>
<td>15.6</td>
</tr>
<tr>
<td>0</td>
<td>-5.5+3.6i</td>
<td>-5.5+3.6i</td>
<td>-379.4</td>
<td>-1563.2</td>
<td>-5.1</td>
<td>-895.7</td>
<td>16.6</td>
</tr>
<tr>
<td>1</td>
<td>-5.2+4.5i</td>
<td>-5.2+4.5i</td>
<td>-367.6</td>
<td>-1504.4</td>
<td>-5.3</td>
<td>-895.7</td>
<td>18.0</td>
</tr>
<tr>
<td>2</td>
<td>-4.7+5.3i</td>
<td>-4.7+5.3i</td>
<td>-355.3</td>
<td>-1450.6</td>
<td>-5.4</td>
<td>-895.7</td>
<td>19.2</td>
</tr>
<tr>
<td>3</td>
<td>-4.2+5.9i</td>
<td>-4.2+5.9i</td>
<td>-342.9</td>
<td>-1401.9</td>
<td>-5.5</td>
<td>-895.7</td>
<td>20.6</td>
</tr>
<tr>
<td>4</td>
<td>-3.7+6.5i</td>
<td>-3.7+6.5i</td>
<td>-331.2</td>
<td>-1360.6</td>
<td>-5.5</td>
<td>-895.7</td>
<td>21.8</td>
</tr>
<tr>
<td>5</td>
<td>-3.0+7.1i</td>
<td>-3.0+7.1i</td>
<td>-318.1</td>
<td>-1318.7</td>
<td>-5.5</td>
<td>-895.7</td>
<td>23.4</td>
</tr>
</tbody>
</table>
For conventional semiconductor lasers, increasing slave laser’s bias current is an effective way to enhance the bandwidth of the modulation system [180]. Here we calculate the frequency response at a fixed injection ratio of 5 dB, with the injection current varying 1.5×, 2×, 3×, and 4×$J_\text{th}$, respectively, and a frequency detuning 0.1 GHz away from the negative detuning edge, as shown in Fig. 6-4. The inset is the corresponding pole/zero diagrams.

Table 6-6 and Table 6-7 list the associated poles and zeros of the modulation responses. The increased bandwidth can be attributed to the increased values of poles and zeros of the frequency response, and the much smaller $z_1$ than any of the poles. Similar effects can also be seen by increasing the injection ratio, for which a much broader modulation bandwidth can be achieved. Our calculation shows that for injection ratio $R=10$ dB and frequency detuning of 0.1 GHz away from the negative edge, over 200 GHz modulation bandwidth can be obtained. However, there is a tradeoff between the injection ratio and the slave laser’s injection current, even though both of which can effectively increase the modulation bandwidth. This is because the increased injection current leads to an increased slave laser output power, so that in order to achieve a high injection ratio, a master laser with a higher output power is required, which in some cases is limited in real applications. We noticed that by direct modulating the injection current using an RF source, Pierre et al demonstrated that the cavity resonance frequency of terahertz QCLs can be injection-locked [193]. However, their scheme is different to ours in that the injection signal in our case is the optical signal from the master laser.

<table>
<thead>
<tr>
<th>$I_0$</th>
<th>$p_1$(GHz)</th>
<th>$p_2$(GHz)</th>
<th>$p_3$(GHz)</th>
<th>$p_4$(GHz)</th>
<th>$z_1$(GHz)</th>
<th>$z_2$(GHz)</th>
<th>$f_{3dB}$(GHz)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.5 $J_\text{th}$</td>
<td>-0.94</td>
<td>-3.8</td>
<td>-182.5</td>
<td>-845.0</td>
<td>-0.25</td>
<td>-522.2</td>
<td>27.6</td>
</tr>
<tr>
<td>2 $J_\text{th}$</td>
<td>-1.0</td>
<td>-4.1</td>
<td>-215.1</td>
<td>-1038.2</td>
<td>-0.27</td>
<td>-522.2</td>
<td>30.0</td>
</tr>
<tr>
<td>3 $J_\text{th}$</td>
<td>-1.1</td>
<td>-4.3</td>
<td>-247.4</td>
<td>-1442.1</td>
<td>-0.29</td>
<td>-522.2</td>
<td>31.8</td>
</tr>
<tr>
<td>4 $J_\text{th}$</td>
<td>-1.1</td>
<td>-4.4</td>
<td>-263.1</td>
<td>-1857.5</td>
<td>-0.29</td>
<td>-522.2</td>
<td>32.6</td>
</tr>
</tbody>
</table>
Table 6-7: Poles, zeros, and $f_{3\text{dB}}$ for Fig. 4 (b).

<table>
<thead>
<tr>
<th>$I_0$ (I$_{th}$)</th>
<th>$p_1$ (GHz)</th>
<th>$p_2$ (GHz)</th>
<th>$p_3$ (GHz)</th>
<th>$p_4$ (GHz)</th>
<th>$z_1$ (GHz)</th>
<th>$z_2$ (GHz)</th>
<th>$f_{3\text{dB}}$ (GHz)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.5 $I_{th}$</td>
<td>-1.5</td>
<td>-9.9</td>
<td>-385.1</td>
<td>-1594.2</td>
<td>-0.50</td>
<td>-895.7</td>
<td>59.2</td>
</tr>
<tr>
<td>2 $I_{th}$</td>
<td>-1.6</td>
<td>-11.3</td>
<td>-434.2</td>
<td>-1971.5</td>
<td>-0.54</td>
<td>-895.7</td>
<td>65.2</td>
</tr>
<tr>
<td>3 $I_{th}$</td>
<td>-1.6</td>
<td>-12.5</td>
<td>-482.0</td>
<td>-2753.1</td>
<td>-0.57</td>
<td>-895.7</td>
<td>69.6</td>
</tr>
<tr>
<td>4 $I_{th}$</td>
<td>-1.6</td>
<td>-13.0</td>
<td>-505.1</td>
<td>-3552.0</td>
<td>-0.58</td>
<td>-895.7</td>
<td>71.4</td>
</tr>
</tbody>
</table>

Fig. 6-4: Normalized modulation response of mid-infrared QCLs under injection locking scheme. (a) QCLs emitting at 4.6 μm. (b) QCLs emitting at 9 μm. The inset show the corresponding poles (x) and zeros (●), with the arrow indicating direction of increasing the injection current of the slave laser. The 3 dB bandwidth is indicated by the dashed line.

It is also noted that there is no unstable locking range in QCLs based on the above rate equation analysis, compared to other types of semiconductor lasers. Mathematically, the stable locking condition is satisfied when the eigenvalues of the frequency response function’s denominators are located at the left half of the complex s-plane. In the entire locking region of QCLs, we find that the roots of the denominator are always negative, making the whole locking region a stable locking system. One of the experimental
evidence is that the injected light induced pulsation effect [185], which is a unique sign of the unstable locking condition in diode lasers [194] caused by the reduced damping and the strong correlation between the phase and intensity of photons, has not been observed in QCLs. The reasons are as follows: first, there is no relaxation oscillation in QCLs which has been experimentally verified, owing to the picosecond carrier lifetime of the laser states [183]. It is equivalent to say that the damping for the relaxation oscillation is so high such that the carrier numbers and photons come to a steady state in a much shorter time than those in conventional diode lasers. Second, using the phasor diagram and small signal analysis (here we express the small change of $N$, $P$, and $\phi$ as $\delta N$, $\delta P$ and $\delta \phi$), Henry pointed out [194] that the increase of $\delta P$ causes an increased $\delta \phi$ in relaxation oscillations. On the other hand, at the negative detuning edge, the increase of $\delta \phi$ causes an enhanced mismatch of $E_0$ and $E_1$, leading to a larger cavity intensity change, i.e. larger $\delta P$. This forms a positive feedback between $\delta \phi$ and $\delta P$, which is enhanced as the relaxation oscillations increase. However, there is no relaxation oscillation in QCLs, thus the interaction between $\delta \phi$ and $\delta P$ is reduced. Therefore the large damping effect and the reduced interactions between $\phi$ and $P$, make the unstable locking range in QCLs disappear, as shown in Fig. 6-2.

In summary, theoretical investigation of injection-locked QCLs at wavelengths of 4.6 $\mu$m and 9 $\mu$m are carried out. By using a three-level rate equations model, we find that the maximum modulation bandwidths of round 7 GHz for QCLs at 4.6 $\mu$m and 20 GHz for QCLs at 9 $\mu$m were obtained. It is shown that by applying the injection locking scheme, the modulation bandwidths can be increased by around three times under a 5 dB injection ratio, compared to the direct modulation scheme. The frequency modulation responses were analyzed using the Bode diagram, based on which it shows that increasing the injection ratio and the injected current of the slave laser are effective ways to enhance the modulation bandwidth of the master QCLs. With a 10 dB injection ratio, more than 200 GHz modulation bandwidth can be obtained for QCLs at 9 $\mu$m. Unlike conventional semiconductor lasers, no unstable locking ranges appear in the locking map, which we attribute to the ultra-short lifetime of the upper laser state of QCLs, due to the nature of intersubband transitions.
Chapter 7
Conclusions and future works

7.1 Conclusions

In this thesis, we presented our preliminary explorations of the feasibility of performance improvements on broadly tunable mid-infrared QCLs. To achieve broadly tunable monolithic QCLs, two novel schemes, e.g. slot-QCLs and SAW-QCLs, were proposed and investigated. Meanwhile, the injection-locked QCLs was theoretically studied to improve the output power and modulation performances of the tunable single-mode QCLs.

We have experimentally demonstrated compact and monolithic broadly tunable single-mode QCLs based on slot waveguide structure. The proof-of-concept device is based on the bound-to-continuum design emitting at wavelength of ~10 μm. Combining the temperature and current tunings, the current slot-QCL device demonstrated a tuning range of 77 cm⁻¹, corresponding to ~7.8 % of relative tuning, which is defined as $2 \times \frac{(\lambda_{\text{max}} - \lambda_{\text{min}})}{(\lambda_{\text{max}} + \lambda_{\text{min}})}$, where $\lambda_{\text{min}}$ and $\lambda_{\text{max}}$ are the cut-off shortest and longest wavelengths within the tuning range. The side mode suppression ratio (SMSR) was ~20 dB within the whole tuning range, with peak output ranging from 30 to 100 mW.

We have also proposed and theoretically investigated tunable single-mode mid-infrared QCLs using the SAW modulation mechanism for broad and fast wavelength tuning. To enhance the coupling between the optical mode and the SAW-induced grating, the air-waveguide and surface plasmon waveguide structures with two-section active regions were proposed, together with Zinc Oxide (ZnO) thin film deposited on top of these devices to enhance the piezoelectricity of the laser materials. Our theoretical calculation
shows a coupling coefficient of ~2.5 cm\(^{-1}\) for both waveguide structures. The moderate coupling strength demonstrates the possibility of achieving tunable single-mode emission by using SAW modulation.

Furthermore, we have theoretically studied the high modulation bandwidth injection-locked single-mode QCLs to improve the single-mode QCLs performances in terms of output power and modulation bandwidth. Two high performance mid-infrared QCLs with wavelengths of 4.6 \(\mu\)m and 9 \(\mu\)m were investigated for comparison in detail. Enhanced modulation bandwidths of ~30 GHz and ~70 GHz were obtained for 4.6 \(\mu\)m and 9 \(\mu\)m, respectively, showing a threefold increase for both wavelengths under a 5-dB optical injection ratio. A modulation bandwidth over 100 GHz was also predicted for 9 \(\mu\)m QCLs with an injection ratio of 10-dB. Current analysis shows no unstable locking region. The nonexistence of unstable locking region was attributed to the ultrafast upper laser state lifetime.

### 7.2 Future works

Some foreseeable future works on the previous works are:

1. For the slot-QCLs, further improvements in the laser performance are follow: first, though our first proof-of-concept slot-QCL demonstrated broad tuning range, the need of temperature tuning hinds its application in various fields. Further improvement will be done to achieve broad RT tuning range by optimizing the structures parameters (e.g. slot width and period). Secondly, for spectroscopy applications, the CW QCLs are highly preferred due to their much reduced laser linewidths. The CW-operated slot-QCLs will be investigated through detailed active region and waveguide designs. Thirdly, the active region will be optimized for broader gain bandwidth which is essential for broad tuning range. Finally, electrical driving system for simultaneous control of the refractive indices of front and back slot arrays is to be developed to enlarge the tuning range of slot-QCLs.
2. For the SAW-QCLs, experiment study will be carried out to test its validity of generating tunable single-mode emission from QCLs. However, a number of challenges in the devices fabrication have to be overcome to achieve SAW-QCLs: first, advanced thin film depositing method has to be developed to deposit high quality ZnO thin film on the top of the waveguide structure. Secondly, nanofabrication process for the IDT patterning needs to be explored. Possible nanofabrication approaches include E-Beam lithography and FIB. Thirdly, waveguide structure with split active region needs further optimization for real wafer growth.

3. For the injection-locked QCLs, recent theoretical and experimental results have demonstrated remarkable performance improvements of QCLs in terms of optical power and laser linewidth by employing injection-locking scheme. In our future work, the corresponding experiment on the high modulation bandwidth injection-locking QCLs could be carried out to verify our theoretical predictions. To achieve that, following issues will be solved: first, AR coating having low residual reflectivity in a broad wavelength range is to be investigated to enhance the coupling between the master and slave lasers. Second, CW DFB-QCL will be developed as the slave laser due to its stable single-mode emission and narrow laser linewidth that are beneficial for stable injection locking effect. Thirdly, high frequency modulation system is to be set up for injection-locked QCLs modulation.
References


Express, 15, 15818 (2007).


Publications

Journal papers:


Conference papers:


2. B. Meng, and Q. J. Wang, “Quantum Cascade Lasers of $\lambda \approx 14 \mu m$ Based on Three-phonon-resonance Design,” Photonics Global Conference (PGC 2012), Singapore.

