Wide-band mid-infrared quantum devices based on intersubband transitions

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**NANYANG
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SINGAPORE

**WIDE-BAND MID-INFRARED QUANTUM DEVICES
BASED ON INTERSUBBAND TRANSITIONS**

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Abstract

The infrared region is the cradle of many vital applications in spectroscopy, medicine and communication. In the Mid-Infrared (MIR) region, innovations and significant improvements of the existing technologies call for advanced behaviour such as frequency comb lasers and high-speed photodetectors with high photoresponse at room temperature. Conventional systems are facing significant challenges due to their intrinsic high-speed limitations and the complexity to reach this domain. The development of quantum devices based on Intersubband Transitions (ISBT) has revealed significant advantages in the MIR reviving intense research in these fields. They allow excellent efficiency in the MIR region, and due to their structures based on quantum wells, they permit an easy tunability of the operation wavelength. In comparison to conventional devices, ISBT exhibits a very short carrier lifetime which makes them suitable for high-speed applications.

The objective of this thesis is to develop and study the high-speed behaviour of quantum devices based on intersubband transitions for emission and detection of coherent light in the MIR region. Additionally, thanks to their wide-band properties, frequency combs and injection locking of Quantum Cascade Laser (QCL) using radio-frequency excitation will be investigated.

In this thesis, we first present the context, and why MIR region is critical for many applications, then we will develop the theory of quantum cascade laser, frequency comb lasers and quantum well infrared photodetector using the unique properties of intersubband transitions.

Secondly, we will give an overview of the process and standard characterisations of QCL devices and then investigate their high-speed behaviour using rectification methods. Using high-frequency setups, we will study the influence of a Radio Frequency (RF) injection at the round-trip frequency on the spectra, especially the generation of sidebands in the optical spectrum for QCLs in the two windows atmospheric (MWIR and LWIR). We will then investigate the injection-locking mechanism and, using high-resolution FTIR, analyse the Free-Spectral Range of the spectrum with and without RF injection.

In the third part, we will develop a Quantum Well Infrared Photodetector (QWIP) based on air-bridge contacts to promote the high-speed modulation. Optical characterisation of the devices has been realised to deeply understand the dependence of the responsivity and photoconductive gain with the temperature and high electric field.

Finally, we tested its wide-band operation by detecting the high-frequency modulation of the QCLs previously characterised. The development of both high-speed emitter and receiver at the same wavelength allow a powerful pair for many applications in the MIR region.

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Abbreviations

IR	Infrared
MWIR	Mid-Wavelength Infrared
LWIR	Long-Wavelength Infrared
QCL	Quantum Cascade Laser
QWIP	Quantum Well Infrared Photodetector
MBE	Molecular Beam Epitaxy
MOCVD	Metallo-Organic Chemical Vapour Deposition
ISB	Intersubband
ISBT	Intersubband Transitions
ISB	Intersubband
FTIR	Fourier Transform Infrared
QW	Quantum well
LO	Longitudinal Optical
NEP	Noise Equivalent Power
BLIP	Background-Limited Infrared Performance
BH	Buried-Heterostructure
IV	Current-Voltage
RC	Resistor-Capacitor
RLC	Resistor-Inductor-Capacitor
DC	Direct Current
AC	Alternating current
CW	Continuous-wave
RF	Radio-Frequency
FSR	Free-Spectral Range
BL	Background-Limited
LT	Low-Temperature
HT	High-Temperature

RT	Room Temperature
LIV	Light-Current-Voltage
GVD	Group-Velocity-Dispersion
WPE	Wall Plug Efficiency
QCD	Quantum Cascade Detectors
WKB	Wentzel-Kramers-Brillouin
BC/BQC/BB	Band-to-Continuum/Band-to-QuasiContinuum/ Band-to-Band
ICP	Inductively Coupled Plasma
PECVD	Plasma-Enhanced Chemical Vapor Deposition
FM	Frequency-Modulated
AM	Amplitude-Modulated
RNGH	Risken-Nummedal-Graham-Haken
FWM	Four-Wave Mixing
AR	Active-Region
NDC	Negative Differential Conductivity
FWHM	Full Width at Half Maximum
FPA	Focal Plane Array
MCT	Mercury-Tellurium-Cadmium
FOV	Field Of View

Symbols

<u>Symbol</u>	<u>Description</u>	<u>Symbol</u>	<u>Description</u>
λ	Wavelength	ΔN	Volume population inversion
k_B	Boltzmann constant	q	Electron charge
h or $\hbar$	Planck constant	T	Temperature
E	Energy	I	Intensity
τ_i	Carrier lifetime at the state i (escape, capture, relaxation...)	Q	Quality factor
ε	Permittivity	N	Electron density
n_i	Electronic densities at the state i	v	Velocity
ν or f	Frequency	L	Length
c	Speed of light	$T(X)$	Transmission probability
η	Efficiency	V_b	Barrier potential
J	Current density	F	Electric field
I	Current	x, y, z	Dimension. With z growth axis.
α	Losses	V_i	Potential at i
z_{ij}	Dipole matrix element between the state i and j	p_i	Probability of i (escape, capture...)
γ_{ij}	Full Width at Half Maximum (FWHM) of the transition i and j	N_{QW}	Number of quantum well
Γ	Overlap factor	g_{photo}	Photoconductive gain
β	Fraction of spontaneous emission emitted	S_i	Current noise power spectral density
n	Refractive index	R	Responsivity or Resistance
σ	Cross-section of the gain per unit of time	C	Capacitance

g_c	gain cross section	NEP	Noise Equivalent Power
R	Reflection coefficient	D^*	Detectivity
Δn	Population inversion	$\Phi_{BG,300K}$	photon flux of a blackbody at 300 K
ω	Pulsation (sometimes named as frequency)	A or S	Surface of the detector/lasers
N_p	Number of periods	P	Power
$\hbar\Delta$	Detuning energy from the resonance	$h(\omega)$	Transfer function
$2\hbar\Omega_{ij}$	anti-crossing gap between state i and j	d	Width of the active region
m^*	Effective masses	$\Delta\omega$	Locking range
$\chi^{(i)}$	i orders susceptibilities	S	Photon flux density
Δk	Phase-matching condition		

Chapter 1 Introduction

1.1. Motivation

1.1.1. Motivation and objective of this thesis

The infrared region is an important area for many applications. Some promising innovations include the use of frequency comb lasers and high-speed modulations of emitters and detectors of coherent light in the MIR. However, conventional systems are facing significant challenges due to their intrinsic frequency response limitations and the complexity to reach this domain. Quantum devices based on ISBT show substantial advantages such as very short lifetimes, making them one of the only devices allowing high-performance and high-speed behaviours at the same time.

The objective of this thesis is to investigate and develop the high-frequency behaviour of two primary quantum devices. These devices are Quantum Cascade Laser (QCL) for the emission and Quantum Well Infrared Photodetectors (QWIP) for the detection. This project aims to reach a high-speed behaviour superior to 22 GHz for both devices corresponding to the beating signal of a 2 mm laser. This wide-band allows to investigate the beating signal of the Fabry-Perot modes of the spectrum and therefore, the frequency comb behaviour of the QCL which appear thanks to the four-wave mixing promoting phase and frequency locked mechanism. Moreover, we will also study the injection locking mechanism achieved by RF injection at the round trip of the cavity. Furthermore,

the high-speed photodetectors should achieve high-performance at room temperature allowing the detection and the high-frequency modulation up to 300 K.

1.1.2. Collaboration and contribution

These works have been possible thanks to the collaboration we had during my PhD candidature. The principal part of my thesis was realised in School of Electrical and Electronic Engineering at Nanyang Technological University in Singapore in Qi Jie Wang's group and at Université Paris Diderot where I have been invited to work on the process and characterisation of intersubband devices with the international research group of Carlo Sirtori in the laboratory Matériaux et Phénomènes Quantiques in Paris, France.

The experiments using the high-resolution FTIR were performed in collaboration with Pascale Roy at Synchrotron SOLEIL and Fridolin Kwabia at LISA. Moreover, finally, the growth of the QWIP wafer in Chapter 3 was done at ETH thanks to M. Beck and J. Faist.

Publications

Broadly continuously tunable slot waveguide quantum cascade lasers based on a continuum-to-continuum active region design (published)

Bo Meng, Yong Quan Zeng, Guozhen Liang, Jin Tao, Xiao Nan Hu, Etienne Rodriguez, and Qi Jie Wang

Coherent emission from integrated Talbot-cavity quantum cascade lasers (published)

Bo Meng, Bo Qiang, Etienne Rodriguez, Xiao Nan, Hu, Guozhen Liang, and Qi Jie Wang

Room temperature, wide-band Quantum Well Infrared Photodetector for microwave optical links at 4.9 μm wavelength (published)

Etienne Rodriguez, Alireza Mottaghizadeh, Djamal Gacemi, Daniele Palaferri, Zahra Asghari, Mathieu Jeannin, Angela Vasanelli, Azzura Bigioli, Yanko Todorov, Mattias Beck, Jerome Faist, Qi Jie Wang, and Carlo Sirtori

Active tunability of the free-spectral range under injection-locking of Mid-Infrared Quantum Cascade Laser by direct microwave modulation (in preparation)

Etienne Rodriguez, Alireza Mottaghizadeh, Djamal Gacemi, Mathieu Jeannin, Zahra Asghari, Qi Jie Wang and Carlo Sirtori

Temperature dependence of the photoconductive gain in Quantum Well Infrared Photodetector at 4.9 μm (in preparation)

Etienne Rodriguez, Alireza Mottaghizadeh, Mathieu Jeannin, Daniele Palaferri, Azzura Bigioli, Zahra Asghari, Djamal Gacemi, Maria Amanti, Mattias Beck, Jerome Faist, Qi Jie Wang, and Carlo Sirtori

Conference:

ITQW 2017: Room temperature, high-frequency mid-infrared quantum well infrared photodetector at 4.7 μm (poster)

Etienne Rodriguez, Alireza Mottaghizadeh, Daniele Palaferri, Maria Amanti, Zahra Asghari, Djamal Gacemi, Azzura Bigioli, Mattias Beck, Jerome Faist, Qi Jie Wang, and Carlo Sirtori

Photonics West 2018: Ultrafast modulation of mid-infrared buried heterostructures quantum cascade lasers (oral presentation)

Carlo Sirtori, A. Mottaghizade, E. Rodriguez, D. Gacemi, D. Palaferri, A. Bigioli, Z. Asghari, Y. Todorov, and A. Vasanelli

IQCLSW 2018: Room temperature, wide-band Quantum Well Infrared Photodetector for microwave optical links at 4.9 μm wavelength (oral presentation)

Etienne Rodriguez, Alireza Mottaghizadeh, Djamal Gacemi, Daniele Palaferri, Zahra Asghari, Mathieu Jeannin, Angela Vasanelli, Azzura Bigioli, Yanko Todorov, Mattias Beck, Jerome Faist, Qi Jie Wang, and Carlo Sirtori

1.1.3. Scope and structure of the thesis report

In this thesis, we will concentrate on the development of wide-band QCL and QWIP in the MIR region. We will also study the parameters of our QCL under external radio frequency signal acting as a master oscillator and the temperature dependence of the responsivity of our QWIP.

After having introduced the motivation and the basic physics involved in this work, we will continue in **Chapter 1** to present a global view of the history, theory and applications of Quantum Cascade Lasers (QCL) and Quantum Well Infrared Photodetectors (QWIP). These both technologies are based on intersubband transitions and emit or detect in the two atmospheric windows in the MIR and LWIR. We will also study frequency comb in QCLs and the behaviour of the optical modes and the beating signal under a master oscillation such as radio frequency signals.

The first part of **Chapter 2** will concern the study of the transient time using rectification measurement and highlight the high-speed behaviour of our QCLs. Then, by using its unique high-frequency ability, investigate its parameters under RF injection at the round-trip frequency such as the evolution of the optical spectra and the variation of the free-spectral range using a high-resolution FTIR. Moreover, high-resolution measurements will give valuable information about its frequency comb performances with or without external injection.

In **Chapter 3** we will describe in detail our work on quantum well infrared photodetector starting by the sample structure and device processing for high-speed behaviour, followed by the measures of the photocurrent, responsivity, photoconductive gain and other background-limited performances of our devices. The last part will be dedicated to the high-speed behaviour of our system.

Finally, the **Chapter 4** will be the conclusion of these works.

1.1.4. Key contributions of this thesis

The key contributions of our work during this PhD candidature is the creation and characterisation of high-speed QCLs and QWIPs in the MIR region. Moreover, important works have been realised on frequency comb mechanism by showing a constant and stable free-spectral range over the spectrum and on injection locking where the RF injection permits a tunability of the free-spectral range over the locking range. Regarding the QWIP, additional to the high-speed modulation up to 30 GHz (limited by the electrical system), we also put in evidence the dependence of the responsivity and photoconductive gain with the temperature and the electric field.

1.2. Background

1.2.1. Infrared domain

The electromagnetic spectrum is the range of frequencies or wavelength of the electromagnetic radiations. It is often displayed from extremely low to high energy radiation (from few Hz to exaHertz), and they have been separated, empirically, in different regions as seen in [Figure 1-1](#) based on the different characteristic of the photon

energy regarding the interactions with the matter and the applications. In this thesis, we will explore a part of the Infrared (IR) spectrum which spreads from hundreds of nanometers up to hundreds of micrometres. The Mid-infrared (MIR) region will be more in the centre of our interest, and more precisely the two first atmospheric windows, included in the Mid-Wavelength Infrared (MWIR) from 3 to 5 μm and Long-Wavelength Infrared (LWIR) from 8 to 15 μm .

Figure 1-1: Energy bands of the electromagnetic spectrum.

The first experiment done in the infrared region dates back to the February 11th, 1800 where William Herschel observed radiations less energetic than the visible [1]. The difficulty in accessing this energy explained the little interest in this field up to the invention of the semiconductor laser in the earlier years of the 1960s. This innovation considerably changes the story and allowed a significant rallying in this field. Some fundamentals advances in the emission [2] and the detection [3] areas permitted a further exploration of the IR world and had contributed to some of the leading innovations in our society such as in telecommunication, spectroscopy, medicine, astronomy and agriculture.

1.2.2. Applications in the MIR region

The two atmospheric windows, shown in *Figure 1-2* (a), are essential for many applications in both emission and detection areas:

In the visible, most objects are secondary sources, they reflect or diffuse the light coming from the sun or other primary sources. However, in the IR domain, thermal emissions are the principal sources which are well described by Plank law explained in the next paragraph. This property allows to:

- Observing a scene without any other light, aside from the radiation of the objects itself; widely used in security surveillance.
- Study the fluctuation of the temperature of an object without contact; used to localise cancers or thermal leakage of buildings.
- Guide missiles using the thermal emission of a motor as a target and disturb IR detectors in military areas.

The spectroscopy is another powerful example of the high interest for these two-atmospheric windows. As shown in *Figure 1-2* (a) and (b), many molecules have their fundamental roto-vibrational modes in these regions. This phenomenon, combined with free water absorption, allows a very sensitive gas sensing for environmental purposes such as air pollution and investigation of volcanic emission [4]. Essential applications have been found in medical to determine the health and disease of the patient by breath analysis [5], diagnostic diabetes [6], investigate lung [7], breast cancer [8] and also leak detection [9].

Figure 1-2: (a) Absorption lines of H₂O molecule in the MIR with the two-atmospheric window in blue. Absorption lines of key molecules; in the first atmospheric window (b) and the second atmospheric window (c). (Data compiled using the HITRAN 2004 databased [10] from [11]).

Another primary application for this domain is the free-space communication which allows the transmission of data without physical support. Beyond the high transmission rate, higher wavelength minimises the losses due to the Rayleigh scattering which is proportional to $1/\lambda^4$. Free-space communication has already been realised using Peltier-cooled QCLs at 9.3 μm [12], and extremely high rates can be transferred using the high-frequency properties of the QCLs and the frequency comb technique [13] as emitters and high-frequency QWIP as receivers.

1.2.3. Blackbody radiation

The blackbody radiation is the theoretical electromagnetic radiation of an object perfectly absorbent, opaque and non-reflective body in thermodynamic equilibrium. The spectrum of the thermal radiation is described by Planck's law [14] and depends only on the

temperature of the object as shown in Figure 1-3. The blackbody spectral radiance is described as the power emitted per unit of area of the opaque object, per unit of solid angle corresponding to the measure of the radiation and per unit of frequency.

$$L(\nu, T) = \frac{2h\nu^3}{c^2} \frac{1}{e^{\frac{h\nu}{k_B T}} - 1} \quad (1-1)$$

Where h is the Planck constant, ν the frequency, c the speed of light, k_B the Boltzmann constant and T the temperature of the blackbody.

Figure 1-3: Blackbody spectral radiance for different temperature as a function of the wavenumber.

As seen in Figure 1-3, the blackbody radiation, very well described by Planck's laws, has a broadband spectrum and the peak position is shifted toward the higher energy with the increase of the temperature. If the temperature of the object is well known, it becomes a standard calibration source for all photoconductive devices in the MIR. This mechanism corresponds to the basis of the thermal imaging.

1.2.4. Intersubband versus Interband transitions

After the development of semiconductor technology, Herbert Krömer suggested incorporating semiconductor materials into heterojunctions [15], [16], [17]. These experiments remained almost impossible until the invention in the early seventies, of the Molecular Beam Epitaxy (MBE) [18] and the Metallo-Organic Chemical Vapour Deposition (MOCVD) [19], [20]. These new technologies allow an extremely low deposition rate which leads to growing epitaxial layers with a precision of the atomic level by, thermal evaporation of materials in ultra-high vacuum conditions for MEB and chemical vapour deposition at very high pressure for MOCVD. A few years later, Barry Levine at the Bell Labs demonstrated the first Quantum Well Infrared Photodetector (QWIP) [21], followed by the first Quantum Cascade Laser (QCL) demonstrated by Capasso group in 1994 [22].

As described in [Figure 1-4](#), we can separate the physics of infrared photoconductors and emitters into two main transitions:

- Interband transition is illustrated in [Figure 1-4](#) (a). They are usually constituted by homojunction or heterojunction semiconductors where the transitions take place between the conduction and valence energy bands, described by Bloch wavefunction [23]. The conduction and valence band are separated by the energy gap of the semiconductor, in the case of a direct band gap, this separation corresponds to the optical transition which is about 1.42 eV for the GaAs [24]. The optical properties are then strictly linked to the semiconductor gap which is also defined as the semiconductor gap slavery [25].

- Intersubband (ISB) transition overcomes this physical limitation by using heterostructure based on junctions of different semiconductors. In this case, the optical transitions occur between the confined electronic states in the subbands of the conduction band as seen in Figure 1-4 (b). This property offers a remarkable degree of freedom for the choice of the wavelength of the emission or detection and for the high-speed behaviour but limits the use to lower photon energy as shown in Figure 1-5.

Figure 1-4: Comparison of (a) interband and (b) intersubband transitions.

Thanks to its physics, Intersubband Transitions (ISBT) have unique properties which make them suitable for infrared and high-speed applications:

- The gap between the confined energies of the Quantum Well (QW) can be tuned by engineering the thickness of the QWs. Low energy transitions can be achieved with excellent efficiency, and the absorption or emission wavelength can be

changed or cascaded in the same structure. This phenomenon will be explained in detail in Figure 1-5.

- Similar parabolicity of the dispersion curves of intersubband transitions exhibits an atomic-like joint density of state leading to symmetric and narrow spectra which can be broadened by the interface roughness scattering and the interaction between the Longitudinal Optical (LO) phonons and electrons of the structure [26].
- ISBT also have an inherent short carrier lifetime which allows the high-speed modulation for QCL and high-speed detection for QWIP [27], [28], [29] which will be clarified in details in the next chapters.

The first point is one of the most exciting properties of the ISB. Indeed, the possibility to tune the absorption or emission wavelength is a powerful tool for many applications which require a specific wavelength.

1.2.5. III-V materials

Figure 1-5: Conduction band offset ΔE_c for different material alloy.

Inset: lattice constant and bandgap for the main III-V semiconductors in quantum well structures. (arranged and reprinted from [30], [31], [32]).

Nevertheless, the energy of the photons is limited by the conduction band offset, i.e. the depth of the QW which depends on the alloy used as displayed in [Figure 1-5](#). Therefore, by a clever choice of III-V materials and considering the constant lattice, it is possible to reach much lower wavelength with high-performance. InGaAs/AlAsSb can be used for wavelength included between 3 and 5 μm , where H. Cockburn group realised room temperature laser emitting at 3.1 μm [33]. Thanks to its important ΔE_c and low effective

mass, InAs/AlSb heterostructure allows the emission of $3\text{ }\mu\text{m}$ [34], [35] as well as $19\text{ }\mu\text{m}$ [36].

Aluminium fraction

To fulfil the gap between the different III-V materials combinations, a strained compensation can be done by changing the Al fraction x which comes to increasing or decreasing the depth of the quantum well. Therefore, Al fraction in the InGaAs/InAlAs structure can permit a gap up to 0.75 eV, QCL with a strained compensated active region using $\text{In}_{0.7}\text{Ga}_{0.3}\text{As}/\text{Al}_{0.4}\text{In}_{0.6}\text{As}$ emitting at $3.6\text{ }\mu\text{m}$ has been realised [37]. This property is often used in the QWIP to be able to reach wavelength absorption not reachable by lattice-matched structures as shown in *Figure 1-6* (a) and (b).

Figure 1-6: (a) Al fraction in the barrier as a function of the well width. Theoretical and experimental (in parentheses) values peak detection wavelength for a doping density of $5 \cdot 10^{11}\text{ cm}^{-2}$ in the QW. (b) Spectral response of QWIPs covering the two atmospheric transmission windows (reprinted from [38]).

Figure 1-6 represents the Al fraction in the case of GaAs well and AlGaAs barrier which permits to cover from 220 meV (5.7 μm) to 41 meV (30 μm). Extending, even more, the wavelength is not beneficial due to the better efficiency of other structure [38].

1.3. Quantum Cascade Lasers

1.3.1. History

As described in Figure 1-4 and Figure 1-5, the conduction band offset is the limiting factor regarding the energy of the photons emitted by the QCL, different materials are used to reach the different part of the IR spectrum. The first QCLs were discovered by J. Faist in 1994 utilising a lattice matched InGaAs/AlInAs structure on InP substrate [22]. Since their invention in the Bell-Labs, QCLs have been intensively studied, and their performances have continued to evolve regarding the output power, wall plug efficiency, working temperature and spectrum width. Several improvements have been implemented, and innovative structures were discovered. First, Faist et al. introduced “three-well-vertical-transition” allowing better carrier injection efficiency due to a better spatial overlap of injector ground state and the upper laser state, the first pulsed operation at Room Temperature (RT) was reported using this design [39]. In 1997, Scamarcio et al. proposed “superlattice active region design” [40], followed by two novel designs “bound-to-continuum” and “two-phonon resonance” studied by Faist group in early 2000 [41], [42] which, so far, are considered as optimal designs. Both approaches improved the carriers extraction and thus, achieve a better population inversion. This design combined with a better waveguide thermal extraction permits to demonstrate the first QCLs at RT [43].

1.3.2. State-of-the-art

Figure 1-7: Total Wall Plug Efficiency (WPE) for pulsed and CW operation for several heat sink temperatures indicated by blue to red colours, the outlined black line shows devices with one or both facets coated (WPE estimated from highly cited articles in years 1994-2012 with an error expected to be about 30%, reprinted from [44]).

The two main figures of merit of the laser are the efficiency and the output power. The Wall Plug Efficiency (WPE) is critical for all room-temperature behaviour. Indeed, the electrical power not converted to light will be transformed on heat and degrade the performance of the laser. The evolution of WPE for pulse and CW operations laser is shown in [Figure 1-7](#) where we can see that some lasers reach a WPE higher than 50% which means that this laser emits more light than heat. [45], [46]

Recently “three-phonon resonance” was demonstrated by Wang et al. in 2009 [47] reducing, even more, the laser state lifetime by adding another LO phonon for extraction of the carrier. This design showed 19% WPE at RT at around 7 μm [48].

1.3.3. Physics of quantum cascade lasers

Active region and injector

Quantum cascade lasers are unipolar semiconductor devices based on intersubband transitions. As explained previously, we can tune the emission wavelength of the laser by a smart choice of materials, Al factor and width of the quantum wells of the active region. Therefore, QCLs currently cover a wavelength range from the SWIR at around 2.5 μm to the THz at about 250 μm . The active area of QCLs is composed of an alternation of hundreds of thin layers of two semiconductors which is generally grown on an InP substrate by molecular beam epitaxy or metalorganic chemical vapour deposition. Figure 1-8 (a) shows a Scanning Electron Microscope (SEM) picture of the active region and its schematic representation in Figure 1-8 (b).

Figure 1-8: (a) SEM picture of the active region of a QCL growth by MEB, dark layers are the barrier and white the quantum well, (b) the schematic representation of the SEM picture with the injector and active area (extract from [49]).

QCLs are generally composed of about 20 periods per structure and as seen in [Figure 1-8](#) (a) and (b) consisting of two main areas:

The active region: designed to force the electrons to go from the higher energy level to the lower energy level by emitting a photon.

The injector area: the quantum wells and barriers are designed to allow a wide mini-band to guide the electron from the first active wells to the next active wells by tunnelling effect, this phenomenon will be detailed in paragraph **1.3.5. Resonant tunnelling and Tunnelling effect.**

The barrier width is vital in the tunnelling process; a wide barrier does not permit tunnelling effects while a relatively thin barrier may allow the coupling effect between

the wavefunctions. If the wavefunctions are aligned, there is the creation of a continuum of energy called miniband surrounded by minigap which guide electrons through the barrier by tunnelling effect as displayed in [Figure 1-8](#) and [Figure 1-9](#).

1.3.4. Rate equations

The emission zone of a quantum cascade laser can be illustrated as a three states system as shown in [Figure 1-9](#). The rate equations are expressed using the rate model [50], [51].

Figure 1-9: Schematic representation of the active region based on a three-level system with the lifetimes involves for the different states in point of view of rates equation model.

The laser parameters as the population inversion, gain and current density can be calculated using the rate equation model:

$$\frac{dn_3}{dt} = \eta \frac{J}{e} - \frac{n_3}{\tau_3} - S g_c(n_3 - n_2) \quad (1-2)$$

$$\frac{dn_2}{dt} = \frac{n_3}{\tau_{32}} + S g_c(n_3 - n_2) - \frac{n_2 - n_2^{th}}{\tau_2} \quad (1-3)$$

$$\frac{dS}{dt} = \frac{c}{n} \left[(g_c(n_3 - n_2) - \alpha_{tot}) S + \beta \frac{n_3}{\tau_{spont}} \right] \quad (1-4)$$

With $\tau_3^{-1} = \tau_{32}^{-1} + \tau_{31}^{-1} + \tau_{esc}^{-1} + \tau_{spont}^{-1}$ and $\tau_2^{-1} = \tau_{21}^{-1} + \tau_{2inj}^{-1}$.

Where n_i corresponds to the electronic densities, τ_i the carrier lifetime at the state i , J the current area density, η the injection efficiency of the injector and S the photon flux density per period and unit.

- $n_2^{th} = n_{inj} e^{-\frac{\Delta_{inj}}{k_B T}}$ represents the electrons thermally activated from the Fermi level of the following injector back to state $n=2$ and n_{inj} the injector ground state population.
- Δ_{inj} the voltage defect which is the energy difference between the lower laser state and the next injector ground state.
- τ_{spont} is the spontaneous lifetime.
- β is the fraction of spontaneous emission emitted into the lasing mode which can be neglected ($\beta = 0.001$). Indeed, in ISBT the relaxation time of the spontaneous emission is in the order of magnitude of nanoseconds while the non-radiative relaxation time is in the order of magnitude of picoseconds.
- α_{tot} is the total mode losses such as $\alpha_{tot} = \alpha_w + \alpha_m$. With α_w the waveguide losses and $\alpha_m = -(1/2L) * \ln(R_1 * R_2)$ the mirror losses where R_1 and R_2 are

the reflection coefficient of the laser facets. For cleaved facets, they can be calculated using Fresnel formula with an effective refractive index of 3.2 which gives 27% of reflection (more precise calculation can be done without assuming a plane wave propagation and give a lower value of 23% [52]).

g_c correspond to the gain cross section written as:

$$g_c = \Gamma \frac{4\pi e^2}{\epsilon_0 n_{eff} \lambda} \frac{z_{32}^2}{2\gamma_{32} L_p} = \frac{\sigma * n_{eff}}{c} \quad (1-5)$$

Where $\sigma = \Gamma \frac{4\pi e^2}{\epsilon_0 n_{eff}^2 \lambda} \frac{z_{32}^2 * c}{2\gamma_{32} L_p}$ is the cross-section of the gain per unit of time and Γ is the total overlap factor between the active region and the optical mode in the cavity, z_{32} is the dipole matrix element between the state 2 and 3, γ_{32} is the Full Width at Half Maximum (FWHM) of the transition and L_p is the length of the period. In the steady state the value $(n_3 - n_2)$ is proportional to J [50].

For a current below the threshold, no photon is generated in the cavity, therefore the photon flux density is zero ($S=0$).

The steady-state regime gives $\frac{dn_3}{dt} = \frac{dn_2}{dt} = 0$, therefore the equations (1-2) and (1-3) give:

$$n_3 = \frac{\eta * J}{e} \tau_3 \quad (1-6)$$

$$n_2 = \frac{\tau_2}{\tau_{32}} n_3 - n_2^{th} \quad (1-7)$$

Using the two-last equations, the population inversion $\Delta n = n_3 - n_2$ is:

$$\Delta n = \frac{\eta * J}{e} \tau_{eff} - n_2^{th} \quad (1-8)$$

With $\tau_{eff} = \tau_3(1 - \frac{\tau_2}{\tau_{32}})$, corresponding to the effective upper state lifetime.

To have the population inversion we need $\tau_{eff} > 0$ and therefore, $\tau_{32} > \tau_2$. This condition can be improved by setting the transition between 2 and 1 in resonance with the energy of the LO phonon of the lattice (~ 36 meV in GaAs). This effect will accelerate the depopulation of the lower laser state and thus improve the population inversion. n_2^{th} is related to the thermal effect, the “re-population” of the lower laser state will increase exponentially with the temperature and in consequence decrease the population inversion which will deteriorate the laser performances.

At the threshold current density $g_c * \Delta n = \alpha_{tot}$ which gives:

$$\Delta n = \frac{\alpha_{tot} * c}{\sigma * n_{eff}} \quad (1-9)$$

Using equation (1-8) and (1-9) we can express the threshold current density as:

$$J_{th} = \frac{e}{\eta * \tau_{eff}} \left(\frac{\alpha_{tot} * c}{\sigma * n_{eff}} + n_2^{th} \right) \quad (1-10)$$

Before the threshold; the higher the current, the higher will be the electric field and thus more aligned are the injector state and upper laser state. Therefore, until arriving at the threshold current, the population inversion increases but when the lasing point ($S > 0$) is achieved the Δn remain constant. Beyond the threshold, the photon density of a period increases linearly with the current injected. The slope efficiency of the optical power per facet is:

$$\frac{dP}{dI} = N_p \hbar \omega \frac{c}{n_{eff}} \alpha_m \frac{dS}{dJ} = \frac{N_p \hbar \omega}{e} \frac{\alpha_m}{\alpha_{tot}} \frac{\tau_{eff}}{\tau_{eff} + \tau_2} \quad (1-11)$$

The efficiency slope for a QCLs is proportional to the laser photon energy $\hbar\omega$, the internal $(\frac{\tau_{eff}}{\tau_{eff} + \tau_2})$ and external $(\frac{\alpha_m}{\alpha_{tot}})$ quantum efficiency and the number of periods N_p .

1.3.5. Resonant tunnelling

After describing the phenomenon happening in the active region, we will explain the second phenomenon to understand the QCL physics: the resonant tunnelling effect. This effect is the basis of the carriers injection and extraction in the structure. It has been described using the tight-binding approximation and the density matrix formalism introduced by Kazarinov and Suris [53], [54], used and applied to QCL by Sirtori et al. [55].

$$J = qn_s \frac{2|\Omega_{g3}|^2 \tau_{\perp}}{1 + \Delta^2 \tau_{\perp}^2 + 4|\Omega_{g3}|^2 \tau_{\perp} \tau_3} \quad (1-12)$$

With $\hbar\Delta$, the detuning energy from the resonance and $2\hbar\Omega_{g3}$ the anti-crossing gap between state g and 3 as represented in the [Figure 1-10](#). n_s represents the sheet carrier density, τ_3 the upper state lifetime and $\tau_{\perp}$ the in-phase dephasing time which is responsible for the loss of phase between the two states (about 0.1 ps).

Figure 1-10: Detuning resonant tunnelling of two quantum wells coupled via an injector barrier

For a specific bias voltage, the energy states g and 3 are aligned, the detuning energy between the two states is equal to zero. The maximal current density is reached when the energy states are in resonance:

$$J_{max} = qn_s \frac{2|\Omega_{g3}|^2 \tau_{\perp}}{1 + 4|\Omega_{g3}|^2 \tau_{\perp} \tau_3} \quad (1-13)$$

From there, two cases can be identified:

- Weak coupling regime: $4|\Omega_{g3}|^2 \tau_{\perp} \tau_3 \ll 1$

$$J_{max} = 2qn_s |\Omega_{g3}|^2 \tau_{\perp} \quad (1-14)$$

In this regime, the transport of electrons is dominated by scattering. τ_3 becomes faster than the injection of electrons from the injector. Therefore, the population inversion is highly deteriorated, and the population of electrons is located in the state g , i.e. the ground state of the injector.

- Strong coupling regime: $4|\Omega_{g3}|^2 \tau_{\perp} \tau_3 \gg 1$

$$J_{max} = \frac{qn_s}{2\tau_3} \quad (1-15)$$

In this regime, the current density is controlled by the upper laser state. The injector provides enough electrons to ensure a good population inversion between the two laser states.

1.3.6. Optimization of the QCL structure

As discussed in Chapter 1, the advanced structures for excellent performance in QCLs were found in the 20th. The population inversion mainly determines the performances of the laser. Therefore, the implementations of a better electrons extraction of the lower state were the key to increase the efficiency of the QCLs. So far, the two optimum design are based on “two-phonon resonance” and “bound-to-continuum” [41], [42]. These two approaches are innovative in the way of electrons extraction of the lower laser energy level:

- Two-phonon resonance as shown in Figure 1-11 (a): The energy gap between the two lower laser energy levels (2-3) are in resonance with the LO phonons of the lattice ($E_{phonon,GaAs} = 36 \text{ meV}$). By enhancing the interaction electron-phonon, it highly reduced the electron lifetime from the level 2.
- Bound-to-continuum as displayed in Figure 1-11 (b): the design of the structure allows a coupling between the low laser energy level (2) and the lower energy levels (3-4...) which create a sort of miniband which permits fast intraband scattering.

Figure 1-11: (a) Two-phonon resonance design; the energy gap between 2 and 1 is in resonance with the phonon energy of the lattice, (b) Bound-to-continuum design; lower laser energy level forming a miniband (from [56]).

The state-of-the-art regarding the efficiency and temperature of operation is mainly following these extractions methods.

1.3.7. Design based on InGaAs/InAlAs on InP substrate

As discussed earlier, the choice of the materials is crucial for the performances and the wavelength emission of the QCLs. The alloy of InGaAs/InAlAs structure is one of the more mature compositions for MWIR QCLs. The matched InGaAs/InAlAs structure allows a band offset of 0.5 eV, while the strain-balanced structure offers a gap of 0.75 eV and improve the confinement of the carriers in the active region. The relatively low effective masses of the materials lead to a significant matrix element, $z_{ij}^2 \propto \frac{1}{m^*}$, and longer non-radiative relaxation lifetime, $\tau \propto \frac{1}{\sqrt{m^*}}$. It consequently, increase the differential gain due to its proportionality to both previous parameters. Therefore, this strained composition has been used to demonstrate very performant lasers [45], [57].

1.4. Quantum cascade laser frequency combs

Frequency comb sources have been actively studied over the last decade; they are typically generated from active or passive mode-locking lasers. The visible and ultraviolet spectra have been covered by conventional lasers [58], [59] but for the MIR region, optical combs operations are usually obtained by down conversion of NIR traditional lasers through nonlinear crystals which renders the system bulky and complicated [60]. Compact MIR frequency combs have been only recently demonstrated [118], [119]. Owing to their compactness, electrical injection characteristic and high-performance in the MIR region, quantum cascade lasers have become the optoelectronic device of choice to fill this electromagnetic spectrum gap. This region is an essential spectral range where; many molecules have their first fundamental vibrational resonances, and where two atmospheric transmission windows are contained [60].

In essence, optical frequency comb is a coherent optical beam composed of particularly narrow and equidistant spectral lines in the frequency domain. It can act as an optical ruler where its teeth can measure unknown frequencies emitted by many sources such as lasers, atoms, molecules, stars or other objects. Thanks to its equidistant mode, combs laser exhibit higher precision than any other tools by measuring the beating signal between the incoming sources and the modes of the laser cavity. *Figure 1-12* illustrates the set of equidistant frequency separated by the comb repetition rate f_{rep} . All the optical modes have a stable phase relationship between each other which allow, depending on the relation, Frequency Modulated (FM) or Amplitude Modulated (AM) output such as extremely short optical pulse largely used.

Figure 1-12: Principle of frequency comb in time and frequency domain, where the phase relationship lead to pulse formation (from: [32], [63]).

A large number of applications are expected including in free-space communication and spectroscopy where comb lasers exhibit outstanding results. It has dramatically simplified

and improved the accuracy of many uses applications such as spectroscopy [64], [65], [66], optical metrology [67], GPS technology and communications [68]. Moreover, they enable a direct link between radio frequency and optical frequency, thus allowing the realisation of ultra-precise optical atomic clocks [69] and ultra-high-rate communication [13]. The interference of the cavity modes with each other in a mixer generates signals in RF domains called beating signal and its harmonics. This RF signal gives useful information on the quality of the comb lasers and contains information on the phase distribution of the modes involving in the comb regime. This RF signal corresponds to the envelope of the output signal.

1.4.1. Mode-locking in QCLs

The term “mode-locking” is most often used in the narrow sense of ultra-short pulse laser, but the general expression only implies that the phases of all modes are locked in a stable relationship between each other. This link can be different and far richer than a constant or linear evolution of the phase over the spectrum and thus can lead to something else than a pulse generation [61] [70]. Indeed, for conventional lasers, i.e. when the gain recovery time is longer than the photon round-trip time in the cavity, the repartition of the amplitudes and the phases’ relation over the spectrum leads to ultra-short optical pulses which can go up to sub-femtosecond pulses for broadband lasers [71]. QCLs are classified in class A lasers, due to their intersubband transition, their gain recovery time (sub-picoseconds range ≈ 0.5 ps) of such lasers is much shorter than the roundtrip cavity (tens of picosecond ≈ 63 ps for 3 mm laser length). Consequently, it leads to a very different output signal in term of instantaneous power and frequency. Indeed, the active region seems not able to store the energy to promote the formation of a pulse in the cavity, the

distribution of amplitudes and phases' relation over the spectrum is a stable and non-trivial function which conducts to a frequency modulated output. We can notice that the very short upper state lifetime of QCLs is responsible for its very wide-band response in frequency domains but also for its incapacity of forming passive ultra-short pulse operation. Relevant experimental and simulation works have been done by J. Faist et al. which demonstrate a good agreement between the mode-locking simulation and experiment on QCL laser [32], [72], [73]. Considering typical time constants in intersubband devices and the effect of the different dispersive sources such as the gain curve and four-wave-mixing process on QCLs, they simulated the behaviour of the amplitude, frequency and phase of the cavity modes for low dispersive regimes (about $500 \text{ fs}^2/\text{mm}$). *Figure 1-13* exhibits the mains figures of the simulations, explaining and predicting the results of QCL in MIR and THz region. Row (1) and (2) display the simulation of passive mode-locked MIR QCL where the upper state lifetime is much shorter than the round-trip cavity, containing respectively 1 and 2 active regions with different central frequencies. Row (3) simulate an active mode-locked THz QCLs where, due to the extremely low energy transition, exhibit upper state lifetime much higher and comparable to the round-trip cavity. These simulations are supported by substantial evidence in experiments data [61], [73], [74]. However, using a specific diagonal transition leading to a much longer upper state lifetime of approximately 50 ps and an actively modulating short section of the device, pulse operation in MIR QCLs has been archived [75].

Figure 1-13: Summary of mode-locked QCL. The rows exhibit in order; the phase value and amplitude of the cavity modes, the RF spectral power of the beating signals with its harmonics and the instantaneous output power. The two first rows are the simulation for passive mode-locked MIR QCLs with respectively 1 and 2 active regions. The instantaneous frequency in (2-c) and the beating signal in the inset (1-c) corresponding to this configuration are added. The last row is the simulation for active mode-locked THz QCL with long upper state lifetime and active RF-injection (extract from [32], [72]).

Column (a) shows the amplitude distribution of the different cavity modes, column (b) the power of the spectral components in RF domain, and column (c) the consequences on the output power in term of instantaneous power or frequency.

This simulation supported by experimental data, draw the non-conventional behaviour of mode-locking in QCLs. In contrast with solid lasers, in the case of intersubband

transitions where the upper state lifetime is much shorter than the round-trip, the amplitude distribution of the cavity modes and the non-trivial phase distribution lead to an FM-modulation output which displays almost constant instantaneous output power and the instantaneous frequency which is periodic at the round-trip frequency. All the harmonics of the beating signal in the radio frequency domains are about 3 orders of magnitude lower than the DC value which is significantly different from pulse operation mode-locked. The inset of (1-c) in *Figure 1-13* shows extremely low and thin amplitude of the beatnote of such configuration extracted from [61]. To finish, in the case of no mode-locked laser, the distribution of phase is random and not stable over the spectrum leading to chaotic instantaneous output power.

1.4.2. Discussion on FM-modulation

The spread of a 4 ps pulse represents about 400 μm in QCL cavities. This value is not negligible compared to the length of a 2 or 3 mm cavity. Longer pulses or multi-pulses can be stable in a QCL cavity which would lead to non-trivial stability somewhere between AM and FM modulations.

The research on this topic is very young, and one needs to keep a critical eye on it. Further, investigations are required to fully understand and validate the complex mechanism of ISBT applied to laser or photodetectors.

1.4.3. Nonlinearity in QCLs

The understanding of nonlinear processes in the QCLs is essential for a better comprehension of the mechanism responsible for the comb formation in intersubband transitions. In this paragraph, we will introduce the nonlinearities in quantum confined

systems and more precisely, the third order nonlinearity corresponding to the Four-Wave Mixing (FWM) process in QCLs which is responsible for the comb operation.

The invention of the laser has been necessary to study the nonlinear effect which appears when the optical properties of materials are modified due to the presence of high electric field radiations [76]. We describe the nonlinear response of the dielectric polarisation due to the electric field of the light by:

$$P(x, t) = \epsilon_0(\chi^{(1)}E(x, t) + \chi^{(2)}E^2(x, t) + \chi^{(3)}E^3(x, t) + \dots) \quad (1-16)$$

Where $\chi^{(i)}$ represents the i orders susceptibilities depending on the material, and E is the incident electric field. The first susceptibility describes the linear response, while other orders describe the nonlinear effects of the medium.

One of the particularities of the intersubband transitions is the large values of the susceptibilities compare to any other materials which allow significant second and third order nonlinear effects. This enhancement is mainly due to the strong dipole matrix elements $z_{i,j}$ in the order of magnitude of few nm which are about one or two orders of magnitude higher than for bulk materials and, the abilities of engineering the energy level. Using these properties, giant second and third order susceptibilities have been demonstrated in intersubband transitions [77], [78], [79].

Due to the significant optical matrix element between the upper and the lower state of the laser transition, QCLs exhibit strong resonant nonlinearities. Substantial values of second and third order nonlinearities have been demonstrated [61], [80]–[83]. For instance, FWM process in a QCLs done by two external excitations with a detuning of about 210 GHz show a significant value of $0.9 * 10^{-15} \text{ m}^2\text{V}^{-2} \text{ per period}$ [83]. FWM can be self-

induced in QCL. The laser transitions can be approximated as an ideal resonance with the incident electric field; therefore, the third order susceptibility per period at the resonance can be written as:

$$\chi^{(3)} = \frac{4\Delta N q^4 z^4 \tau_1 \tau_2^2}{3\varepsilon_0 \hbar^2} \quad (1-17)$$

With ΔN the volume population inversion, τ_1 the relaxation time, τ_2 the dephasing time, ε_0 the vacuum permittivity, $\hbar$ the Planck constant, q the electron charge, and z the matrix element corresponding to the laser transition. For a typical value of laser transition in microstrip QCL ($\Delta N = 1.9 * 10^{16} \text{ cm}^{-3}$, $\tau_1 = 0.29 \text{ ps}$, $\tau_2 = 0.15 \text{ ps}$) third order susceptibility can exhibit a very strong value of $8 * 10^{-14} \text{ m}^2 \text{ V}^{-2}$ per period [82].

Furthermore, the remarkable feature of the third order nonlinearity is the powerful mechanism of the four-wave mixing to generate stable frequency comb in the mid-infrared region due to the self-mode-locking process more detail in the next paragraph [12], [83], [15], [16]. Besides, FWM process had an extremely broad bandwidth due to the short upper state lifetime of intersubband transitions.

Nonlinear processes are also dependent on the dispersion as we will describe in the next paragraphs, the FWM can appear only in the case of the respect of the phase-matching condition (Δk). For the red resonance shown in [Figure 1-14](#) the phase matching condition may be express as:

$$\Delta k = \frac{n_3 \omega_3}{c} + \frac{n_4 \omega_4}{c} - \frac{n_2 \omega_2}{c} - \frac{n_5 \omega_5}{c} \approx 0 \quad (1-18)$$

With n_i the effective refractive index of the correspondent modes and ω_i the optical frequencies. The phase condition depends on the effective refractive index for the

different optical modes. Therefore, this mechanism is strongly linked to the dispersion in our QCL as described in **1.4.7. Dispersion in QCL**. As we will explain in the next paragraphs, regarding our QCLs, the low dispersion of these different sources facilitates the respect of the phase-matching condition and thus, enhance the FWM process in the low-dispersion area.

1.4.4. Four-wave-mixing process

The FWM effect occurs when three different frequencies propagate together in a nonlinear medium. The interaction between these wavelengths will produce a 4th wavelength such as:

$$P_{NL} = \epsilon_0 \chi^{(3)} E_i E_j E_k \quad (1-19)$$

The wavelength generated will follow:

$$\omega_{ijk} = \omega_i \pm \omega_j \pm \omega_k \quad (1-20)$$

Figure 1-14: (a) Non-degenerate and degenerate generation of frequency by FWM on the laser spectrum, (b) zoom-in on dispersed Fabry-Perot modes with injection-locking in yellow and pulling-effect in purple, (c) diagram showing the resonant FWM process in the laser transition and (d) injection-locking process at the round trip cavity corresponding to the free-spectral range.

As described in [Figure 1-14](#) (a) and (c), three incident electric fields can create, through the FWM a large number of new or already existing frequencies through different processes;

- Non-Degenerate generation process $\omega_{ijk} = \omega_i + \omega_j - \omega_k$
- Degenerate generation process $\omega_{jk} = 2 * \omega_j - \omega_k$
- Self-generation process $\omega_i = 2 * \omega_i - \omega_i$

Due to the broadband process the new frequencies generated in the gain curve, can be amplified by the gain medium, enhance existing modes and contribute to a generation of a new frequency through the same process.

Theoretically, the frequency generated by the FWM process at ω_i has no reason to be at the exact same frequency than the Fabry-Perot modes at ω_i' which see the dispersion of the medium. If the dispersion is low enough, i.e. the frequency generated is close enough to the Fabry-Perot modes, the frequency generation can act as a master oscillator and lock the frequency of the cavity modes corresponding to the slave oscillator such as $\omega_i = \omega_i'$. This process is illustrated in [Figure 1-14](#), the critical aspect is; locking the Fabry-Perot frequency on the generated frequency, goes along with fixing the spacing between the modes to the spacing submitted by the FWM process ($\Delta\omega_{FWM}$) or the RF injection in the locking range. Therefore, thanks to the FWM mechanism in a low-dispersion region, this nonlinear effect will not only generate new frequency but also create a comb regime with a fixed spacing between modes such as $\Delta\omega_{i,i+1} = \Delta\omega_{FWM}$.

As shown in [Figure 1-14](#) (d), this process can be seen in the radio-frequency range corresponding to a frequency of the mode spacing. The Free-Spectral-Range (FSR) of modes locked by the FWM becomes equal to the mode spacing submitted by the nonlinear process. In this sense, in a low-dispersion regime, the FWM mechanism will correct the dispersion which tends to disperse the FSR of the cavity modes.

1.4.5. FWM as an injection-locking mechanism

The phenomenon of injection locking is well described by Adler's equation on coupled oscillators [84]. Indeed, this phenomenon can be related to the locking frequency between

a master oscillator (here the frequency generated by FWM for the optical modes) and the slave oscillator (here the Fabry-Perot cavity modes).

As illustrated in Figure 1-14 (b), to lock the cavity modes, the frequencies generated have to be close enough to the cavity modes. We define the locking range (red domains in Figure 1-14 and Figure 1-15) as the frequency range where the Fabry-Perot modes (slave oscillator) can be locked by FWM generation (master oscillator) such as:

$$\Delta\omega_{lock} = \frac{2\omega_0}{Q} \sqrt{\frac{I_1}{I_0}} \quad (1-21)$$

Where ω_0 and I_0 represent respectively, the frequency and the intensity of the Fabry-Perot modes, I_1 the intensity of the FWM generation and Q the quality factor of the slave laser resonator.

The pulling effect illustrated in Figure 1-14 (b) and Figure 1-15, is explained for a frequency outside of the locking range ($|\omega_1 - \omega_0| > \frac{\Delta\omega_{lock}}{2}$) we can express the pulling effect as:

$$\omega_0 - \omega_{osc} = \frac{\omega_0^2 I_1}{Q^2 I_0} \frac{1}{2(\omega_0 - \omega_1)} \quad (1-22)$$

Where ω_{osc} is the frequency of the slave oscillator pulled by the injection. We can also notice the creation of additional weak sidebands at $\omega_{osc} \mp \Delta\omega_b$ with $\Delta\omega_b =$

$$\sqrt{(\omega_1 - \omega_0)^2 - \left(\frac{\Delta\omega_{lock}}{2}\right)^2} \approx \omega_1 - \omega_0 - \frac{\left(\frac{\Delta\omega_{lock}}{2}\right)^2}{\omega_1 - \omega_0}.$$

Figure 1-15: Illustration of the pulling effect due to the injection out of the locking range with the creation of sideband frequency.

This is how FWM can contribute to the formation of frequency comb operation in QCLs with low dispersion. However, this phenomenon is the same mechanism governing the injection locking by microwave modulation in the radio-frequency domains. Indeed, in this case, the slave oscillator is the beating signal of the Fabry-Perot modes and the master oscillator is the RF injection thanks to the RF generator. These phenomena will be experimentally observed in Chapter 2. The injection in the locking range will generate sidebands in the optical spectra and tune the FSR of the laser while for an injection out of the locking range only a small part of the spectrum will be locked, and it may potentially introduce dispersion in the optical spectra.

1.4.6. Instabilities in QCLs

Due to fast gain recovery time, the multimode dynamics in QCLs is different from other common lasers. The instabilities into the medium transform its emission from a single mode to a multimode operation. Fast gain recovery of QCLs promotes two multimode

regimes: One is related to the Risken-Nummedal-Graham-Haken (RNGH) instability and the second is the spatial hole burning. This phenomenon is due to the oscillation of the population inversion at the Rabi frequency which modulates the gain. However, this multimode operation is essential for the comb formation.

The narrow width of the ridges, as well as the high temperature and high intensity in the cavity, the multimode behaviour tends to be governed and favoured by the RNGH instability shown by the Rabi splitting observable in many spectra (including our spectrum). The spectral hole burning also plays an important role (especially at low temperature) in the amplitude distribution of the modes over the spectrum, it tends to suppress the central peak and replace it by a multitude of lower intensity peaks. The studies also show that as the active region width is increased, the RNGH instability and spatial hole burning tend to be attenuated [85] [86]. It seems that the mode-locking, the instabilities, the nonlinear effects are linked by a non-trivial mechanism.

1.4.7. Dispersion in QCL

It is critical to have a good understanding of the different sources of dispersions in QCLs. The dispersion is the variation of the refractive index of a material with the frequency, all the cavity modes of the lasers do not see the same refractive index, and therefore, each longitudinal mode can have a different propagation speed in the medium. It introduces a shift in the frequencies and phases in the Fraby-Perot modes which is the central problem in frequency comb formation. We study the dispersion by analysing the FSR over the spectrum; if there is no dispersion, the cavity modes will all have the same value of the mode spacing and thus will be perfectly equidistant (constant group refractive index over the spectrum).

However, if the medium is dispersive, the cavity modes will be slightly shifted compared to their normal position. That is why, in the frequency domain the optical modes are governed by the dispersion and are not equidistant anymore whereas, in a temporal domain, this phenomenon leads to a shift of the phase of each mode and prevent them from stable interferences which could lead to a short pulse or other stable output. The group refractive index is written as:

$$n_g = n + \omega \left(\frac{dn}{d\omega} \right) \quad (1-23)$$

With n is the phase refractive index. The Group Velocity Dispersion (GVD), generally displayed in fs^2/mm , which is the variation of the group refractive index in the frequency domain is defined as:

$$GVD = \frac{1}{c} * \frac{dn_g}{d\omega} \quad (1-24)$$

The GVD in a QCL is composed of three sources of dispersion, described as:

- The dispersion of the materials is the variation of the refractive index as a function of the wavelength of each element composing the waveguide. We must weigh the dispersion introduced by each layer by the confinement factor of the transverse mode propagating in the waveguide.
- The waveguide GVD or the modal dispersion comes from the fact that the transverse modes are different for each wavelength. That introduces a variation on the effective refractive index for each transverse mode.
- The gain and loss dispersion is often the most critical dispersion for devices without specific design, based on many active regions, to achieve a flat gain curve.

The gain curve is linked to the imaginary part of the susceptibility by the Kramers-Kronig relations.

An essential study of the sources of dispersion in QCLs has been realised by Faist's group [73]. They showed that the dispersion introduced by the InP and other materials constituting the active region as GaAs is very low for the mid-infrared range (from 3.3-10 μm) and exhibit a value which can be well below 1000 fs^2/mm . The dispersion introduced by the waveguide is highly dependent on the width of the ridges where a trade-off must be found between the high-dispersion and the number of transverse modes oscillating in the FP cavity. Therefore, for an "empty" waveguide, i.e. considering the laser cavity empty of photons, an active region wider than 8 μm would introduce a negligible waveguide dispersion for our lasers. In such cases, the main source of dispersion becomes the gain spectrum, especially if no specific design to flatten the gain curve is done (single active region). Using the Kramers-Krönig relations, a single active region with a modal gain of about 2 cm^{-1} exhibits a dispersion oscillating of about $\pm 300 \text{ fs}^2/\text{mm}$.

Nevertheless, the "empty" waveguide does not consider the self-consistent control of the gain curve in QCL, the gain saturation under lasing operation can be calculated using Maxwell-Bloch model, and this mechanism can introduce strong dispersion under the resonant laser action. That is also why the observation of a thin beating signal in QCL is not always stable all over the I-V.

A necessity for frequency comb generation is a low dispersion medium. The remarkable feature is that zero-dispersion is not always required as long as the nonlinear effects are strong enough to compensate it. Indeed, the dispersion tends to pull the frequencies away

from their equidistant values, and in the frequency domain, nonlinear effects such as FWM manages to lock the frequencies and phases of all modes and thus mitigates the impact of dispersion [70].

Figure 1-16: (a) Diagram showing the impact of dispersion on the free-running optical modes and (b) simulation of the frequency offset for three values of dispersion considering FWM (from [72]).

Considering the different parameters of the laser transitions and the cavity, a rough idea of the correction of the dispersion introduced by the FWM process can be given. J. Faist et al. calculated that the GVD must remain below $560\text{ fs}^2/\text{mm}$ for the modes to stay efficiently coupled by FWM in their experiment. This value is a rough estimation of the order of magnitude of the dispersion that is relevant for comb operation in QCL [73], supported by the simulation shown in *Figure 1-16* (b) and (c) [72]. For a GVD of $500\text{ fs}^2/\text{mm}$, the FWM will be able to lock the mode and generate a frequency comb laser. For MWIR QCLs, the dispersion introduced by the materials and the waveguides remain lower than the one from the gain medium. For a single active region, only the modes around the maximum of the gain curve (can be shifted by the other sources of dispersion) will exhibit a low dispersion and be locked thanks to the FWM.

1.5. Quantum well Infrared photodetectors

1.5.1. History

As we discussed earlier, the history of infrared photodetector started 200 years ago with a report in the Royal Society as Dark heat [1], and since this time research never stopped to bring brilliant innovation and discovery in this field [3]. In the next 100 years after the first observation of infrared phenomena, Smith discovered the first photoconductive effect due to a photons flux in selenium [87], followed by the invention of the first bolometer in 1880 by Langley. At this time, the thermal detector such as thermocouples and bolometers were highly used and were the most performant detector [3]. Lately, due to the potential applications of IR photodetectors in the army, the world war II has been a springboard for the development of infrared photodetector where many materials have been studied [88]. The first infrared detector, based on Lead Sulphide (PbS) quantum detectors have been invented in Germany by Gudden, Görlich and Kutscher for military applications and followed by the first IR image converter in 1934. Both discoveries were significantly improved during the world war II for ships and tanks imaging [3]. Since this invention, two main categories of IR detectors can be recognised and are listed below.

- Thermal detectors: These detectors respond to an incident IR light by a change of their physical properties. Golay cells based on mechanical deformation due to pneumatic effect [89], pyroelectric detectors based on the generation of temporary voltage in a pyroelectric crystal due to a change of temperature [90] and bolometers based on the variation of the electrical conductivity of the semiconductor [91].

- Photovoltaic and photoconductive detectors: These detectors in the IR are based on photoelectrical effects introduced by Einstein in 1905 [92], [93]. As described earlier they can be separated into two semiconductor devices:
 1. Interband detectors, with many important innovations in the first part of the 20th century as HgCdTe [94], II-VI and III-V photodiodes [95], [96], [97], [98], and recently nanotube [99].
 2. Intersubband detectors, which were possible thanks to the improvement of the growing facilities as MBE and MOCVD in the second part of 20th century. The central devices based on ISB transitions chronologically listed are Quantum well infrared photodetectors [21], quantum cascade detectors [100], charge sensitive infrared detectors [101] and quantum dot infrared photodetectors [102].

Figure 1-17: History of the development of infrared photoconductive detectors and focal plane array technology (extract from [3]).

In this paragraph, we will focus on photovoltaic and photoconductive detectors which are more relevant to our work. [Figure 1-17](#) shows the history of the development of IR detector. The first demonstration of photoconductive sensor is followed by indium antimonide (InSb) detectors at 3-5 μm . The year 1959 made a turn for IR detector with the discovery of the Mercury-Tellurium-Cadmium detectors (MCT also called HgTeCd) by Lawson and co-workers [103], particularly attractive due to their high-sensitivity and the finding of variable band-gap HgCdTe alloy permitting a considerable degree of freedom in a wide range of wavelength. These systems are currently used mainly for thermal imaging covering from 2 to 15 μm . As described earlier, the large enhancement of growing technics in the second half of the 20th century (thanks to MBE and MOCVD) led to the development of devices based on intersubband transitions. These new technologies require a precision of the atomic layer such as the first superlattice QWIP based on GaAs/GaAlAs by B. Levine in 1987 [21]. The unique properties of ISB

transition already discussed in the previous paragraph make them one of the most reliable alternatives to MCT technologies. The intense interest on this technology allows quick development and the years following their discovery; numerous enhancements have been realised, especially in the absorption wavelength which, described in [Figure 1-5](#), allow to tune the wavelength by either using different semiconductor or changing the Aluminium fraction in the composition. Therefore, QWIPs covering the first atmospheric transmission windows (3-5 μm) are typically composed of InGaAs/AlGaAs while for the second atmospheric transmission windows (8-12 μm), they are generally formed of GaAs/AlGaAs, both using different aluminium concentration for relatively smaller tune of the wavelength [28], [38], [104]. Due to their properties, they can reach very a low energy transition and are good candidates for THz detection with the first demonstration by H.C. Liu et al. in 2005 using very low aluminium fraction on GaAs/AlGaAs quantum wells [105]. The main drawback of QWIPs is their dark current which increases exponentially with the temperature and therefore degrades the signal to noise ratio at high temperature, to eliminate or reduce the dark current they need to be cooled to cryogenic temperature. QWIP growth on GaAs is now a mature technology [106], the development and the commercialisation of Focal Plane Arrays (FPA) for thermal imaging systems is a good judge for the maturity of a system. Several important companies are developing these products for the two transmission windows such as Jet Propulsion Laboratory for the United-States, Sofradir for France and IR-nova for Sweden.

1.5.2. State-of-the-art

This paragraph will give a state-of-the-art concerning the principal figure of merits for IR detectors. The detectivity is one of them which take into account several critical

parameters including the signal to noise ratio and the responsivity. Figure 1-18 summarised the background-limited detectivity of various detectors including the theoretical curves of ideal thermal, photoconductive and photovoltaic detectors using the radiation of a blackbody at 300 K.

Figure 1-18: Comparison of background-limited detectivity of variously available IR detectors with the temperature indicated ($FOV = 2\pi$ for 300 K background). Ideal thermal, photoconductive and photovoltaic detectors are also shown in dashed lines (extracted from [3]).

Thermal detectors mostly have a constant detectivity covering a vast domain in the infrared region, the uncooled bolometers and thermopiles have a detectivity of about 10^8 - $10^9 \text{ cm} \cdot \sqrt{\text{Hz}}/\text{W}$ while hot-electron bolometers at Helium temperature reach about 10^{10} - $10^{11} \text{ cm} \cdot \sqrt{\text{Hz}}/\text{W}$ up to THz domains. Golay cells along with the pyroelectric detectors

can have a detectivity at room temperature of 10^9 - $10^{10} \text{ cm} \cdot \sqrt{\text{Hz}}/\text{W}$. Nevertheless, due to their physical properties, these IR detectors do not allow high-speed behaviour, which for the best case reach few kHz of modulation. The interband detectors listed beyond display relatively high-performance with detectivities of 10^{10} to $10^{12} \text{ cm} \cdot \sqrt{\text{Hz}}/\text{W}$ up to high temperatures but show some strong limitation regarding the spatial uniformity and significant 1/f noise [107]. Another considerable barrier of interband detectors is their intrinsic low-speed response which makes them not suitable for all applications based on high-speed detections. Indeed, the intrinsic high-speed response of a detector is limited by the smaller value of the photoelectron lifetime or the transit time. For HgCdTe it is in the order of magnitude of microseconds, InAs/GaInSb superlattice structure, a serious alternative to HgCdTe [3], [73], shows a photoelectron lifetime of about 100 ns.

Due to their intrinsic properties, QWIP shows a photoconductive gain higher than 1 which provides competitive parameters regarding the responsivity and detectivity of 10^{10} - $10^{11} \text{ cm} \cdot \sqrt{\text{Hz}}/\text{W}$ for a typical GaAs/AlGaAs superlattice and outstanding parameters regarding the high-speed behaviour due to its very short intrinsic carrier lifetime of about 5 ps. The carrier lifetime of QWIP is more than 5 orders of magnitude lower compared to the other alternative [28]. QWIP hold the unique position of high-speed behaviour and high-absorption for the thermal infrared region without any competitive options. Since 1995 a significant amount of efforts have been invested in the LWIR, where high-room temperature heterodyne detection up to 110 GHz [29] has been demonstrated and recently, heterodyne measurement with high-sensitivity at room temperature operation using metamaterial [109]. Meanwhile, in the MWIR region, peak-responsivity of 1 A/W for QWIP stack [110] and high background-limited detectivity of $2 \times 10^{11} \text{ cm} \cdot \sqrt{\text{Hz}}/\text{W}$ at 77K [111] were achieved. But so far, no MIR devices could gather high sensitivity,

high responsivity, high-speed behaviour and high-temperature response in one [112], [113].

The two primary intersubband detectors are QWIPs and Quantum Cascade Detectors (QCD). QCD works on the same principle that the QCL devices, the excited electrons of the QW absorber tunnel through thin barriers equivalent to the injector for QCL and decays by LO phonon emission on the consecutive wells down to the second QW absorber and so on. Due to its high photoconductive gain, QWIPs achieve better performance [114], but its behaviour of photoconductor introduces a sizeable dark current which in general constrains them to low-temperature use. However, QCDs are photovoltaic devices which limit the dark current and permit higher temperature use [115], [116]. Detectivity of 10^8 and $10^9 \text{ cm} \cdot \sqrt{\text{Hz}}/\text{W}$ has been demonstrated for respective devices at $9 \text{ } \mu\text{m}$ at 70 K and $5.3 \text{ } \mu\text{m}$ at 150 K [116]. However, QCDs showed high-speed measurements with a cut-off frequency of few GHz for heterodyne detection [117] and rectification method [118] which not meet the performances of the QWIPs.

1.5.3. Physics of Quantum well infrared photodetectors

Principle

Even though the physics is relatively similar, QWIP call for a much straightforward design than QCL and QCD due to the absence of injector. QWIP is a photoconductive unipolar device using intersubband transitions to absorb a particular wavelength according to the energy level in the structure. Figure 1-19 shown an illustration of the principle of the QWIP, the incident photon at specific energy is absorbed by an electron in the lower state of the quantum well and send to the continuum where it is swept by the

electric field toward the collector. It is usually composed of a stack of n-type III-V materials which form quantum wells sandwiched between two highly n-doped GaAs contact layers also called emitter and collector. The quantum well structure is constituted by InGaAs/AlGaAs for absorption in the first transmission windows in the MWIR and GaAs/AlGaAs for the second atmospheric windows in the LWIR. To have a sufficient absorption between 10 and 100 quantum wells are cascaded. One of the crucial parameters is the doping concentration in the active quantum well, used to populate the lower energy state. If the doping is too important, it will lead to an unwanted high dark current due to the lower thermal activation energy required to send the electron to the continuum.

Figure 1-19: Illustration of QWIP principle (reprinted from [28]).

We will now present the theory of the Quantum well infrared photodetectors. Several textbooks describing more in details the intersubband and quantum well physics, with few specialised in QWIP have been written and were used to write this paragraph [38], [119]–[121].

1.5.4. Transport model: Dark current

Dark current is the weightiest factor in the QWIP, it directly related to the signal to noise ratio and the temperature of operation of the QWIP. Several relatively sophisticated models have been established to formulate relevant law in the physics of QWIPs, but none can express the entire complexity of the principle. Indeed, QWIP principle is non-trivial due to its combination between drift-diffusion and ballistic transport mechanism in the QWs and the continuum, added to that the high doping and highly non-uniform distribution of the electric field over the structure. Here we will focus on the model which are the most relevant for our analyse:

- 3D carrier drift model to estimate the 3D electron density in the continuum considering the doping level of the QW, i.e. transport on the top of the barrier.
- Emission-capture model to build up the main equations governing the QWIP under illumination, i.e. the transport in the quantum well.
- Self-consistent drift-diffusion model will be briefly introduced at the end of this section. This model is essential to understand the limitations of the two previous models.

QWIP is photoconductive devices; a non-null bias voltage applied between the contact layers is required to make the carrier flowing through the structure.

Vertical current flow: Under dark condition or illumination, the drift carrier model defines the vertical current of the electrons flowing in the continuum from the two contact layers.

The dark current density is given by:

$$J_{dark} = eN_{3D}v(F) \quad (1-25)$$

With N_{3D} the 3D electron density on the top of the barrier and $v(F)$ the drift velocity depending on the electric field F where $v(F) = \frac{L_p}{\tau_{trans}(F)}$. L_p corresponds to the length of the period composed of the barrier and the quantum well and τ_{trans} the transit time through one period which is around 0.43 ps using typical values of $v(F) = 10^7 \text{ cm.s}^{-1}$ and $L_p = 43.5 \text{ nm}$.

Regarding the electron density inside the quantum well, two phenomena need to be considered: emission and capture phenomena and the tunnelling effect of the electrons through the barriers. At first, we will simplify the calculation using only the emission and capture model described by H.C. Liu in [114] which requires some reasonable assumptions in a first approach and for low bias voltage. Then we will explain the tunnelling effect which can occur through the barrier that we neglect in the first approach.

1. The barriers are thick enough to neglect the interwell tunnelling effect in the dark current.
2. The electron density in each well remains constant.
3. The emitter and collector are supposed to be perfect injection contact (due to their high doping level).
4. Only one bound state is confined in the quantum well; the upper state is in resonance with continuums (bound-to-continuum or bound-to-quasicontinuum).

The first assumption will be discussed more precisely in the next paragraph while the two following hypotheses will be more detailed in Chapter 3.

Figure 1-20 illustrates the total current through a period under illumination or in dark condition. When applying a bias voltage to the structure under illumination or not, we observe a dark current which is strongly related to the temperature. This phenomenon is due to the high doping in the quantum well, the electrons are not only distributed in the quantum well but also out of the well and on the top of the barrier as shown Figure 1-20 (a). Higher is the temperature, and more delocalized are the electrons to higher energy and therefore, are more likely to contribute to the dark current. This thermal emission is mainly caused by the interaction with the LO phonons of the lattice which create an inelastic deexcitation/excitation. It also tends to lower the electron density in the quantum well which must be balanced by the capture of electrons coming from the previous QW. Therefore, under the steady state, the density of electron captured is equal to the density of electron emitted ($j_c = j_e$). The dark current is constant over the structure, then we can write:

$$J_{3D} = j_c + (1 - p_c)J_{3D} \quad (1-26)$$

With j_{3D} the current flowing in the three dimensions on the top of the barriers (in dark condition $j_{3D} = J_{dark}$) and p_c the capture or trapping probability such as $j_c = p_c * j_{3D}$. Similarly, p_e is the emission probability with $j_e = p_e * j_{3D}$.

Figure 1-20: (a) Distribution of electrons in the quantum well, the broad arrows correspond to the delocalized electrons which contribute to the dark current. Representation of the current indicated by arrows for dark condition (b) and under illumination (c) (reprinted from [38]).

Figure 1-20 (b) and (c) represent the current flowing through the structure in dark condition or under illumination. Electrons can be excited by the thermal activation, i.e. contribute to the dark current as represented by the black arrow, or by photoexcitation. In the latest case, the carriers in the lower state absorb an incident photon with its energy matching the energy gap with the higher energy state represented by the red arrow. The vacant electrons in the quantum well will create an extra injection also translated by extra trapping represented by the black arrow for the thermal excitation and the thin black arrow for the photoexcitation.

Regarding the emission-capture model, Figure 1-21 introduces the different concept of time-probabilities of capture, scattering, transit, escape and relaxation. Figure 1-21 (a)

shows the scattering process from the ground state to the continuum ($1/\tau_{scat}$) and re-captured by the lower state ($1/\tau_{capt}$), the capture phenomenon can only occur to the vacant place in the lower band. V_b represents the barrier height, mainly the electrons with an energy higher than the barrier height will be scattered from the 2D ground state to the 3D continuum with a characteristic scattering time of ($1/\tau_{scat}$).

Figure 1-21: (a) Process of scattering and capture from the ground state into the continuum states and vice versa (extract from [38]). Illustration of the emission and capture model with lifetime involves of different states in dark condition (b) and under illumination (c).

The escape current density is proportional to the electron density in the 2D ground state N_{2D} with higher energy than the barrier height and the scattering constant time such as:

$$J_e = \frac{eN_{2D}}{\tau_{scatt}} \quad (1-27)$$

The capture probability will counterbalance the scattering process to fulfil the missing electrons in the quantum well and preserve the current. Therefore, the probability of capture is the capture rate divided by the sum of the possibilities which are the capture rate and the transit rate.

$$p_c = \frac{1/\tau_{capt}}{1/\tau_{capt} + 1/\tau_{trans}} = \frac{\tau_{trans}}{\tau_{trans} + \tau_{capt}} \quad (1-28)$$

With τ_{trans} is the transit rate for an electron to cross one period and arrive at the next quantum well. At dark current, we can assume that $p_c \ll 1$, in other words, $\tau_{capt} \gg \tau_{trans}$. Therefore, the dark current density using the equations (1-27) and (1-28) becomes:

$$J_{dark} = \frac{j_e}{p_c} = eN_{2D} \frac{\tau_{capt}}{\tau_{trans} * \tau_{scatt}} = eN_{2D} \frac{\tau_{capt}}{L_p \tau_{scatt}} v \quad (1-29)$$

p_c is directly inversely proportional to the photoconductive gain and has a typical value between 0.06 and 0.1. We can thus establish a relation between N_{2D} and N_{3D} such as:

$$\frac{N_{2D}}{\tau_{scatt}} = N_{3D} \frac{L_p}{\tau_{capt}} \quad (1-30)$$

A good estimation of N_{2D} will allow us to calculate the dark current density. Using the important results of Meshkov [122] which take into account the tunnel transmission probability, we can formulate an expression of the electron density in the 2D ground state of the quantum well under applied electric field:

$$N_{2D} = \int_{E_1}^{\infty} \frac{m^*}{\pi \hbar^2} T(E, F) \left[1 + \exp\left(\frac{E - E_F}{k_B T}\right) \right]^{-1} dE \quad (1-31)$$

$T(E, F)$ represents the transmission probability; this phenomenon will be more details in the next paragraph. To simplify the equation, we will neglect the electronic tunnelling contribution through the barrier or a part of it and use a pure thermionic emission regime, i.e. $T(E, F) = 0$ for $E < V_b$ and $T(E, F) = 1$ for $E > V_b$. Therefore, the expression of the charge carrier density and the dark current density using the equation (1-31) and the condition $(E - E_F)/k_B T \gg 1$ for $E > V_b$ become:

$$N_{2D} = \frac{m^*}{\pi \hbar^2} k_B T \ln \left[1 + \exp\left(\frac{E_F - E_1}{k_B T}\right) \right] \quad (1-32)$$

$$J_{dark} = \frac{e v \tau_{capt}}{\tau_{scatt}} \frac{m^*}{\pi \hbar^2 L_p} k_B T \exp\left(-E_{act}/k_B T\right) \quad (1-33)$$

With $E_{act} = V_b - E_F$ the activation energy of the structure. As expected, the dark current density increases exponentially with the temperature. The model described here has been compared with experimental values and showed a good agreement with the model [123].

Tunnelling effect

The electronic tunnelling in QW is an essential factor to consider. It is the primary factor involving in injector area for the QCL or QCD and must be taken into account for a better understanding of the behaviour of the structure of QWIPs. Wentzel-Kramers-Brillouin (WKB) approximation was developed in 1926 [124]–[126], this model allows us to write the transmission probability of an electron to tunnel through a barrier potential $V(z)$ with z the growth axis:

$$T(E) = \exp\left(-2 \int_0^z dz \sqrt{2m^*[V(z) - E]}/\hbar\right) \quad (1-34)$$

We define the transmission probability according to the barrier thickness, the energy of the electrons and the electric field F . Regarding the energy of the electrons; we can recognise different regimes exposed in [Figure 1-22](#). If the carrier energy is higher than the barrier height, it will be excited to the continuum, and therefore, the transmission is maximal. If its energy is included between $V_b - eF(z - L_w/2) < E < V_b$ we are in the triangular regime where the electric field will bend more or less the structure and reduce the effective barrier. In this case, depending on the different factors the transmission can be near the unity. However, for a trapezoidal regime, $E < V_b - eF(z - L_w/2)$, i.e. the electrons must go through the entire barrier, we can usually neglect this part since the barriers are thick enough to avoid this regime which could lead to dark tunnelling which would increase the noise of the detector.

The different regimes give:

- $T(E, F) = 1$ if $E > V_b$
- $T(E, F) = \exp\left(-2 \int_0^{z=z_c} dz \sqrt{2m^*[V_b - E]}/\hbar\right)$
if $V_b - eF(z - L_w/2) < E < V_b$ for **triangular regime**
- $T(E, F) = \exp\left(-2 \int_0^{z=z_B} dz \sqrt{2m^*[V_b - E - eF(z - L_w/2)]}/\hbar\right)$
if $E < V_b - eF(z - L_w/2)$ for **trapezoidal regime**

Figure 1-22: Schematic representation of the different tunnelling regimes occurring in QWIP's structure according to the WKB approximation.

Resonant tunnelling is an essential aspect used mainly in QCLs to design the injector area. Indeed, two barriers, placed in series may show, as for Fabry-Perot resonator in optics, a strong resonance which can give very high tunnelling transmission.

1.5.5. Transport model: Photocurrent

The photoconductive phenomena are not new and have already been explained for other devices. The QWIP is different due to its intersubband structure where the incident light is only absorbed between discrete energy levels in the conduction band of the quantum well. When the photons are absorbed by the QW, electrons from the ground state are photoexcited to the continuum as shown in [Figure 1-21 \(c\)](#). The photocurrent has a spectral dependence associated with the design of the intersubband transitions and therefore, the QW is not capable of absorbing photons with any energy. The photoconductive gain is defined as the number of electrons going through the structure

for each photon absorbed. It includes the electrons photoemitted but also the electrons added due to the extra-injection. This phenomenon is represented in [Figure 1-21](#) (c) and [Figure 1-23](#) for one quantum well. When an infrared source illuminates the structure a large number of electrons are added from the ground state to the current. An extra-injection current is needed to counterbalance the loss of electrons in the QW due to the photoexcited carriers. The amount of extra-current must be higher than the number of excited electrons to repopulate the wells due to the probability of capture ($p_c < 1$). For a cascade of QWs, the photocurrent is independent of the number of periods assuming that the photoexcited electrons are identical in each QWs. In other words, since the number of electrons of the extra-injection current captured in the well must counterbalance the photoexcited electrons, the extra injection flowing toward the first or last QW must be the same. The mechanism which causes the extra injection is a self-consistent process shown [Figure 1-23](#); The depopulation of electrons photoexcited in the QW enhances the electric field at the barrier until it satisfies the photocurrent requirement. Therefore, different potential drops are build up into the structure. The potential drop in the first barrier, V_1 , may be much higher than V_2 , the one after the QW, while with a homogenous repartition of charge we would have V_1 which would be equal to V_2 . This phenomenon is already largely described in the literature [38], [127], [128], [129], [130].

Figure 1-23: Illustration of the effect of the non-homogenous repartition of charges on the drop of potential in the structure under illumination. The arrows correspond to tunnelling effect to refill the QW. Blue arrow and solid line for the trapezoidal regime in dark condition and dashed arrow and solid line under illumination.

The extra-injected current is added to the photoemitted current and cannot be distinguished, it, therefore, contributes to the observed photocurrent. That is why and how the photocurrent gain can be higher than the unity. [Figure 1-21](#) explains the mechanism of photoexcitation, the escape rate ($1/\tau_{relax}$) corresponds to the characteristic time of an excited electron to remain in the excited state while the escape rate ($1/\tau_{esc}$) corresponds to the time characteristic for an excited electron to escapes to the continuum. The escape probability is the ratio between the escape rate and the sum of the relaxation and escape rate.

$$p_e = \frac{1/\tau_{esc}}{1/\tau_{esc} + 1/\tau_{relax}} = \frac{\tau_{relax}}{\tau_{esc} + \tau_{relax}} \quad (1-35)$$

At steady state, the variation of the density of excited electron n_{ex} in the upper state is equal to zero, the rate equation for the higher state gives us an expression of the incident flow absorbed as a function of the excited electron density in the upper state:

$$\frac{dn_{ex}}{dt} = \phi\eta^{(1)} - \frac{n_{ex}}{\tau_{esc}} - \frac{n_{ex}}{\tau_{relax}} = 0 \quad (1-36)$$

Therefore:

$$n_{ex} = \phi\eta^{(1)} \frac{\tau_{relax}\tau_{esc}}{\tau_{esc} + \tau_{relax}} \quad (1-37)$$

The expression of the photoemission current for one well is the excited electron density over the escape rate:

$$i_{photo}^{(1)} = \frac{en_{ex}}{\tau_{esc}} = e\phi\eta^{(1)} \frac{\tau_{relax}}{\tau_{esc} + \tau_{relax}} = e\phi\eta \frac{p_e}{N_{QW}} \quad (1-38)$$

With $\eta^{(1)} = \eta/N$ the absorption coefficient for one quantum well assuming that each quantum well has the same absorption coefficient and N_{QW} the number of quantum wells.

Since the extra-injection flowing toward the first and last QW is the same, the photocurrent is equal to the injection current which counterbalances the photoexcited electrons considering the capture probability. Consequently, we have:

$$I_{photo} = \frac{i_{photo}^{(1)}}{p_c} \quad (1-39)$$

With p_c defined by the equation (1-28). The photocurrent becomes:

$$I_{photo} = e\phi\eta \frac{p_e}{N_{QW} p_c} = e\phi\eta g_{photo} \quad (1-40)$$

The photoconductive gain is a crucial parameter for the QWIP and is defined as:

$$g_{photo} = \frac{g_{photo}^{1QW}}{N_{QW}} = \frac{p_e}{p_c} * \frac{1}{N_{QW}} \quad (1-41)$$

A simplified expression can be written under some assumptions such as;

$$p_e \approx 1 - p_c \approx 1, \text{ and } \tau_{capt} \gg \tau_{trans} \text{ then } p_c \approx \frac{\tau_{trans}}{\tau_{capt}} \ll 1$$

$$g_{photo} \approx \frac{1}{p_c N_{QW}} \approx \frac{\tau_{capt}}{N_{QW} \tau_{trans}} = \frac{\tau_{capt}}{\tau_{trans}^{tot}} = \frac{\tau_{capt} v}{N_{QW} L_p} \quad (1-42)$$

With $\tau_{trans}^{tot} = N_{QW} \tau_{trans}$ which is the transit time over the entire structure. This expression of the photoconductive gain corresponds to the conventional theory of photoconductivity given in the textbook. The [Table 1-1](#), summarises the typical value of essential phenomena in the physics of QWIPs.

Phenomenon	Time duration	Reference
τ_{scat} : Scattering time ($= 1/\tau_{life}$: excited electron lifetime)	1 to 2 ps	[90], [133]
τ_{capt} : Capture time	1-7 ps	[132]–[134]
$\tau_{transit}$: Transit time through one period	0.4 ps (about 17 ps for 40 periods)	[132]
τ_{esc} : Escape time	~ 0 ps (BQC and BC)	
τ_{relax} : Relaxation time	0.5 and 1 ps	[135]–[137]

Table 1-1: Typical values of essential phenomena in QWIP principle based on theory and experiment on GaAs/AlGaAs design.

Self-consistent model

A more complex model has been developed by Ershov et al. [129], [138]. This model considers the accumulation or depopulation of electrons by, not only, solving the continuity equation but also by adding the Poisson equation. As shown in *Figure 1-24*, this is especially important for the first period of the QWIP where the electric field is concentrated to extract carrier from the contact layer. This approach will be imperative to understand the results of Chapter 3.

Figure 1-24: Simulation using self-consistently method for QWIP structure composed of 4, 8 and 16 QWs. The bias voltage applied is 10 kV.cm^{-1} shown in dashed line (extracted from:[38], [129], [138]).

Optimisation of the structure

The previous assumptions done for the calculation of the photoconductive gain in equation (1-42) are especially true for the optimum designs of the quantum well structures. As shown in Figure 1-25, three different designs can be identified in the QWIP configurations:

- **Bound-to-bound (BB)** design: the upper state is positioned in the QW ($E_2 < V_B$). Therefore, the electrons excited must tunnel through the barrier or a part of the barrier to escape toward the continuum.
- **Bound-to-quasicontinuum (BQC)** design: the upper state is located in resonance with the top of the barrier ($E_2 = V_B$).
- **Bound-to-continuum (BC)** design: the upper state is sited above the top of the barrier ($E_2 > V_B$).

Figure 1-25: Three different QW structures the thickness of the well influence the position of the upper state.

QWIP designs have considerably improved since the first design which was a bound-to-bound structure [139]. By raising the barrier thickness to avoid tunnel-induced dark current and by reducing the width of the well to bring the second energy level beyond the barrier and consequently, drastically increase the escape time ($p_e \approx 1$) of the excited electrons which has the consequence of improving the performances of the QWIP. The Bound-to-quasicontinuum design [140] were followed and enhanced by placing the upper state in resonance with the top of the barrier instead of beyond the barrier. So far, this structure is known as the optimal configuration leading to a better oscillator strength and lower dark noise current than BC design. The BQC design was introduced by Gunapala and Bandara in 1995 [141]. The BB structure leads to a slightly thinner FWHM than BCQ design while BC QWIP is characterised by a broader spectrum, especially toward higher energy.

1.5.6. Photoconductive transport and figure of merit

Noise Spectral density in QWIP

In this section, we will describe the different sources of noise in the QWIPs and define their figure of merits using the previous calculation thanks to the rate equation model.

Photoconductive devices display several sources of noise;

- $1/f$ noise which is intrinsic to all electrical circuits, this source is a complex noise and is an ongoing research topic still under debate [142]. Experiments on GaAs QWIPs show that it can be neglected due to its low contribution to the current.
- Johnson-Nyquist noise is an electrical noise due to the thermal agitation of the carriers and is defined as; $i_{n,J}^2 = \frac{4k_B T}{R} \Delta f$. It is present in all resistive devices, its contribution remains lower than the dark and background current [143] and will be neglected here.
- Dark current noise.
- Background noise.

Other mechanisms can introduce noise to the system but, as the contribution of the dark current and background photocurrents are the main sources which limit the detector, we will focus our description on these two mechanisms. As we saw previously, the dark current is due to the thermal excitation of electrons in the quantum well, due to its exponential character with the temperature it is, far ahead, the primary sources at high temperature. At low temperature, the main contribution is the photoexcited electrons due to the background radiation of the surrounding object at 300 K described earlier.

Therefore, the total noise current flowing in the circuit is the sum of these two factors and defines as:

$$I_{total} = I_{background} = I_{dark} + I_{ph,bg} \quad (1-43)$$

Moreover, the noise spectral density within the detector bandwidth Δf can be written as:

$$\begin{aligned} i_N &= \sqrt{4e(g_{noise} I_{dark} + g_{photo} I_{ph,bg}) \Delta f} \\ &= \sqrt{4eg(I_{dark} + I_{ph,bg}) \Delta f} = \sqrt{S_i \Delta f} \end{aligned} \quad (1-44)$$

Where S_i is the current noise power spectral density, I_{dark} the current introduced by the thermal excitation, $I_{ph,bg}$ the photocurrent generated by surrounding objects and, $I_{background}$ the sum of these two contributions. In the case of photoconductors, we can approximate that the photoconductive gain is equal to the noise gain ($g_{noise} = \frac{1}{N_{QW} * p_c} \approx g_{photo} = \frac{p_e}{N_{QW} * p_c} = g$). Therefore, g is the photoconductive gain, using $p_e \sim 1$ (as defined in the previous paragraph). To eliminate the dark current, we need to decrease the temperature of the detectors, while to remove the background photocurrent, it is a necessity to reduce the temperature of the surrounding object, and consequently, it is required to use a cryo-shield cooled at low temperature. The background photocurrent will be the one coming from blackbody radiation of the cryo-shield at low temperature.

In the case of wanted sources which can be very narrow as lasers or very broad as blackbody radiations, the photocurrent introduced by the sources depends not only on the factors introduced earlier, but also on the solid angle that the source does with the active area. It is related to the flux of photons “seen” by the detector also described as the field

of view. The background photocurrent is often used to quantify the sensitivity of the photodetector. In this case, the maximal radiation collected by the detector corresponds to a field of view of 2π due to surrounded object at 300 K. The responsivity of the detector is the ratio between the power “seen” by the detector and the photocurrent generated by it. One must consider the spectral overlap between the absorption spectrum of the detector and the emission spectrum of the source. We can write it as:

$$R = \frac{I_{photo}}{h\nu\Phi} = \frac{e}{h\nu} \eta g_{photo} \quad (1-45)$$

With R in $[A/W]$, $g_{photo} = \frac{p_e}{N_{QW} \cdot p_c}$ the photoconductive gain and $\eta \propto N_{QW}$ the absorption coefficient. Consequently, the responsivity does not depend on the number of quantum wells (N_{QW}) and the detector area but depends only on the internal properties of the detector. To optimise the responsivity, the probability of escape p_e should be equal to the unity, i.e. the escape rate ($1/\tau_{esc}$) much higher than the relaxation rate ($1/\tau_{relax}$) which is verified for the structures BC and BQC. The BB structure needs an upper state very close to the top of the barrier to have a tunnelling escape time much faster than the relaxation time.

1.5.7. The figure of merit: Background-limited detectivity and BLIP regime

Using the equation (1-44), the background-limited signal to noise ratio can be written as:

$$S/N_{BL} = \frac{I_{ph,bg}}{i_N} = \frac{I_{ph,bg}}{\sqrt{S_i \Delta f}} \quad (1-46)$$

With I_{photo} the signal analysed. The background-limited (BL) Noise Equivalent Power (NEP) is the signal power required to obtain the same signal and noise power for an integration of a bandwidth of $\Delta f = 1 \text{ Hz}$.

$$NEP_{BL} = \frac{\sqrt{S_i}}{R} \quad (1-47)$$

With NEP in $[W/Hz^{1/2}]$. The NEP still depends on the detector dimension, to make a universal figure of merit which would not rely on the area, we define the specific detectivity as:

$$D^* = \frac{\sqrt{A_{det}}}{NEP} \quad (1-48)$$

Therefore, the background-limited detectivity (which help to determine the temperature resolution of a thermal imager) is:

$$D_{BL}^* = \frac{\sqrt{A_{det}}}{NEP_{BL}} = \frac{R\sqrt{A_{det}}}{\sqrt{4eg(I_{dark} + I_{ph,bg})}} \quad (1-49)$$

With D^* in $[cm.\sqrt{Hz}/W]$, we remind that this formula is specific to QWIP because different noise currents have been neglected in the previous paragraph which may be dominant or non-negligible for other photoconductive devices. The detectivity is strongly dependent on the temperature, we can, therefore, observe two main regimes:

- At Low-Temperature (LT) where $I_{background} \approx I_{ph,bg} \gg I_{dark}$ the background-limited detectivity is:

$$(D_{BL}^*)_{LT} = \frac{R\sqrt{A_{det}}}{\sqrt{4egI_{ph,bg}}} \propto \sqrt{\frac{\eta}{\Phi_{BG,300K}}}$$

- At High-Temperature (HT) where $I_{dark} \gg I_{ph,bg}$ the Dark-Limited (DL) detectivity is:

$$(D_{BL}^*)_{HT} \approx D_{DL}^* = \frac{R\sqrt{A_{det}}}{\sqrt{4egI_{dark}}} \propto \frac{1}{\exp^{-\frac{E}{k_B T}}}$$

Figure of merit

As shown in [Figure 1-26](#) the LT regime is independent of the temperature and inversely proportional to $\Phi_{BG,300K}$, the 300 K photon flux, while at high-temperature regime is highly-dependent of the temperature and inversely proportional to the exponential behaviour of the dark current.

Figure 1-26: Theoretical background-limited detectivity with the Low

Temperature (LT) and High-Temperature (HT) regimes. The BLIP

temperature is defined as the crossing point between the two linear curves.

The intersection of the two regimes corresponds to Background-Limited Infrared Performance (BLIP) temperature where the photocurrent is equal to the dark current

$I_{dark} = I_{ph,bg} = \frac{I_{background}}{2}$. Thus, the detectivity corresponding to this specific temperature is:

$$D_{BL}^*(T_{BLIP}) = \frac{R\sqrt{A_{det}}}{\sqrt{8egI_{ph,bg}}} = \frac{(D_{BL}^*)_{LT}}{\sqrt{2}} \quad (1-50)$$

T_{BLIP} is particularly important to determine the operating temperature of a device. This regime corresponds to a domain where the dominant noise current is the background current, to keep a good sensitivity, detectors are used in this regime. For imagery applications, it is not necessary to cool down the detector well below the T_{BLIP} to reduce

the noise as below this temperature the limiting factor is the background photocurrent. An important factor for imagery application is the limiting temperature variation measurable by the detector also called NETD. In other words, the ΔT which induce a background power equals to the NEP:

$$NETD = \frac{NEP_{BL}}{dP_B/dT} \quad (1-51)$$

With $NETD$ in $[K]$, P_B the background power and NEP_{BL} the background-limited noise equivalent power. The typical value for uncooled micro-bolometer cameras is about 40 mK, while the photoconductive cameras cooled at cryogenic temperatures on the market have an NETD of about 20 mK. H. Schneider and al. demonstrated high-resolution QWIP FPAs with an NETD of 9.6 mK for the 8-12 μm and 14.3 mK for the 3-5 μm [144].

Chapter 2 Study of high-speed behaviour and injection locking in quantum cascade laser

2.1. Process and main setups

2.1.1. Fabrication process

In this section, we will briefly present the, already well known, the fabrication process for standard ridges QCLs. The quality of the fabrication is critical to avoid any leakage current and other artefacts which can degrade the performance of the laser. The fabrication process of ridge QCL can be summarised by the following steps represented in Figure 2-1:

a) and b) illustrate the deposition of hard mask in SiO₂ to pattern the ridges using lithography process with a positive photoresist. c) Shows the etching step through the active region done by dry etching using Inductively Coupled Plasma (ICP) system, we then remove the hard mask using HF solution. d), e) and f) are the top windows opening by a lithography process. The insulator layer used, in our case, is 300 nm of SiO₂ by Plasma-Enhanced Chemical Vapor Deposition (PECVD). This layer is etched on the top of the ridge to force the current going through the active region. g) And h) display the deposition of the ohmic contact composed of Ti/Au on the top of the ridges with a lift-off process to remove the conductive layer between ridges and isolate them. To improve the thermal management and allow higher temperature operation of the sample, we polished

the device to remove a thick layer of the InP substrate. Finally, i) express the deposition of the back ohmic contact.

Figure 2-1: Flow diagram of the MWIR QCLs fabrication process.

The final step is to mount the wafer on a holder. The mounting process has significant influences on the heat extraction and therefore, can drastically reduce the temperature of use. Standard cryogenic lasers are mounted on copper holder using thin indium layer, while higher temperature operation requires better thermal extraction as described later. The contacts are ensured by wire bonding on gold pads for low-frequency use, while high-frequency behaviour requires other configurations also detail in the next paragraph. In

this chapter, the lasers used are standard Buried-heterostructure. The process is different and requires a regrowth of Fe-doped InP to increase the thermal extraction.

2.1.2. Characterisation setups

In order to use the QCL in CW operation, we place them in a cryostat and cool them down to cryogenic temperature. The devices are placed on a copper holder with thermal probe linked to a Peltier for precise control of the temperature. Moreover, thermal paste is used between all contacts with the mount to improve the thermal extraction. We applied a pulse current with a low duty cycle for pulse operation use. A DC current can be applied if CW operation is wanted. The current is recorded using a current transducer at the entrance of the cryostat, and moreover, the voltage is read by the oscilloscope to a resistance placed in parallel to the laser. The oscilloscope is synchronised with the pulse generator and read the relevant data. The pertinent information is reported using computer-assisted data acquisition software written by LabView through GPIB instruments.

Figure 2-2: Standard characterisation setup; QCL is shining on the FTIR for spectrum characterisation and focusing on the thermopile for Power-Current-Voltage (LIV) characterisation.

The two-principal typical setups are shown in [Figure 2-2](#):

- Dot circle in [Figure 2-2](#) represents the configuration for standard LIV curve measurements; the shortest focal length lens is placed to collimate the laser beam and the second will focus it on the thermopile of the power meter. If the focal length is not able to collect the entire highly divergent beam, the calibration is done by doing the ratio of the power obtained by putting the detector right at the laser end and after the optical system at the same current. The power as a function of the current and voltage is recorded using the GPIB system. The standard LIV curve is displayed in [Figure 2-3 \(a\)](#).

- The Figure 2-2, excluding the dot circle, illustrates the spectrum characterisation. The collimated beam is sent to the Fourier-Transform InfraRed spectroscopy (FTIR) instrument which analyse the Fourier-transform of the time domain interferogram. The spectrum is, then, collected for different configuration, a typical multimode spectrum is displayed in Figure 2-3 (b).

For a better understanding, the influence of the bias voltage on the structure is illustrated for the different region of the LIV curve in Figure 2-3 (a).

Figure 2-3: Typical (a) LIV curve with structure corresponding to different regimes and (b) spectrum of a QCL emitting at 4.7 μm .

As seen in [Figure 2-3](#) (a), at a low bias voltage, i.e. before the threshold, the structure is not aligned and meagre current flow across the structure. The upper energy level of the laser transition is not occupied. At higher bias voltage, the structure is aligned, and allow the electrons to flow from lower energy state to the injector and then to the upper laser

state where the population inversion is built. After the roll-over, the energy levels are not any longer aligned, and the electrons are not allowed to cascade the structure.

2.1.3. List of samples

During my thesis I worked on QCLs process at NTU (EEE) and MPQ, using commercial wafer. *Table 2-1* is a summarised on the main lasers used in this thesis.

Sample name	Origin	Dimension (μm^2)	Composition (InP substrate)	Wavelength absorption (μm)
QCL-9-3-1A	MPQ	3mm*10 μm	InGaAs/InAlAs (commercial wafer)	9.2 μm
QCL-9-3-1B		3mm*12 μm		
QCL-9-3-1C		3mm*12 μm		
QCL-9-4-2A		4mm		4.7 μm
QCL-4-3-1A		3mm*10 μm		
QCL-4-4-1B		4mm		
QCL-4-3-2A		3mm		

Table 2-1: List of samples used.

2.2. High-speed behaviour

2.2.1. Transient time

In this paragraph, we will have a closer look at the transient behaviour of QCL and study the theoretical frequency response of our quantum devices to a small modulation of the driving current. Using the rate equation developed in Chapter 1, we can define the frequency response as:

$$|h(\omega)|$$

$$= \frac{1}{\sqrt{1 + \omega^4 \tau_{phot}^2 \tau_{stim}^2 + \omega^2 \tau_{phot} \tau_{stim} \left(\frac{\tau_{phot} \tau_{stim}}{\tau_3^2} + \frac{\tau_{phot}}{\tau_{stim}} + 2 \frac{\tau_{stim}}{\tau_3} - 2 \right)}} \quad (2-1)$$

$$\tau_{phot} = \frac{n_{eff}}{c\alpha_{tot}} \text{ and } \tau_{stim}^{-1} = \left(\frac{J}{J_{th}} - 1 \right) \tau_3^{-1} \quad (2-2)$$

With τ_{phot} the lifetime of the photon in the cavity which is inversely proportional to the total loss in the cavity, τ_{stim} the lifetime of the stimulated emission directly linked to the structure alignment before the gain saturation and τ_3 the upper state lifetime. For a QCL in the MIR region, the upper state lifetime is inferior to the picosecond ($\tau_3 = 0.6 \text{ ps}$), the typical losses are in the order of 10 cm^{-1} corresponding to a lifetime of the photon of $\tau_{phot} = 11 \text{ ps}$.

Figure 2-4 study the frequency response dependence to the different parameters.

Figure 2-4: Simulation of the small signal response of the photon population to a modulation current of the QCL using $\tau_3 = 0.6$ ps, $\tau_{phot} = 11$ ps and τ_{stim} following the equation (2-2) for (a) variation of the upper state lifetime at $\frac{J}{J_{th}} = 2$ and (b) different value of the current ration $\frac{J}{J_{th}}$.

As illustrated Figure 2-5 (b) the lasers used in this chapter are standard Buried-Heterostructure (BH) of InGaAs/InAlAs active region grown on InP substrate by MBE. The wavelength emissions are in the two atmospheric windows at 4.7 and 9.2 μm . The active region is about 2 μm thick, and the width of the laser is from 10 to 14 μm . The size of the cavity is chosen according to the frequency of the beating signal wanted, corresponding to a cavity length of 2, 3 and 4 mm. Furthermore, the device is thin down up to 70-80 μm , and the active region is embedded in a 50 Ω impedance microstrip line to improve the RF injection.

2.2.2. Electronic cut-off frequency

As for all electronic system, the electrical circuit can act as a low-pass filter preventing the propagation of high-frequency waves. As discussed in Chapter 3, wire bonding

introduces high inductance which drastically decreases the cut-off frequency of the circuit; therefore, we ensure the contact by an RF launcher. The resistance and the capacitance of the devices usually can be improved only by a specific design for high-frequency behaviour. In consequence, the cut-off frequency is the RC electrical circuit of the device including the resistance and capacitance of the active region and the insulator layer which is InP:Fe in our case. *Figure 2-5* (a) shows the equivalent circuit of the QCL, the total capacitance is the sum of the capacitance of the InP:Fe and the active region. Due to the high resistance of the insulator layer, $R_{AR} \ll R_{InP:Fe} \approx 280 \text{ M}\Omega$, the resistance of the circuit is the resistance of the active region.

Figure 2-5: (a) Equivalent electrical circuit of the quantum device and (b) illustration of the devices; in red the active region and in green the insulator layer (InP:Fe) with the dimension.

The resistance of the active region is the differential resistance calculated with the variation of the IV curve of the laser: $R_{AR} = \frac{dV}{dI}$ which in our case is relatively low. *Figure 2-6* (a) exhibit the differential resistance of the QCL-9-3-1B emitting at $9.2 \mu\text{m}$. Our QCLs at $4.7 \mu\text{m}$ show lower differential resistance. For the rectification measurement,

the RF generator injects the signal via a 50Ω impedance into the QCL. An estimation of the capacitance of the circuit can be calculated using the equation:

$$C = \frac{\epsilon_0 \epsilon_r * S}{d} \quad (2-3)$$

With S and d the surface and width of the active region or insulator layer. ϵ_r the permittivity of the materials; we took $\epsilon_{r,AR} = 10.9$ and $\epsilon_{r,InP:Fe} = \epsilon_{r,InP} = 12.5$.

Therefore, both capacitances are dependent on the dimension of the device. Figure 2-6 (b) displays the evolution of the capacitance and thus, the cut-off frequency at -3 dBm as a function of the dimension for a differential resistance of 2Ω . The capacitance of the QCLs are relatively high, three orders of magnitude higher than the capacitance of the QWIP. Due to the low differential resistance for high current density ($> J = 2 J_{th}$), the cut-off frequency of the electrical circuit remained relatively high.

Figure 2-6: (a) Variation of the differential resistance in black and cut-off frequency of the RC circuit in blue as a function of J/J_{th} (for QCL-9-3-1B and a capacitance of 4 pF) and (b) capacitance of the active region as a function of the surface for a differential resistance of 2 Ω . Both are done for a 2 μm thick active region.

The frequency response of the QCL is the multiplication of the intrinsic transfer function of the QCL, $h(\omega)$, and the transfer function of the electrical circuit.

2.2.3. High-speed setups

As described earlier, the high-speed behaviour requires a specific configuration. Furthermore, the characterisation of CW-QCLs often need cryogenic temperature; the cryostat must be adapted for high-speed transmission. All the connectors and cables used have a bandwidth of more than 30 GHz. The AC and DC injection are done using the RF launcher that we carefully place on the ridge using a microscope as seen [Figure 2-7](#). We used a 45 GHz bias-T to ensure simultaneity the DC and AC injection. The characterisation of the IV curves and spectrum of the lasers is done as explained in 2.1.2. Characterisation setups.

Figure 2-7: High-speed adaptation in the cryostat, RF launcher ensuring the AC and DC current.

Figure 2-8 (a) illustrates the rectification setup used to study the high-speed behaviour of the QCLs. The microwave rectification technique is a way to explore the frequency response of our device by applying an RF signal to the QCL and measuring the variation of its DC biasing current. This value reflects the frequency roll-off behaviour of the electrical circuit limited by the device capacitance and differential resistance, along with other parasitic effects [38], [27]. To reduce the noise of the measurements we use a lock-in technique with the electrical modulation of the microwave source. This setup is more explained in Chapter 3.

Figure 2-8: (a) Rectification method for high-speed characterisation and (b) setup used to inject and analyse the beatnote.

Figure 2-8 (b) shows the standard setup used for the investigation of the RF injection on the spectrum. To be able to inject RF frequency and see the effect of this injection on the beatnote we employ an RF circulator which allows separating the different signal. Indeed, the injection is, as previously, directly injected in the QCL via the launcher, but the beatnote which is irradiated from the mixer (here the QCL itself) can be detected and send to the spectrum analyser via the RF launcher and through the circulator. We realised most of the 2D injection-lockings by collecting the RF frequency and the beatnote on the high-speed QWIP presented in Chapter 3.

2.3. QCL characterisations

First, in this paragraph, we will present the standard IV curves and the spectrum as a function of the current. We will also characterise the high-speed behaviour using the rectification method up to 30 GHz and draw a new high-speed characterisation method by mixing two frequencies. Second, we will study the evolution of the spectrum under RF injection for different injection power and frequencies, and then, we will explain the

different effect observed. The characterisation will be done for QCL at 4.7 μm and 9.2 μm separately while for the experiment using RF injection, results will be presented in parallel.

2.3.1. QCL at 4.7 μm

Using the standard setup, coupled with the high-speed implementation we can characterise the electrical and optical part of the laser. *Figure 2-9* (b) represents the IV curve in CW operation at 80 K of the *QCL-4-3-1A* (3 mm * 10 μm), the vast dynamic range is a good indicator of the quality of the laser. The spectrum, in *Figure 2-9* (a), is recorded at each dot of the IV curve; at the threshold (220 mA) the spectrum is composed of a single mode operation while when increasing the current the apparition of multimode regime emerges and becomes broader with the current.

Figure 2-9: Characterisation of the QCL-4-3-1A at CW operation at 80 K (a) spectrum as a function of the current following the colour dots on the IV curve and (b) IV curve.

The high-speed performances of our QCLs are investigated using the rectification technique explained in the previous section. *Figure 2-10* (a) exhibits the normalised rectification curves recorded at several $\frac{J}{J_{th}}$ ratios. The RF injection power is 10 dBm, and the step frequency is 100 MHz. As described by the theory, before the gain saturation the cut-off frequency increases which introduce an intensification of the normalised amplitudes with the $\frac{J}{J_{th}}$ ratio as seen *Figure 2-10* (b).

Figure 2-10: QCL-4-3-1A; (a) Normalised rectification curves for different $\frac{J}{J_{th}}$ ratio, (b) the evolution of the normalised amplitude of the rectification curves at 21.6 GHz, (c) the stimulated lifetime calculated with the fitted curve (in black) and the theoretical value (in red) and (d) the superposition of the rectification curve and their fits.

The simulations are done by taking into account the inherent frequency response described in the equation (2-1) and (2-2). $\tau_3 = 0.6 \text{ ps}$, τ_{phot} is calculated and fixed at 11 ps, this value corresponds to a typical values for MWIR QCLs [145]. The differential resistance and capacitance of the devices are fixed at a calculated value of respectively 1.2Ω and 1.44 pF . For the frequency response of the RC circuit we added the 50Ω to the differential resistance of the QCL. τ_{stim} is settled as a free parameter. *Figure 2-10* (c) shows the theoretical and the fitted values of the stimulated lifetime. Due to the of the

different parameters, the uncertainty on the absolute value of τ_{stim} is relatively high. However, the variation of its value follows the theoretical trend. *Figure 2-10* (d) corresponds to the superposition of the experimental curve and the fitted curves. Due to the alignment of the structure which does not allow the current to flow through it, the rectification below the threshold sees a differential resistance of the active region much higher than after the threshold. This is translated in a lower cut-off frequency of the electrical contribution for this current.

The electrical resonances, corresponding to a centimetre scale, are not inherent to the device but are due to the reflections in the electrical circuit and are often experimentally observed [145], [146].

2.3.2. QCL at 9 μm

We realised the same experiment at a wavelength of 9 μm . *Figure 2-11* shows the IV curve of the laser in (b), and the spectrum as a function of the current in (a). As for the 4.7 μm , we notice a significant widening of the spectrum at high current. For this QCL, the threshold is at 230 mA and the roll-over at about 800 mA.

Figure 2-11: Characterisation of the QCL-9-3-1A at CW operation and 80 K (a) spectrum as a function of the current following the colour dots on the IV curve and (b) IV curve.

Figure 2-12 (a) shows the rectification of the QCL for different $\frac{J}{J_{th}}$ ratio. We notice that the amplitude of rectification point does not increase up to the roll-over, we notice in *Figure 2-12* (b) a drop of the amplitude as soon as $2 J_{th}$. This also can be seen in *Figure 2-12* (c) where the fitted stimulated emission does not follow the theoretical value. This phenomenon may be due to the variation of the capacitance with the current density. Indeed, differential resistance is higher for QCL at $9 \mu\text{m}$ (1.2Ω at $4.7 \mu\text{m}$ for 2 to 3Ω at $9.2 \mu\text{m}$), plus the decrease of the capacitance with the current density will decrease the cut-off frequency of the signal faster than for QCL at $4 \mu\text{m}$ [146].

Figure 2-12: QCL-9-3-1A; (a) Normalised rectification curves for different $\frac{J}{J_{th}}$ ratio, (b) the evolution of the normalised amplitude of the rectification curves at 21.6 GHz, (c) the stimulated lifetime calculated with the fitted curve (in black) and the theoretical value (in red) and (d) the superposition of the rectification curve and their fits.

The parameters of these simulations are; $\tau_3 = 0.6$ ps, and τ_{phot} fixed at 11 ps. The differential resistance and capacitance of the devices are adjusted at a calculated value of respectively 2 Ω and 1.44 pF. As previously, τ_{stim} is settled as a free parameter.

2.3.3. RF mixing and demodulation

Another way to investigate the high-speed behaviour could be to mix two RF frequencies in the QCL and study the amplitude of the beating signal between the two RF signals. In

this experiment, we combined in a mixer (here the QCL) the beatnote of 2 mm in *Figure 2-13* (a) and 3mm in *Figure 2-13* (b) with the injection of an RF signal at a fixed frequency. Therefore, the mix of the two frequencies will generate a beating frequency at a frequency corresponding to the interval between them.

Figure 2-13: (a) beating signal generated from the mix of the 2 mm QCL beatnote and a 10 dBm injection at 230 MHz, (b) study of the generated frequencies from the beatnote of a 3 mm QCL and RF injection at 18 dBm at different frequencies, inset: beatnote of the 3 mm QCL.

The amplitude of the generated frequency will be dependent on the intensity of the two signals. This generation of a new frequency is very beneficial for some applications such as the demodulation of a high-frequency signal. The demodulation process can require a complex and bulky system in the MIR. Such dispositive may be a solution to simplify such system in the MIR. Furthermore, due to the similarity with QWIP, we can most probably achieve same results using our detectors.

2.4. Injection-locking by microwave modulation

The development of high-speed QCLs has many advantages, including the abilities to inject RF signals at the round-trip frequency of the cavity of the laser. In certain circumstances, it can generate sidebands and significantly broaden the spectrum. Moreover, it can stabilise the spectrum, and in a specific range of frequency around the round-trip cavity can tune the FSR over a part or the entire spectrum. To investigate the RF injection at the round-trip cavity, we use the setup seen in *Figure 2-8* (b). In the second part of this chapter, we will study the high-resolution spectra with and without external injection.

2.4.1. Evolution of the spectrum under RF injection

Figure 2-14 (a) and (b) show the evolution of the spectrum with the RF injection at the round-trip cavity of respectively, 4 mm QCL-4-4-1B emitting at 4.7 μm and 3 mm QCL-9-3-1C emitting at 9.2 μm . The injection mechanism is explained in detail in 1.4. Quantum cascade laser frequency combs. It generates sidebands of the free running modes which can be amplified by the gain curve and redistribute the energy of the powerful modes over the spectrum. The consequences of this effect are a broadening the spectrum and creating a more homogeneous distribution of amplitude in the modes. Therefore, as displayed in *Figure 2-14* (a) the spectrum of the QCL at 4.7 μm goes from about 25 cm^{-1} without injection to 54 cm^{-1} with 30 dBm injection. Regarding the QCL at 9.2 μm in *Figure 2-14* (b) goes from about 29 cm^{-1} to 92 cm^{-1} with injection at 30 dBm.

Figure 2-14: Evolution of the spectrum for several power injections for (a) 4 mm QCL-4-4-1B at 4.7 μm with a current of 300 mA corresponding to 1.6 Jth and (b) 3 mm QCL-9-3-1C at 9.2 μm with a current of 450 mA corresponding to 1.8 Jth.

The RF injections at the round-trip frequency increase the number of the modes in the spectrum. *Figure 2-18* studies the augmentation of the number of modes as a function of the injected power.

We studied this effect with a threshold of 1 % and 10 %. The inset is the percentage of modes within an amplitude of 90 % of the highest peak. Considering 1 % as the total number of modes, we can see that the percentage of modes included in this range goes from 20 % to 60 % with the RF injection.

Figure 2-15: Evolution of the number of modes for the 4 mm QCL-4-4-1B in the previous configuration. The black curve is the number of them within an amplitude of at least 10 % of the highest. The inset represents the percentage of the modes included in that range.

The most majority of the cavity modes are generated up to an injection of 20 dBm, the important increase at low power corresponds to the redistribution of the energy of the principal cavity modes by the generation of sidebands frequency. Figure 2-16 shows the mechanism of sidebands generation at the round-trip frequency of the spectrum without injection (in red) through production of sidebands after an injection at 0 dBm (in black) which correspond to a power of 1 mW. The generations of the sidebands are locked at the frequency of the RF injection which is the round-trip cavity. We expect an injection efficiency of few percents, which means, that tens of μW are enough to have a non-negligible effect on the spectrum. After the injection of 20 dBm (100 mW) the generation

of new modes slow down, but the distribution of intensity in the spectrum continue to operate.

Figure 2-16: Evolution of the high-intensity group of modes under injection. The dashed arrow illustrates the sidebands generations.

This configuration has the potential to stabilise the frequency and phase of the cavity modes leading to a redistribution of the amplitudes of the cavity modes.

Figure 2-17: Evolution of the beating signal (a) and the spectra (b) with the injection power.

However, the generation of sidebands using the high-power RF injection can broaden the spectrum enough to generate optical modes where the dispersion is high enough to not be compensated by the FWM mechanism (most probably due to the dispersion introduced by the gain curve as described earlier). This phenomenon is observed by the creation of a pedestal area around the injection. *Figure 2-17* (b) represents the broadening of the spectrum as a function of the injection power, a part of the generation of new optical modes present in the yellow area see higher dispersion creating the pedestal area around the injection observed in *Figure 2-17* (a). [147], [73] Same effects have been observed with lasers emitting at $4.7\ \mu\text{m}$.

Nevertheless, for some cases the pedestal introduced by high RF injection can be a phase-noise pedestal due to a collective instability involving all the modes simultaneously [148]. In the next section, we will detail the injection process in and out of the locking range.

2.4.2. Injection-locking mechanism

Figure 2-18 (a) and (c) show the injection-locking by increasing the injected power at a fixed frequency. As discussed earlier the locking-range of the slave oscillator (here the

beatnote) is proportional to the square intensity of the master oscillation (here the RF injection). We can see here the creation of sideband (in green) which indicate a good injection, at an injected power of 26 dBm the beatnote is fully injected. *Figure 2-18* (b) and (d) display the injection locking by tuning the frequency injection around the round-trip frequency at a fixed power. As earlier, we can see the generation of a sideband in the first part (green colour), the second part (red colour) indicate the injection-locking and the third part (in blue) the release of the beatnote with again the creation of sidebands.

Figure 2-18: Injection locking by; (a) and (c) increasing the power injected and (b) and (d) varying the gap between the injection and the beatnote. The 2D injection-locking does not correspond to the same current but illustrate well the phenomena.

According to the equation (1-21), the locking range is proportional to the square roots of the injected power $\Delta\omega_{lock} \propto \sqrt{I_{injection}}$. Figure 2-19 shows the square root dependence of the locking range with the injected power from -5 dBm to 14 dBm. The QCL used is a laser emitting at 9 μm with a current of 650 mA, and the operating temperature was stabilised to 80 K. The X-axis is the measurement of the signal on the spectrum analyser, and the Y-axis is the injected microwave signal. The colour scale shows the intensity of the signal in dBm unit and the acquisition has been done by 100 times averaging, the resolution bandwidth has set to 3 KHz.

Figure 2-19: Radio frequency spectra as a function of injected microwave frequency for different powers from a) -5 dBm, b) 0 dBm, c) 5 dBm, d) 10 dBm up to e) 14 dBm and f) Locking range measurement; the dots correspond to the different injected microwave powers reported in figures a-e. The red solid line corresponds to the square root fit of the data.

The sidebands generation, observed in these figures, correspond to an injection which is not in the locking range of the slave oscillator. In this case, the creation of sidebands in the spectrum is too far from the next cavity mode and does not contribute to the evolution

of the spectrum. Figure 2-20 illustrates the spectrum under an RF signal out of the round-trip frequency. This phenomenon is also explained in detail in 1.4. Quantum cascade laser frequency combs.

Figure 2-20: Influence of the RF injection, relatively far from the round-trip frequency, on the spectrum.

Figure 2-21 shows the amplitude of the beatnote while decreasing its gap with the RF signal. The green area shows a good injection-locking where the beatnote is fully absorbed by the RF injection, in the opposite the red zone indicates a destabilisation of the spectrum by the injection, the beatnote does not seem to be injected in the master oscillator anymore. The evolution of the beating signal with the gap shows that, even though the beatnote is not fully locked, a certain percentage of its amplitude is locked in the master oscillator.

Figure 2-21: (a) spectrum corresponding to different injection frequency, (b) the evolution of the beating signal as a function of the gap between the beating signal and the RF injection at 10 dBm and (c) beatnote of the laser without injection.

Figure 2-22 shows the destabilisation of the injection on the bigger RF signal scale and for different examples. The 2 D schemas are not related to this laser but exhibit the same instability over a specific range of injection. The green area corresponds to an injection locking while the red zone indicates a destabilisation of the beatnote. This injection seems to destabilise the FSR of the multimode laser, the beatnote is not injected anymore and does not appear around the round-trip cavity. This effect is not well known but seems to be emerging at high injection power. Additionally, contrary to injection at a lower

frequency than the beatnote, the one at higher tends to favour the destabilisation of the FSR.

Figure 2-22: (a) 2D injection-locking for a 2 mm QCL, (b) the evolution of the beating signal as a function of the delta between the beating signal and the RF injection and (c) 2D injection locking for a 3 mm laser. All these experiments were done with an injection power of 10 dBm.

More works are needed to understand this mechanism deeply. Nevertheless, injection to the lower frequency of the beatnote seems to exhibit a better injection than for higher frequency.

2.4.3. Thermal and pulling-effect

The beating signal of the laser is inversely proportional to the length of the cavity and the group refractive index. The variation of the beating signal with the current is related to the thermal parameter of the sample. The increase of the current will lead to an augmentation of the Joule effect and therefore the temperature of the sample. The refractive index of the devices is consequently modified since $\frac{dn}{dT} = 10^{-4} K^{-1}$. In consequence, an increase of the temperature will decrease the free-spectral range of the

spectrum and therefore, drift the beating signal of the laser toward lower frequency as shown by the red arrow in Figure 2-23. The injected RF signal used can exhibit high power and goes from -20 to 30 dBm. A small percentage of this power is absorbed by the sample and increase its temperature. As explained earlier, this phenomenon leads to a drift of the beatnote; toward the master oscillator in the case of injections at a lower frequency as in Figure 2-23 (b) and (c) or in the opposite direction as in Figure 2-23 (a) and (d).

Figure 2-23: (a) Illustration of the lock-in phenomenon and 2D injection-locking for a 2 mm QCL under RF injection at (a) lower frequency and (b) higher frequency than the beatnote.

This phenomenon explains why the injection is, most of the time, more efficient when it is located at a lower frequency than the beatnote, and that, especially for high injection power.

2.5. High-resolution spectrum analysis

The next measurements have been realised using a high-resolution FTIR allowing us to capture the spectrum emission of our laser with an absolute resolution of about 60 MHz. However, the implementation of zero-padding is largely increasing the peak precision. The crucial point for these measurements is not the resolution of the FTIR which gives the absolute frequency error of the points but the precision on peak positions which gives the relative error of one point with the next one. Therefore, this error may be below the resolution of the FTIR.

The analysis of the FSR using this technique can provide the evidence of RF pulling and injection-locking. However, this method does not give information on the phases of each mode and therefore, won't be able to make evidence of mode-locking behaviour.

2.5.1. FTIR Resolution and higher-order transverse modes

The zero-padding allows us to overcome the resolution by adding zeros on the interferogram and consequently, adding virtual points on the spectra. Due to a large number of points in the interferogram, the computers used could not support more than a zero-padding of 8. Obviously, the resolution will remain unchanged while the precision on the peaks will be improved by a factor 8. It is equivalent to define a peak position for a curve defined by 20 points or by 8×20 points [147].

In order, to have a smooth curve and improve a bit more the precision we can interpolate the curve by a cubic spline method as in *Figure 2-25* (a). The real precision on the peak is hard to determine. We assume:

- No zero-padding: $1,9\text{E-}3 \text{ cm}^{-1}$ (57 MHz)
- 8 zero-padding: $1,3\text{E-}4 \text{ cm}^{-1}$ (4 MHz)
- 8 zero-padding + cubic spline interpolation: $3\text{E-}5 \text{ cm}^{-1}$ (0.9 MHz)
- Error bars: $2\text{E-}4 \text{ cm}^{-1}$ (6 MHz)

Simulation of the active region has been realised for the QCLs used at 4.7 and 9.2 μm . The small width of the ridge is supposed to allow only the fundamental optical transverse mode to propagate in the waveguide, but the not-rectangular shape due to the etch during the process increases its width from 14 μm to about 30 μm as shown in the simulation *Figure 2-24*. This broadening of the active region allows the propagation of higher-order transverse modes leading to higher-order cavity modes in the spectrum.

Figure 2-24: High-order transverse modes propagating in the QCL cavity for (a) QCL at 9 μm and (b) QCL at 4.7 μm .

Most of the time, the higher-losses introduced to these optical modes make them small enough to permit a study only the main cavity mode. However, for the case of higher-order modes with an amplitude of the order of magnitude of the fundamental modes, the

investigation may become complicated and give unwanted slope as we can see in the next data (especially at the extremity of the spectrum).

2.5.2. Analysation process of the dispersion and variation of the FSR

In this section, we will first introduce the analysis of the FSR of the spectrum and the shift of the FSR under RF injection. As explained previously, the modes are recorded with the highest resolution of the FTIR, the 8 zero-padding, and the curving interpolation increase the precision on the peak position as we can see in *Figure 2-25* (a). In this work, each peak position of the spectrum is recorded and analysed. To study the FSR of a spectrum, the peak position is compared to the next or previous modes from the same spectrum. Nevertheless, regarding the variation of the FSR under RF injection it is compared to the peak position of the referent spectrum which is the no-injected one as illustrated in *Figure 2-25* (b) and (c). However, a second approach to study the shift of the FSR by RF injection would be to compare the FSR of the referent spectrum with the one of the spectrum under RF injection. But, this approach shows values of the order of magnitude of the resolution FTIR while the first analysis exhibits a variation of N times the gap between the RF injection and the beatnote with N the number of modes in the spectrum. Besides, the slope of the variation of peak position curve gives the value of the gap between the beatnote and the RF injection, i.e. the shift of FSR. The analysis process is shown and explained in *Figure 2-25* (b) and (c).

Figure 2-25: (a) Zoom-in in the peak of a mode with different interpolation. Analysis process to investigate the FSR of the laser; (b) peak position for the two spectra and (c) the variation of the peak position between S1 and S2, the slope corresponds to the shift of the FSR.

As details in 1.4.7. Dispersion in QCL, the dispersion is related to the variation of the FSR over the spectrum since the $FSR \propto \frac{1}{n_g}$. In the case of no-dispersion, n_g is constant, and the plot of the peak position as a function of the wavenumber is a perfect linear curve with a slope matching the constant value of FSR. For a dispersive spectrum, n_g vary with the frequency, every peak position of the modes which are not on the linear curve will

introduce a variation in the first derivate. This approach gives same results than different approach such as investigate the FSR directly by computing $\nu_{S2}^{N+1} - \nu_{S2}^N$.

2.5.3. Dispersion analysis

Using the high-resolution FTIR, we can see the dispersion into the MHz range. By using the method explained above; *Figure 2-26* shows the dispersion analysis of the spectrum of the QCL-4-3-2A emitting at 4.7 μm with and without injection at a current of 400 mA for (a) and (b) and 810 mA for (c). An averaging is applied to the FSR to smooth the effect of the resolution, and the yellow area represents a frequency of 10 MHz. The injected power was 27 dBm and located at the extremity of the locking range, corresponding to the measurements M4 in the next paragraph. The injection-locking is at about 7 MHz in higher frequency than the beating signal.

Figure 2-26: Upper part; study of the FSR of the free running modes, the yellow region corresponds to a frequency of 10 MHz and lower part; spectrum corresponding. QCL-4-3-2A at 400 mA without injection (a), at 400 mA with injection in the locking range at 27 dBm (b) and at 810 mA without injection (c).

As studied in [Figure 2-24](#), due to the higher-order transverse modes propagating in the cavity and the finite resolution of the FTIR, we can observe an electronic pulling effect due to the overlapping of higher-order and fundamental transverse modes. This phenomenon leads to a slight shift of the position of the maximum of the fundamental mode which produces an unwanted variation of the FSR seen in [Figure 2-26](#) (a), (b) and (c). This artefact has also been observed in other articles [147], and it does not allow us to conclude on the origin of the oscillations of the free-spectral range.

Based on the thin beating signal of the free running modes (below 1 MHz, represented in red dashed-line in *Figure 2-27* (b)) and the *Figure 2-26* (a), (b) and (c) we have strong evidence of that the QCL operate at frequency comb regime. However, a phases analysis of the modes is required to claim a frequency comb regime. With or without injection in the locking range, the high-resolution measurement does not show any dispersion under the resolution of the FTIR. Therefore, over more than 90 % of the FSR of the optical power is under 10 MHz, corresponding to a GVD below 2000 fs²/mm. These measurements show that most of the modes are involving in the thin beating signal and are frequency-locked by the FWM.

Nevertheless, for an injection out of the locking range, as displayed in *Figure 2-27*, we can observe in bleu the sideband due to the master oscillator and, in red, the pulled beatnote due to the RF injection.

Figure 2-27: QCL-4-3-2A at 400 mA with injection out of the locking range at 27 dBm; (a) upper part; study of the FSR of the free running modes, the yellow region corresponds to a frequency of 10 MHz and lower part; optical spectrum corresponding. (b) Radio Frequency spectrum corresponding to the superposition of the free-running beatnote and RF injection out of the locking range.

The study of the high-resolution spectrum for an injection out of the locking range (13 MHz away from the beatnote) in [Figure 2-27](#) (a) shows that the 40 first optical modes of the spectrum are injected and follow the new FSR submitted by the external excitation out of the locking range, but the next 70 modes (most of the optical power) are not locked and oscillate around a value following the pulled beatnote. The pulled beating signal seems to be wider than the original beatnote, this injection out of the locking range seems to introduce GVD in the spectrum.

Same measurements have been done on QCLs emitting at 9 μm for different current density and give the same conclusion. A second study of the locking process will be done by studying the variation of the FSR under external injection near the round-trip frequency shown in [Figure 2-29](#) for MWIR QCLs and [Figure 2-30](#) for LWIR QCLs.

2.5.4. The shift of the FSR under RF injection

In this paragraph, we will present the main results of the measurements done with the high-resolution FTIR for the laser at 4.7 and 9 μm . These results are the most relevant one, but same effects were found for different currents and different lasers length. For a better understanding, each measurement is noted as MXX; (lower number corresponds to the measurement without injection).

QCL at 4.7 μm

Figure 2-28 illustrates the measurement realised with the high-resolution FTIR. We recorded the high-resolution spectrum for each measurement. Then the peak position of each mode was compared with the same peak number of the referent spectrum without injection (M1). The variation of the peak position, giving the FSR, was investigated for injection at the extremity of the locking range (M5 and M4), on the beatnote (M2) and out of the locking range (M3) with an RF power of 27 dBm.

Figure 2-28: Illustration of the measurement with RF injection at different frequencies.

For the injection in the locking-range, we made sure that the beatnote was fully injected in the RF signal. However, concerning the injection out of the locking range, we observed

the creation of the sidebands in the RF spectrum. *Figure 2-29* (a) shows the histogram of the repartition of the FSR over the spectrum following the frequency of injection.

Regarding the measurement M2, M5 and M3 for an RF injection at a lower frequency than the beatnote we see the FSR shifting toward the lower frequencies. In the locking range, most of the FSR stay concentrated in a range of 10 MHz, nevertheless for the injection out of the locking range we observe a significant broadening of the histogram. *Figure 2-29* (b) displays the analysis of the variation of the peak position under RF injection compare to the reference value without injection: ΔMX . The injection at a lower frequency than the beatnote will increase the value of the FSR (in cm^{-1}), and thus, display a positive slope corresponding to the difference of frequency between the beatnote and the injection if the beatnote is locked. In the opposite, the injection at a higher frequency will decrease the value of the FSR and then introduce a negative slope. We can noticeably observe the evolution of the peak position which shows the adaptation of the modes to the RF injection over the entire spectrum or a part of the spectrum. The dashed line corresponds to the expected slope calculated in *Figure 2-28* with the value shown in *Table 2-2*.

The modes following the slopes are locked by the injection, the break of the slope may be due to a part of the spectrum which is not locked by the injection or due to the “pulling-effect” of higher order modes as discussed earlier.

Figure 2-29: QCL-4-3-2A at 400 mA M1 without and M2, M3, M4 and M5 with RF injection at 27 dBm. (a) represent the histogram of the FSR for each measurement and (b) the variation of the peak position with the expected slope in dashed line.

The Table 2-2 lists the expected values of the slope corresponding to the ΔMXX in Figure 2-28 and the experimental values given by a linear fit over the stable part of the spectrum for the analysis concerned. The experimental values are in an excellent agreement with the expected value.

Measurements	Slope expected (cm^{-1})	Real slope (cm^{-1})
$\Delta M5$ (extreme low frequency of the range)	$1.9 * 10^{-4}$ (5.8 MHz)	$1.87 * 10^{-4}$ (5.6 MHz)
$\Delta M4$ (extreme high frequency of the range)	$-1.94 * 10^{-4}$ (7.8 MHz)	$-2 * 10^{-4}$ (6 MHz)
$\Delta M3$ (Out of the range)	$-4.3 * 10^{-4}$ (12,8 MHz)	$-4.7 * 10^{-4}$ (14.1 MHz)

Table 2-2: Summary of the experimental and calculated values of the shift of the FSR under microwave modulation for QCL-4-3-2A.

The configuration with RF injection in the locking range exhibit an injection locking over most of the spectrum, the curves show that the FSR is imposed by the injection. As discussed earlier, the resolution is not always limited by the FTIR but can be degraded by the effect of higher order mode which can generate an electrical pulling effect.

QCL at 9 μm

The same measurements have been realised on the 9 μm QCL. *Figure 2-30* (b) show the beatnote and the injection frequencies. As seen explained previously *Figure 2-30* (a) exhibits the histograms showing the variation of the FSR for each measurement while in *Figure 2-30* (c) the variation of the peak position express a slope following the shift of FSR.

Figure 2-30: QCL-9-4-2A at 725 mA M18 without and M19, M20 and M21 with RF injection at 15.8 dBm. (a) represent the histogram of the FSR for each measurement, (b) RF spectrum with the beatnote and the different injection frequencies and (c) the variation of the peak position with the expected slope in dashed line.

Table 2-3 summarises the FSR value expected using Figure 2-30 (b) and the experimental value of the FSR.

Measurements	Slope expected (cm^{-1})	Real slope (cm^{-1})
$\Delta M21$ (extreme low frequency)	$1.63 * 10^{-4}$ (4.9 MHz)	$1.6 * 10^{-4}$ (4.8 MHz)
$\Delta M20$ (extreme high frequency)	$-4.3 * 10^{-5}$ (1.3 MHz)	$-5 * 10^{-5}$ (1.5 MHz)

Table 2-3: Summary of the experimental and calculated values of the shift of the FSR under microwave modulation for QCL-9-4-2A.

Same results have been found for the QCL emitting at 9 and 4.7 μm . The radio-frequency injection enables to tune the FSR over the locking range with a more efficient injection toward the lower frequency of the beating signal as explained previously. The thin beating signal of the free running FP modes and the absence of dispersion over the entire spectrum, under the high-resolution measurements, allow us to show a comb-like operation of our QCLs. Moreover, the injection in the locking range does not introduce GVD and provide enough evidence to show the RF pulling due to the external injection.

Chapter 3 High-speed Quantum

Well Infrared Photodetector

This paragraph will describe the design, process, characterisation and high-speed behaviour of quantum well infrared photodetectors in the MWIR. We will start with the essential parameters for the design of the QWIP active region and the optimisation of the performances considering the applications. Then we will express the fabrication process of the devices, in term of standard QWIP (MESA) and high-speed QWIP which ask more complicated steps. The third part will explain the different setups to study the main parameters, to calibrate and to examine the figure of merits of our device. Finally, the last section will describe and show the high-speed behaviour of our photodetector by direct and indirect methods. The direct method will be done using high-speed QCL in MWIR detailed in Chapter 2, the beatnote and injection-locking of the laser will also be shown in this chapter. Regarding the indirect method, it consists of the rectification methods, realised here using different setups to remove the resonance of the circuits. LWIR QWIP will be treated succinctly here, using standard process.

3.1. Design of the active region

Despite the choice of materials and aluminium fraction, the doping concentration and the QWIP structures are significant for the performances of the device. The upper state in the QW must be precisely positioned to maximise the photocurrent by optimising the escape probability. The structure design we choose here is the bound-to-quasicontinuum where, as we explained in Chapter 1, the upper state is in resonance with the continuum.

3.1.1. Optimisation of the doping concentration

The doping concentration is one of the most critical factors for the performances of QWIPs. Most of the figures of merits are related to the electron density and thus the doping in the well. These dependencies allow us to optimise the performances of the detector considering the temperature of use or the signal to noise ratio. The optimum doping concentration as a function of the temperature operation is shown in Figure 3-1 (a) [38];

- The maximisation of the detectivity will be given by;

$$E_F = 2k_B T, \text{ i.e. } n_{2D} = \frac{2k_B T}{\pi \hbar^2} m^*$$

- The optimisation of the BLIP temperature will be given by;

$$n_{2D} = \frac{k_B T}{\pi \hbar^2} m^*$$

With m^* the effective mass of the electron in $\text{In}_{0.1}\text{Ga}_{0.9}\text{As}$.

Figure 3-1: (a) Optimum doping concentration regarding the detectivity (solid black line) and BLIP temperature (in red dash line). (b) The behaviour of the detectivity as a function of the temperature for different doping concentration (extract from [104]).

A trade-off according to the applications must be done, slight change on the doping concentration can help us to improve the signal to noise ratio (detectivity) or the temperature of use of the detector (BLIP temperature). The influence of the doping concentration on the detectivity showed in the equation (1-40). The absorption coefficient is proportional to the electron density in the QW, the higher the doping concentration, the

higher will be the absorption per wells and therefore, increase the detectivity. The influence of the number of quantum wells on the detectivity come from the fact that the photoconductive gain is inversely proportional of the number of quantum wells, the current noise will decrease with the augmentation of periods and therefore, $D_{BL}^*(T) \propto \sqrt{N_{QW}}$ [149].

3.1.2. Presentation of the active region

The structure presented here is based on 40 periods of strained InGaAs/AlGaAs alloys grown on GaAs substrate. The wafer was grown by molecular beam epitaxy on a GaAs substrate. The active layers are composed of 40 periods of the following sequence: 5 Å In_{0.1}Ga_{0.9}As, 25 Å In_{0.1}Ga_{0.9}As (doped: $5 \times 10^{11} \text{ cm}^{-2}$), 5 Å In_{0.1}Ga_{0.9}As separated by a 400 Å Al_{0.4}Ga_{0.6}As barrier. The 1.78 μm thick active region is cladded between highly-doped GaAs contact layers. This structure is designed as a bound-to-quasicontinuum transition as seen in the simulation in [Figure 3-2 \(b\)](#). We can make the approximation of complete ionisation which means, for p-doped semiconductors, all the impurities have donated an electron. In this case, electron density in the quantum well is equal to the doping concentration and assuming that $E_{21} - E_F \gg k_B T$, we can safely relate the doping density to the Fermi energy by:

$$n_{2D} = N_D = (m^*/\pi\hbar^2)E_F \quad (3-1)$$

In the literature review, empirically and experimentally, the ratio of the effective mass for the ternary composition of In_{0.1}Ga_{0.9}As is $\frac{m^*}{m_0} = 0.058$. We can, therefore, obtain a value of the Fermi energy; $E_F = 20.4 \text{ meV}$ referenced to the first eigenlevel E_1 at 0 K. The equation (3-1) shows the dependence between the Fermi energy with the temperature

through its relationship with the electron density which follows Fermi-Dirac statistic [119]:

$$n_{2D}(T) = \frac{m^*}{\pi \hbar^2} k_B T \ln[1 + \exp\left(\frac{E_F - E_1}{k_B T}\right)] \quad (3-2)$$

When the temperature increases, the electron density decreases following the Fermi Dirac distribution, however, this temperature dependence remains relatively small.

Figure 3-2: Simulation of the energy level in QWs for our structure.

As seen in [Figure 3-2](#) the activation energy corresponds to $E_F - V_B$ the energy required for thermal activation of electrons from the ground state to the continuum. The energy $E_2 - E_1$ corresponds to the optical transition; it is the energy necessary to photoexcite the carriers of the ground state. As we will see later, these two values are directly related to respectively, the dark noise current and the absorption spectra of this QWIP.

3.1.3. Presentation of the QWIP structure

The structure is a bound-to-quasicontinuum structure, and the upper state energy is designed in resonance with the continuum. According to the simulation, the upper state energy is located at about 17 meV from the top of the barrier at 0 V. This energy must remain low to ensure a high probability of escape ($p_e \sim 1$) and therefore, excellent performances of the device.

Figure 3-3: Simulation of the structure with 3 QWs and a bias voltage of 2 V. Relevant parameters are displayed for 0 V.

QWIP are photoconductive devices; a bias voltage is required to allow the electrons to flow from the first contact layer, the emitter, to the second contact layer, the collector. Indeed, the bias voltage will tend the structure according to the difference of potential between the two electrodes. The [Figure 3-3](#) shows the simulation of the structure with a

bias voltage of 2 V. To simplify the approach, we only simulated 3 QWs without using Poisson equations and keeping a homogenous distribution of the electric field in the structure. The fact that the structure is bent decrease the gap between the energy state and the top of the barriers which for 2 V decrease of few meV.

3.2. Fabrication method

The standard fabrication method for the QWIP is not as sophisticated as the QCLs fabrication. However, the process for high-speed QWIPs calls for a more complex architecture due to the relatively small size of the detector and the new contact which is done using air bridge instead of the conventional wire bonding.

3.2.1. Standard MESA fabrication

Figure 3-4 (a) shows SEM pictures of the standard QWIP using relatively large MESA of about 300 μm , Figure 3-4 (b) shows a cleaved MESA which permits us to see the different compositions as described at the beginning of this chapter.

Figure 3-4: (a) SEM pictures of 300 μm diameter MESA, (b) section of the QWIP with different layers indicated on the SEM picture.

The primary steps of fabrication will be detailed in the next section where we will explain the fabrication of high-speed QWIP.

3.2.2. High-speed fabrication

The dimension of the structure will not be explained in this section; they will be calculated and detailed on the high-speed paragraph at the end of this chapter. The fabrication will be illustrated using Scanning Electron Microscope (SEM) pictures from [Figure 3-5](#) to [Figure 3-10](#). Square MESAs were made using dry-etching techniques on an annealed Ge/Au/Ni/Au top contact. 2.08 μm were etched up to reaching the bottom contact using Inductively Coupled Plasma (ICP) etching system as seen in [Figure 3-5](#) (a) and (b) where the different layers of the structure are noticeable. The top contact is annealed at 420° C to diffuse it into the contact layer.

Figure 3-5: (a) Square MESA after dry etching, (b) close look at the etched structure.

The dry etching is stopped at the ground of the MESA which is, as the top contact, a heavy doped GaAs layer corresponding to the second electrode. A second etch using wet-technic is required to isolate the coplanar waveguide from the bottom contact. First, a positive photoresist is deposited to protect the MESA from the wet agent as seen [Figure 3-6 \(a\)](#), then the entire layer of the bottom contact is removed in front of the active region as seen [Figure 3-6 \(b\)](#).

Figure 3-6: (a) Positive photoresist layer to protect the MESA during the wet etching process, (b) bottom contact removed in front of the MESA.

The air-bridge will connect the top contact to the coplanar waveguide. This process is critical because if the bridge is not done correctly and touch the bottom contact, the two contact layers will short-cut the active region. The air-bridge is realised using two different photoresists, a positive one used as a pillow to support the ridge in front of the MESA showed [Figure 3-7 \(a\)](#), and a negative photoresist to do the lift-off after a Ti/Au thick deposition by E-beam technique shown [Figure 3-7 \(b\)](#). The additional bakes of the positive photoresist are essential to soften the angle of the remaining part and to make it strong enough to support the development of the negative photoresist. Therefore, 5 min for each bake from 110 to 150° C every 10° C is done. The E-beam deposition must be done with three different angles (+30, 0 and -30° without rotation) following the direction of the coplanar waveguide to ensure good adhesion between the bridge and the substrate.

Figure 3-7: (a) Positive photoresist used as a pillow to support the air-bridge, (b) a negative photoresist is added to ensure the lift-off process after gold deposition, inset: microscope photo of both photoresists.

After the lift-off, both photoresists are removed, it remains the air-bridge plus the coplanar waveguide seen [Figure 3-8 \(b\)](#). [Figure 3-8 \(a\)](#) shows a zoom-in on the air-bridge passing over the bottom contact. The bridge ensures the link between the top contact of the MESA to the microstrip which is designed to widen to a width adapted to the high-frequency launcher while keeping an impedance of 50Ω .

Figure 3-8: (a) Zoom-in on the active region with the air-bridge “jumping” over the bottom contact, (b) global view of the device with the coplanar waveguide.

At this point, as seen in [Figure 3-9 \(a\)](#), all the devices are linked by the gold deposition, a precise cleavage needs to be done slightly after the start of the coplanar waveguide to isolate them. Finally, we polished the facet at 45° to couple the light, and the sample is bonded to a copper base using a thin layer of InP as seen in [Figure 3-9 \(b\)](#). During the polishing part, the air-bridge is protected using thick positive photoresist.

Figure 3-9: (a) Row of devices with different square size, (b) devices cleaved and bonded to copper-based using InP thin layer, inset: devices after polishing the facet at 45°.

Figure 3-10 shows a comprehensive view of the devices in the electrical and the optical point of view. The top contact is linked to the coplanar waveguide where the signal will be detected using an air-bridge. The bottom contact (the ground) is separated from the coplanar waveguide by a precise gap corresponding to an impedance of $50\ \Omega$ matching with the impedance of the high-frequency components as described in the high-speed paragraph. The incoming light is coupled to the active region through the 45° facet.

Figure 3-10: Global view of the devices with the electrical and the optical interactions, inset: picture of the sample.

In this way, we made samples with different square sizes, [Table 3-1](#) will list the sample used in this thesis.

3.2.3. List of samples

Sample name	Dimension (μm^2)	Composition	Wavelength absorption (μm)	Number of quantum well
4-4B	25x25	$In_{0.1}Ga_{0.9}As/Al_{0.4}Ga_{0.6}As$	4.9	40
4-4D	15x15			
4-4F	25x25			
4-7C	20x20			
9-1A	$\pi 150^2$	$Al_{0.35}Ga_{0.75}As/GaAs$	9	10

Table 3-1: List of samples available.

The optical characterisations have been realised on both wavelengths. However, the high-frequency responses have been measured only on the sample at 4.9 μm . We will present the results of the QWIP at 9 μm in the paragraph 3.3.4. LWIR QWIP at 9 μm .

3.3. Optical characterisation

The optical characterisation has been done on the sample 4-4D and permits to highlight the different contributions to the current. As described in the first chapter, at low temperature (below 70 K for MIR QWIP) the main contribution is due to the background radiation (see black-body radiation Chapter 1) which photoexcite the electrons from the ground state to the continuum. At higher temperature, the dominant noise factor becomes the dark current which increases exponentially with the temperature due to the thermal activation which excites the electrons from the ground state to the continuum.

3.3.1. Dark current density and activation energy

We positioned the QWIP on a cold finger in a cryostat which is pumped and cooled down at cryogenic temperature. We controlled the temperature by using a Peltier in the cryostat.

Figure 3-11: (a) Setup for dark current measurements with cryo-shield at low temperature and (b) the setup for background current measurements depending on the FOV.

To remove the background current and study only the dark current, we use a cryo-shield cooled at low temperature as illustrated in [Figure 3-11](#) (a). This assembly will act as black-body radiation at low temperatures and therefore, according to Planck-law seen in Chapter 1 drastically reduce the photoexcited electrons compared to the room-temperature radiation. As described in Chapter 1 the dark current density can be written as:

$$J_{dark} = J_0 T \exp(-E_{act}/k_B T) \quad (3-3)$$

With $J_0(T) = \frac{ev(T)\tau_{capt}}{\tau_{scatt}} \frac{m^*}{\pi\hbar^2 L_p} k_B$. We clearly see its exponential behaviour due to the temperature. The E_{act} has been described in the first chapter and will be measured by fitting the dark current using the equation (3-3).

Figure 3-12: (a) Evolution of the current as a function of the temperature fitted by the exponential curve, inset: Activation energy as a function of the

*bias voltage, (b) dark current versus the voltage for different temperature
(sample 4D with 15 μm MESA).*

Figure 3-12 (b) expresses the dark current versus the bias voltage for temperature going from 80 K to 300 K. Figure 3-12 (a) shows the study of the dark current as a function of the temperature, with exponential fit according to equation (3-3) to extract the activation energy. The inset of Figure 3-12 (a) represents the activation energy as a function of the bias voltage, as explained earlier, the increase of the bias voltage will bend the structure and reduce the activation energy by reducing the gap between E_F and the top of the barrier. The fitting curve gives the activation energy at low bias voltages of about 263.2 meV, which is in excellent agreement with the simulation of the structure.

3.3.2. Background-limited current and BLIP temperature

As described in the first chapter the background current is the sum of the photoexcited electrons due to the background radiation and the dark current.

$$I_{background} = I_{ph,bg} + I_{dark} \quad (3-4)$$

Figure 3-11 (b) shows the setup for background current measurements. We removed the cryo-shield, and the detectors see the black-body radiation of the surrounded objects at RT, in this situation the Field Of View (FOV) is equal to 2π . In order to have the background photocurrent as the limiting factor of the detector, it has to be cooled at low temperatures to remove the dark current. Therefore, we can investigate the ratio between the dark current previously recorded using the cryo-shield and the background current of the setup without the cryo-shield. Figure 3-13 (a) shows the I-V curve in dark and background conditions at 110 K and Figure 3-13 (b) records the ratio between the two

currents. While increasing the temperature, the ratio tends to 1 which correspond to the regime where the dark current becomes the limiting factor. As described earlier the BLIP temperature, is archived when the photoexcited electrons are equal to the thermally excited carriers. Therefore, the BLIP temperature corresponds to $I_{ph,bg} = I_{dark} \equiv \frac{I_{background}}{I_{dark}} = 2$ shown in Figure 3-13 (b) by dashed purple line.

Figure 3-13: (a) Dark and background currents as a function of the voltage for 110 K (b) ratio between background current over the dark current versus the temperature for +/- 2 V.

BLIP condition is the temperature where $I_{ph,bg} > I_{dark}$ indicated in purple in [Figure 3-13](#) (b). For the sample 4-4D, the BLIP temperature is around 130 K, up to this temperature, the current is limited by the background radiation, and beyond this temperature, the background current is becoming negligible compared to the dark current which makes the BLIP condition the privileged regime for imagery applications.

3.3.3. Spectrum as a function of the bias voltage and the temperature

The absorption spectrum is studied using the internal sources of the Fourier Transform Infrared spectroscopy (FTIR) which is a black-body at about 1200° C. The setup is illustrated in [Figure 3-14](#).

Figure 3-14: Illustration of the setup for spectrum analysis.

The sample is mounted on a cryostat and cooled down at cryogenic temperature. The spectra for different temperatures and bias voltages have been measured by imaging the globar on the detector using lenses. To remove the background noise, we optical chopped the sources at 500 Hz and use a lock-in technic [150]. The detector signal was pre-amplified by a low-noise current amplifier DLPCA-200 and sent to the lock-in amplifier, Stanford research SR830, and finally, the output is sent to the external port of the FTIR as shown in [Figure 3-14](#).

Figure 3-15: (a) Spectra for different bias voltages at 80 K, inset: the relative amplitude of the spectra as a function of the bias voltage.

Figure 3-15 (a) shows the normalised spectra for different bias voltage at a temperature of 80 K. The amplitude of the spectra is presented in the inset of Figure 3-15 (b) and follow the evolution of the photocurrent versus the bias voltage. The peak absorption is at about 250 meV which corresponds to the energy gap $\Delta E = E_2 - E_1$ simulated in the previous paragraph. The temperature of the sample is ensured by a Peltier and feedback loop placed in the cryostat.

Figure 3-16: (a) Spectra for different temperatures, (b) red-shift of the peak position versus the temperature and (c) FWHM of the Lorentz fit as a function of the temperature.

The behaviour of the absorption spectra with the temperature gives a good idea of the performance of the devices. After the calibration performed below, considering the dimension of the detector and the spectral response of the detector, we calculated the optical input on the QWIP of 52 nW. As shown in [Figure 3-16](#) (a) spectra are observed up to room temperature using a low input power, this is substantial evidence for room temperature behaviour of our devices. [Figure 3-16](#) (b) shows the central wavelength of the QWIP slightly shifting toward lower energy with increasing the temperature (from 254.17 meV at 80K to 249.20 meV at 250 K) which is due to the depolarization effect coming from Coulomb interaction with the oscillating electrons [38]. While the linewidth

is stable and starts to increase from 130 K (from 27.03 meV up to 130 K to 33.48 meV at 250 K) which is mainly due to electron-phonon interaction which broadens the spectrum exhibited in [Figure 3-16](#) (c).

For the spectrum at 300 K, the signal read on the Keithley is about 5 nA which corresponds to a power of about 52 nW with a responsivity of 0.1 A/W. The noise current is written as $I_{noise} = \sqrt{2egI_{dark}\Delta\nu}$.

At a bias voltage of -5 V the photoconductive gain and dark current of our devices are respectively 0.5 and 3×10^{-4} A. Therefore, with an integration time of 100 ms the noise current is 2.2×10^{-11} A, the ratio between the signal and the noise is more than 20, which is enough to generate a spectrum at room temperature.

3.3.4. LWIR QWIP at 9 μm

Here we present the results of a QWIP designed in the second atmospheric windows, at 9 μm . Here, we present a structure based on 10 periods of AlGaAs/GaAs alloys grown on GaAs substrate. The wafer was grown by molecular beam epitaxy on a GaAs substrate. The active layers are composed of 10 periods of the following sequence: 6 Å GaAs, 40 Å GaAs (doped: $7 \times 10^{11} \text{ cm}^{-2}$), 6 Å GaAs separated by a 350 Å $\text{Al}_{0.25}\text{Ga}_{0.75}\text{As}$ barrier. This structure is displayed with a bias voltage of 2 V and 0 V (in inset) in [Figure 3-17](#) (a). We accomplished same experiments to study the background current and plot it as a function of the bias voltage for different temperatures in [Figure 3-17](#) (b), the inset at the bottom left, is the activation energy and bottom right the photocurrent at 77 K vs the voltage applied.

Figure 3-17: (a) Band structure of LWIR QWIP with a bias voltage of 2 V, inset: QW at 0 V. (b) Background current versus the bias voltage for different temperatures, inset: (bottom left) the activation energy and (bottom right) photocurrent at 77 K vs the bias voltage.

As for the MWIR QWIP, we designed a bound-to-quasicontinuum structure where the upper state is in resonance with the continuum. We designed this QWIP with a relatively high doping concentration to increase the temperature operation and therefore, as we can

see in Figure 3-17 (b), and in the insets, the low activation energy leads to a high current noise due to the thermal activation. In the opposite with our QWIP in the MWIR, the photocurrent shows a critical negative differential conductivity due to the decrease of the drift velocity of the electrons in GaAs at a particular voltage as discussed in the paragraph **3.4.5. Performance at high bias voltage.** Figure 3-18 presents the spectra behaviour versus the voltage in (a) and versus the temperature (b).

Figure 3-18: (a) Spectra for different applied voltages, inset: the amplitude of the spectra as a function of the voltage, (b) spectra for several temperatures at -2 V.

The amplitude of the spectra versus the voltage follows the photocurrent behaviour and displays a peak position at about 129,4 meV, and an FWHM of 27 meV at -0.8 V and 77 K. The spectra have been done using the FTIR in rapid scan mode, and as expected we have spectra up to relatively high temperature.

3.4. Calibration and figure of merit

We are not able to fully picture the performances of our devices without calibrating it with a well-known system. We calibrated the sample 4-4B using a commercial MCT already calibrated. To calculate the photoconductive gain and other parameters such as the responsivity and the detectivity, we need to know the amount of power which give the photocurrent induced by an incoming IR light.

3.4.1. Blackbody calibration at 1000° C and photocurrent

The incident sources used for the calibration is a 0.05 inch aperture blackbody at 1000° C (OL 480 Blackbody calibration standard composed of an internal conical cavity which is electrically heated up). This IR broadband source is 1:1 imaging on the detector using two lenses, the image of the blackbody on the QWIP will be much larger than the active area of the detector as illustrated in [Figure 3-19](#).

Figure 3-19: Illustration of the setup to investigate the photocurrent and other important parameters.

We applied the bias voltage of the QWIP using a 2450 source Meter KEITHLEY, and the photocurrent is collected by the lock-in amplifier through a Shunt resistance placed in

series with the QWIP. The values of the resistance are chosen to be comparable to the resistance of the QWIP at the specific temperature range.

Figure 3-20: (a) Experimental electrical circuit, (b) equivalent electrical circuit using the photoconductive detector as a current generator and (c) Norton equivalent circuit.

As we explained earlier, the lock-in technique is a powerful technique to remove the noise by measuring the signal at a specific frequency which corresponds to the chopping frequency. Therefore, the measured read is the root-mean-square value at the first harmonics of the square wave signal decomposed into a Fourier series; it is translated into a $\pi/\sqrt{2}$ factor in the calculation of the current seen in the equation (3-5). Moreover, the cut-off frequency of the AC lock-in entrance is an RLC circuit, and according to the resistance used, is in the order of MHz. Therefore, our AC photocurrent due to the optical chopping at 500 Hz is not attenuated by the low-pass filter. Figure 3-20 (a) and (b) show the equivalent electrical circuit with the QWIP seen as a current generator and Figure 3-20 (c) show the equivalent circuit using Norton theorem. The lock-in will see only the

AC current which is proportional to the photocurrent and the current read on the DC source (Keithley) is the total current, I_{tot} , flowing through the electrical circuit which will help us to calculate the differential resistance of the QWIP.

Therefore, we have:

$$I_{photo} = \frac{\pi}{\sqrt{2}} \frac{V_{Lock-in}}{R_{eq}} \quad (3-5)$$

With $R_{eq} = \frac{R_{QWIP} \cdot R_{SHUNT}}{R_{QWIP} + R_{SHUNT}}$, QWIP differential resistance is calculated by $R_{QWIP} = \frac{dV_{QWIP}}{dI_{tot}}$. The difference of potential at the terminal of the QWIP is $V_{QWIP} = V_{in} - r_s I_{tot}$ with V_{IN} the voltage of the DC source.

The shunt resistance R_{SHUNT} must be chosen comparable to the QWIP resistance in the specific temperature range. Thus, we used a resistance of 22 k Ω below 200 K and 1 k Ω for higher temperature due to the resistance of the QWIP at room temperature which is about few k Ω . If the resistance is too high, the photocurrent shows higher noise due to the smaller current sent to the lock-in as we can see in [Figure 3-21](#).

Figure 3-21: Comparison of the photocurrent extracted from two different shunt resistances.

Using this method, we measured the photocurrent of our devices, plotted in log scale, as a function of the bias voltage for different temperatures up to RT seen [Figure 3-23](#). As described in the first chapter, the photocurrent increases with the bias voltage; a theoretical optimal value can be calculated, considering, bent of the structure due to the electric field, the transition energy for a bound-to-quasicontinuum transition and the number of wells such as:

$$V_{opt} = \frac{E_{12} * N_{QW}}{e} \quad (3-6)$$

For $E_{12} = 250 \text{ meV}$ and 40 quantum wells, the optimum voltage is about 10 V. Indeed, our photocurrent increase up to 10 V as seen in this experiment, but further experiments will show an increase of the photocurrent up to 24 V which is explained by the absence of negative differential conductivity in AlGaAs with Al fraction superior to 0.35 and a relatively strong non-homogeneous electric field in the structure. This behaviour is not

always observed in the QWIP. However, in the MWIR region, some works show non-usual behaviour which for our case will be explained in detail in paragraph 3.4.6.

Discussion about the increase of the photocurrent with the temperature.

3.4.2. Responsivity

To analyse the responsivity of our devices, the optical input power is calculated using a calibrated MCT detector with the same configuration. We used a Teledyne Judson J15D22 MCT at the same place than the QWIP with a tabulated responsivity of $R_p = 30 \text{ V/W}$ at low frequency and a detector surface of $A_{MCT} = 2 \text{ mm}^2$. The signal measured by the MCT is internally amplified by a gain $G = 100$.

The photocurrent of the MCT photo-generated by the black-body radiation is:

$$V_{MCT} = V_{lock-in} \frac{\pi}{\sqrt{2}} \frac{1}{G} \quad (3-7)$$

We can calculate the optical power using the responsivity of the MCT $P_e^{MCT} = \frac{V_{MCT}}{R_p}$. The optical power shining on the QWIP can be written as:

$$P_E^{QWIP} = P_E^{MCT} \frac{A_{QWIP} \Sigma_{QWIP}}{A_{MCT} \Sigma_{MCT}} \quad (3-8)$$

With $A_{QWIP} = d^2 \cos(\theta)$ the detector geometry with $d = 25 \text{ }\mu\text{m}$. $\frac{\Sigma_{QWIP}}{\Sigma_{MCT}}$ represents the ratio between the spectral response of the QWIP over the one of the MCT. The spectral responses have to take into account the temperature of the blackbody radiation used for the photocurrents and the spectra. This calculation is shown in [Figure 3-22](#).

Figure 3-22: Curves used to calculate the spectral response of the QWIP and the MCT. The purple and red areas represent the integration of the spectral response.

We calculated the spectral response of $\Sigma_{QWIP} = 6.5 * 10^{-21} J$ for the QWIP and $\Sigma_{QWIP} = 2.3 * 10^{-20} J$ for the MCT, the ratio between the two spectral responses is $\frac{\Sigma_{QWIP}}{\Sigma_{MCT}} = 0.28$.

Due to the [1:1] imaging of the black-body source we are in the configuration where the image of the sources is smaller than the active region of the MCT, thus, $A_{QWIP} < A_{source} < A_{MCT}$. Table 3-2 summarises the different parameters which led to the power on the QWIP.

Aperture (inch)	A_{source} (mm^2)	$V_{lock-in}$ (V)	P_E^{MCT}	A_{QWIP} (μm^2)	P_E^{QWIP}
0.0125	0.07917	0.2345	0.17 mW	328.3	166 nW
0.025	0.31669	0.957	0,7 mW		169 nW
0.05	1.26677	3.5866	2.6 mW		158 nW

Table 3-2: Different parameters used to calculate the power on the QWIP.

We can see that in the configuration where $A_{source} < A_{MCT}$, the power is strongly dependent on the aperture of the source. This effect is largely reduced when $A_{QWIP} < A_{source}$. The optical power shining on the QWIP allows us to calculate the responsivity of our device such as:

$$R_{QWIP} = \frac{I_{photo}}{P_E^{QWIP}} \quad (3-9)$$

Figure 3-23 displays the responsivity vs the bias voltage for different temperature.

3.4.3. Photoconductive gain

The responsivity along with other intrinsic parameters of our QWIP allow us to calculate the photoconductive gain which is given by the equation (1-41) where:

$$g_{photo} = \frac{RE_{12}}{e\eta} \quad (3-10)$$

with $\eta \propto N$ the absorption coefficient and the QWIP responsivity $R_{QWIP} = t\xi R$ which consider, ξ the polarisation coefficient for the intersubband structure due to epitaxy growth direction and t is the transmission coefficient of the GaAs substrate at $4.9 \mu m$, in our case is $t = 0.71$.

$$g_{photo} = \frac{R_{QWIP} E_{12}}{t \xi e \eta} \quad (3-11)$$

The polarisation selection rules for our geometry based on a facet at 45° allow the absorption of only about 50% of unpolarized light, thus; $\xi = 0.5$. η . The absorption coefficient for an IR optical beam with a propagating angle of θ with the growth axis of a structure composed of N_{QW} QWs and considering the peak transitions is defined as [38]:

$$\begin{aligned} \eta(\omega) &= N_{QW} \eta_1 \\ &= N_{QW} \frac{e^2 \hbar}{2 \epsilon_0 n_r m^* c} \frac{\sin^2 \theta}{\cos \theta} n_{2D} f_{12} \frac{\Gamma}{(E_2 - E_1 - \hbar \omega)^2 + \Gamma^2} \end{aligned} \quad (3-12)$$

Due to the intersubband properties, using the peak transition defined by E_{21} is a good approximation. f_{12} is the oscillator strength of the peak transition, from the ground state to the upper-state, is $f_{12} = 0.96$, ϵ_0 the vacuum permittivity, $n_r = 3.6$ the refractive index in $\text{In}_{0.1}\text{Ga}_{0.9}\text{As}$, $m^* = 5.35 * 10^{-32} \text{ kg}$ the effective mass of the $\text{In}_{0.1}\text{Ga}_{0.9}\text{As}$, and Γ the FWHM with the spectrum fitted by a Lorentz (ISB transition).

Considering, the resonance absorption where $E_2 - E_1 - \hbar \omega = 0$, we calculate:

$$\eta(\omega) = N_{QW} \eta_1 = 40 * 0.205 \% = 8.2 \%$$

We can see that the absorption is directly proportional to the doping concentration and the number of quantum wells. By using higher doping concentration and by using a large number of quantum wells we can reach very high absorption of around 100 %.

Figure 3-23: (a) Photocurrent, responsivity and photoconductive gain as a function of the bias voltage from 80 K to 310 K and (b) variation of the parameters with the temperature for several bias voltages.

Figure 3-23 (a) displays the central parameters of the QWIP described earlier. These figures allow us to see the evolution of the photoconductive gain, the responsivity and the photocurrent as a function of the voltage and the temperature for several temperatures (a) and bias voltages (b). The data show an exponential increase with the temperature; we

will explain this behaviour in detail in the paragraph **3.4.6. Discussion about the increase of the photocurrent with the temperature.**

3.4.4. Detectivity and NEP

These factors allow us to calculate the leading figure of merits of photoconductive devices. The background-limited detectivity define as:

$$D_{BL}^* = \frac{R\sqrt{A}}{\sqrt{4egI_{background}}} \quad (3-13)$$

With $I_{background} = I_{ph,bg} + I_{dark}$ the current measured at each temperature and voltage.

The background-limited NEP is defined as:

$$NEP_{BL} = \frac{\sqrt{A}}{D_{BL}^*} \quad (3-14)$$

We plotted these two parameters in [Figure 3-24](#). We could comment that the divergence of the detectivity at low bias voltage is due to the Johnson-Nyquist current noise which is not anymore negligible compared to the other current sources and must be taken into consideration.

Figure 3-24: (a) Detectivity as a function of the bias voltages for different temperatures, (b) detectivity and NEP versus the temperatures at -5, -7 and -8 V.

In [Figure 3-24](#) (b) we can see the different regions theoretically explained in Chapter 1, the plateau of $5 \times 10^{10} \text{ cm}\sqrt{\text{Hz}}\text{W}^{-1}$ at low temperature where the current is limited by the photoexcited electrons from the background and the second regimes where the current is mostly dominated by the dark current. This value is near to the ideal photoconductor limit of $2 \times 10^{11} \text{ cm}\sqrt{\text{Hz}}\text{W}^{-1}$.

3.4.5. Performance at high bias voltages

High-voltage photocurrent has been performed using the internal source of the FTIR. As we saw previously, this source introduce an optical power of about 50 nW on the QWIP.

Figure 3-25: (a) Background current as a function of the electric field at 80 K. Inset: total current under dark and illuminated conditions as a function of the bias voltage for the different incident optical powers, after [38], [151]. (b) Photocurrent, responsivity and photoconductive gain versus the voltage at 80 K.

Figure 3-25 (a) shows an important increase of the dark current after 100 kV/cm (about 17 V). High electric field measurements allow us to highlight the different transport effect of the carrier in the QWIP under illumination already well described in GaAs/AlGaAs structure [38], [151]. We can identify different transport effect while increasing the voltage, at high electric fields the limitation is due to the increase of the dark current as seen in Figure 3-25 (a) after 100 kV/cm. The equation (1-25) shows the dependence of the dark current with n_{3D} which increases with the reduction of the effective barrier due to the electric field. The prominent plateau at relatively low bias voltage observed in Figure 3-25 and in the inset, appears only under illumination and shows a limitation by the electron mobility. We can notice that the prominent plateau in the inset is flat and can exhibit Negative Differential Conductivity (NDC) relatively common for structure based on GaAs/AlGaAs but our device shows a plateau with a positive slope identical to the photocurrent in Figure 3-25 (b). This difference will be the subject of the next paragraph.

Figure 3-26: Photocurrent versus the electric field with a linear fit represented by the red dashed line.

At low electric fields, the dependence of the photocurrent with the applied electric field is linear for BCQ and BC structure as we can see in [Figure 3-26](#). This behaviour is due to the linear increase of the drift velocity with the bias voltage, and it corresponds to the maximum point calculated at the equation (3-6) with $V_{max} = \frac{E_{12} * N_{QW}}{e} = \mp 10 \text{ V}$ which is the linear part of our photocurrent. For electric field higher than 50 kV/cm strong nonlinearity in the photocurrent has been observed in III-V bulk semiconductors such as GaAs, corresponding to a negative differential conductivity so-called Gunn-effect. In GaAs, experiments show a decrease of the photocurrent after the voltage threshold given by the Γ -L intervalley energy. Indeed, high energy is given to the electrons, due to the electric field, induce a scattering effect from Γ -minimum into the L- or X- minima of the conduction band. This is translated by an increase of the effective mass of the electrons and therefore lower mobility and velocity [151], [152]. S. Hava and M. Auslender realised relevant works on the drift velocity relation in $\text{Ga}_x\text{Al}_{1-x}\text{As}$ as a function as Aluminium

fraction. They highlighted the Gunn effect for the x fraction up to 0.3 from relatively low electric fields (5 kV/cm). Regarding the Al fraction $x \geq 0.4$, no negative differential mobility has been found up to about 200 kV/cm [153] and Monte Carlo simulation by Brennan et al. suggest no velocity saturation up to 400 kV/cm [154].

These analyses and simulation are consistent with our experiments which show an increase of the photocurrent up to high electric fields, indeed [Figure 3-25](#), and [Figure 3-26](#) does not show NDC up to 150 kV/cm, i.e. 25 V for our sample. We may add here that the 150 kV/cm may not be homogenous over the structure, and a significant part of it can be concentrated in the first barriers, we will explain more in detail this phenomenon in the next paragraph.

3.4.6. Discussion about the increase of the photocurrent with the temperature

We highlighted the increase of the photocurrent with the temperature using different setups and different samples. [Figure 3-27](#) (a) presents the photocurrent introduced by a QCL, emitting in the absorption spectrum of the QWIP, at 80 K and 300 K and that for different laser output powers on the sample 4-4D seen [Figure 3-27](#) (b).

Figure 3-27: (a) Photocurrent as a function of the bias voltage for different laser powers at 80 and 300 K, inset: linearity between the photocurrent generated and incoming power (b) the absorption spectrum of the QWIP at 80 K and -8 V with QCL spectrum emission.

These measurements have been done using the lock-in amplifier to detect only the photocurrent. As shown on the previous results with the blackbody sources, an apparent gap is visible between the photocurrent at 80 K and 300 K for the same incoming light,

the inset shows the linearity between the absorption of the QWIP and the incoming IR light. The thermal evolution of the photocurrent with the temperature, have an exponential behaviour. Fitting its evolutions allow us to release this activation energy of the photocurrent link to this phenomenon.

Figure 3-28: Evolution of the photocurrent, responsivity and photoconductive gain as a function of the temperatures up to 310 K for different bias voltages. The solid lines are the fits corresponding to each voltage.

These increases follow an exponential behaviour with the temperature and match correctly to the exponential fits $(b + J_0 * T * e^{\frac{-E_a}{k_B * T}})$. They express a thermal activation which corresponds to energies close to the first contact barrier. The factor b reprints the background photocurrent at low temperature and J_0 the exponential factor described previously. Due to the low number of points, here the uncertainty on J_0 is high and therefore, the one on the activation energy as well.

Figure 3-29: (a) Factor b of the exponential fits corresponding to the background current at low temperature, (b) the activation energy of the photocurrent as a function of the voltage, inset: activation energy of the dark current.

Figure 3-29 reveals in (a) the factor b of the fits which correspond to the photocurrent at low temperature and in (b) the activation energy of the photocurrent as a function of the bias voltage. This parameter at low bias voltage corresponds to about 310 meV which matches to the contact barrier height, and at higher bias voltages, this parameter decreases quickly. We can observe two regimes already described in Figure 1-22 and Figure 1-23.

- At low bias voltages, the activation energy remains relatively constant and start to decrease slowly up to 6 V. Where the transition of electrons from the contact to the structure is governed by trapezoidal regime and electron “jumping” over the barrier.
- At high bias voltages, the activation energy drops faster after 6 V. This regime is governed by triangular tunnelling regimes where the barrier thickness is greatly reduced by the strong electric field.

This behaviour is not comparable to the activation energy of to the dark current which tends to decrease with a high slope at low electric fields due to the diminution of the gap between the Fermi energy and the top of the barrier seen in the inset of [Figure 3-29](#) (b). The activation energy of the dark current is corresponding to energy which takes place in the QWs.

The activation energy of the photocurrent takes place in the contact. [Figure 3-31](#) (left) shows the effect of the electric field;

- At low bias voltages the activation energy match to contact barrier and stay relatively constant because the electric field does not bend the contact.
- At high bias voltages, the rapid decrease of the activation energy corresponds to the energy required to make the electron seeing a tunnelling transmission near unity as explained in the paragraph of the tunnelling effect in Chapter 1.

Two factors are playing essential roles in the capacity of the emitter to provide electrons to the structure; the electric field explained earlier in [Figure 3-31](#) (left) and the temperature which enhances the electric field effect and delocalized a portion of the carriers at higher energy levels.

Considering the thickness of the contact layer, we can use calculation for bulk GaAs. The Al fraction of 0.4 gives a barrier height of 316 meV which is consistent with the activation energy measured in [Figure 3-29](#). In comparison, the barrier height in our 9 μm QWIP with an Al fraction of 0.25 is 197.5 meV and 50 nm thick. The electron density is calculated using:

$$N_c = 2 \left[\frac{2\pi m^* k_B T}{h^2} \right]^{3/2} \quad (3-15)$$

With N_c the effective density of state in the conduction band which is equal to $N_c = 5.7 * 10^{17} \text{ cm}^{-3}$ in GaAs. The Electron density and effective density of state are linked with:

$$n = n_{2D} = N_c \exp\left(\frac{E_F - E_c}{k_B T}\right) \quad (3-16)$$

The electrons density for an n-type semiconductor is equal to the doping concentration which is $n_{2D} = 1 * 10^{18} \text{ cm}^{-3}$ in the top contact layer. This formula allows us to calculate the Fermi energy as a function of the temperature. The Fermi Dirac distribution gives the probability of occupation of a vacant state by an electron according to the thermal energy given by $k_B T$ such as:

$$P(E, T) = \frac{1}{1 + \exp\left(\frac{E - E_F}{k_B T}\right)} \quad (3-17)$$

The probability of occupation for an energy E and a temperature T is shown in [Figure 3-30 \(a\)](#). [Figure 3-30 \(b\)](#) represents the distribution of the electron density in log scale for 80, 200 and 300 K. We can see the spreading of the electrons due to the thermal energy in the system. Therefore, if we take a limit of $0.01 * n_{2D} = 1 * 10^{16} \text{ cm}^{-3}$, these electrons see activation energy of about 180 meV.

Figure 3-30: Simulation for the top contact layer, (a) probability of occupation by an electron versus the energy in the quantum well, (b) electrons density in log scale as a function of the energy in the QW. The Fermi energies for 80 and 300 K and the quantum well are indicated by respectively, black, red dash line and full black line.

The effect of the temperature is seen in [Figure 3-30](#) where at higher temperature electrons are delocalized in the higher energies of this bulk GaAs contact layer. This calculation can be underestimated due to the annealing of the top contact defused into the top layer.

The two effects of the increase of the photocurrent described earlier are summarised in a comprehensive illustration in [Figure 3-31](#) which highlights the behaviour of the electric field (a, b) and temperature (c, d) on the efficiency for the contact to provide electrons to the structure.

Figure 3-31: Illustration of the influence, on capability for the contact to provide electrons to the structure, of; (a, b) the electric field supported by the activation energy versus the bias voltage and (c, d) the temperature reinforced by the increase of the photocurrent with the current.

The higher part of the figure explains the contribution of the electric field supported by the curve of the activation energy of the photocurrent in the inset. The lower part shows the effect of the temperature reinforced by the evolution of the photocurrent with the temperature. The height of the first barrier after the contact blocks the dark current that refills the wells that have been emptied by the incoming photons. At low temperatures,

the rate of refill is lower than that the rate of depopulation of the well and therefore, a space charge effect, due to the excess of positive charge in the QWs strongly bend the first barrier and allows to drive more current as shown in the upper right part of *Figure 3-32*. This inhomogeneous repartition of charges, however, has the effect of creating a strong inhomogeneity in the applied bias that drops almost all on the first few QWs, while leaving the rest of the structure with much lower average electric field [127]–[130], [155] as illustrated *Figure 3-32*. Therefore, this configuration does not allow an optimised operation of the entire structure.

Figure 3-32: Left; the activation energy of the photocurrent as a function of the low bias voltage in yellow and high bias voltage in purple, the right part illustrate the effect on the QWIP structure for low (upper part) and high (lower part) bias voltages.

This phenomenon can be accentuated by the impact ionisation which will increase the depopulation of the firsts QWs due to the high energy provided to the electrons [156]. When the temperature increases, the broadening of the Fermi level allows electrons in the contact to occupy higher energies and therefore, raises the probability of i) thermionic current as the number of electrons able to “jump” above the barrier is enormously increased [155] and ii) tunnel current as the effective barrier traverse is highly reduced. The ease of refill due to the temperature leads to a more homogenous repartition of the electric field across the structure at high temperature as shown in [Figure 3-33](#).

Figure 3-33: Illustration of the influence of the temperature on the QWIP structure.

This phenomenon is more significant for QWIPs in MWIR [155], indeed in LWIR, the height of the first barrier is lower and therefore, the electrons from the contact can more easily reach the structure. The extra electric field concentrated at the contact barrier need to be stronger for QWIP in the MWIR which leave a relatively low electric field for the rest of the structure [128]. This phenomenon adding to the absence of NDC for AlGaAs with Al fraction > 0.4 can explain why the responsivity of our devices continues to improve even with a large bias voltage of 25 V shown in [Figure 3-25](#) and [Figure 3-26](#), while is it not the case for LWIR QWIP seen in inset of [Figure 3-17](#) (b).

3.5. High-speed behaviour

The frequency dependence of the QWIP is due to two principal factors as describe [27], [157] :

- Photoconductive lifetime: $\alpha(\omega) = \frac{1}{1+(\omega\tau)^2}$ with τ the excited electron lifetime already describes in the first chapter.
- The electrical response of the circuit: $\beta(\omega) = \frac{1}{(1-\omega^2 LC_D)^2 + \omega^2 (R_L C_D + \frac{L}{R_L})^2}$ with L the wire bound inductance and C_D the QWIP capacitance.

The intrinsic response time of the detector is limited by the smaller value of the photoelectron lifetime or the transit time. For a relatively large number of QWs, the total transit time is higher than the carrier lifetime and, due to the properties of ISB, the electron lifetime is in the order of magnitude of about 5 ps [27], [29]. Therefore, the intrinsic lifetime limited cut-off frequency is given by:

$$f_{max} = \frac{1}{2\pi\tau} \approx 32 \text{ GHz} \quad (3-18)$$

This last is an inherent characteristic that allows a new range of applications including heterodyne detection [29], [109], [117], characterisation of ultrafast laser pulses [61] and wideband communication [158]. The electrical response is generally limited by the electrical properties and the geometry of the device. We will, consequently, develop an electrical circuit with a cut-off frequency higher than the intrinsic limitation, i.e. which the only limiting factor will be the carrier limitation. Thus, in a case of a low-pass electrical circuit of the first order (as typical RC circuit) at a frequency higher than f_{max} the frequency response will be proportional to $1/\omega^2$.

3.5.1. Electrical circuit

The equivalent circuits for high frequency considering the QWIP as a current generator is represented in [Figure 3-34](#) with (right) and without (left) air-bridge.

Figure 3-34: Equivalent circuits considering the high-frequency behaviour with (right) and without (left) air-bridge.

Where C_D is the device capacitance, R_C the access resistance of the coplanar line which is designed to be 50 Ohm to match the impedance of the system load R_L and L , the parasite inductance which is negligible for an air-bridge design. The differential resistance of the QWIP is negligible compared to the load resistance, therefore, we will not take it into account. Any wire in the circuit will introduce an inductance which will profoundly reduce the cut-off frequency of the electrical circuit; the first step is to reduce the parasitic inductance by avoiding any wire bonding and using an air-bridge connected to a 50 Ω microstrip line. By doing so, the electrical response is not anymore, an RLC filter but become an RC filter with a cut-off frequency at -3 dBm of $\frac{1}{2\pi R_L C}$.

To see the influence of the gold wire we can calculate the inductance using:

$$L = \frac{\mu_0 l}{2\pi} \left[\ln\left(\frac{2 * l}{r}\right) - 1 \right] \quad (3-19)$$

With l the length and r the radius of the gold wire. $\mu_0 = 4\pi * 10^{-7} \text{ H/m}$ is the magnetic constant. Using typical values of a wire bonding; whit $l = 3 \text{ mm}$ and $r = 12.5 \text{ }\mu\text{m}$. The inductance introduced by this gold wire is about $L = 3 \text{ nH}$. The influence of the inductance gave by standard wire bonding are shown in [Figure 3-35](#) (a). Moreover, a wire

bonding design would introduce large losses due to the impedance mismatch with the hardware.

The second parameter is to decrease the capacitance of the QWIP which is linked to the geometry of the structure and its composition. The capacitance introduced by the active region can be calculated using the parallel-plate approximation:

$$C = \frac{\epsilon_0 \epsilon_r * A_{QWIP}}{d} \quad (3-20)$$

With $\epsilon_0 = 8.85 * 10^{-12} F.m^{-1}$ the vacuum permittivity, the relative permittivity AlGaAs/InGaAs is $\epsilon_r \approx 12.9$, A_{QWIP} the dimension of the sample and d its thickness which depends on the number of QW.

Figure 3-35: (a) Influence of the wire bonding on the cut-off frequency at -3 dBm as a function of devices geometry and (b) the influence of the number of quantum wells on the cut-off frequency at -3 dBm.

The influence of the capacitance, which is inversely proportional to the number of quantum wells is shown in [Figure 3-35](#) (b), and [Table 3-3](#) summarised the data for our sample with 40 QW.

Square MESA (μm)	Capacitance for 40 QWs (fF)	The cut-off frequency at -3dbm (GHz)
10	5.5	580
15	12.3	257
20	22	145
25	34.3	92
100	549	5.8

Table 3-3: Capacitance and cut-off frequency depending on the size of the MESA.

We realised the dimension, and the electrical circuit of the device to not be the limiting factor of the system in term of cut-off frequency. We, therefore, will be limited by the intrinsic properties of the materials up to the cut-off frequency of the electrical circuit where the two factors will degrade even faster the detection of these frequencies.

3.5.2. The ultrafast dynamic of QWIP

QWIP ultrafast dynamic is composed of 2 main photocurrents, one “fast” photocurrent which is due to the absorption of photons and a “slow” photocurrent which correspond to the refilling current due to the nonequilibrium space charges. Important works have been done by H. Schneider et al. on the properties of dynamic photocurrents by studying the transient intersubband photocurrent response after ultrafast IR pulses [38], [133], [159], [160].

Figure 3-36: Measure photocurrent transient at 5.2 V and 77 K, containing the “fast” and “slow” photocurrent for $In_{0.3}Ga_{0.7}As/GaAs$ [159]. In the inset, the illustration of the different regimes (a) photoexcited carrier emission, carrier capture and (c) redistribution of the space charges [38].

Depending on the probability of capture, the slow current can be as fast as the fast photocurrent or can last for a relatively long time.

3.5.3. Direct modulation

Figure 3-37: Schematic illustration of the setup used to investigate the direct high-speed modulation of the QWIP using a high-speed laser (QCL).

In this section, we present the high-speed characterisation of our detector. The setup for this experiment is shown in [Figure 3-37](#). A high-speed QCL shining at $4.7\ \mu\text{m}$ is mounted on a high-speed cryostat and cooled down to cryogenic temperature, a bias-T allows the injection of the DC and the microwave currents. The laser is modulated using a launcher and is detected by the QWIP after focusing it using two convex lenses. We also positioned the QWIP in a high-speed cryostat connected at bias-T to apply the bias voltage on the QWIP and measured the high-frequency photocurrent on a spectrum analyser Agilent E4407B which limits the detection to 26.5 GHz. The injection is tuned from 100 MHz to 26.5 GHz using a Signal generator Anritsu MG3693B with a step of 300 MHz at different power injection.

Figure 3-38: (a) Output power of the QCLs as a function of the current with AM-modulation illustrated at 400 mA, inset: superposition of the RF signal detected and photocurrent using blackbody radiation as a function of the voltage. (b) I-V curve with or without laser at 80 and 300 K, inset: picture of the high-speed device.

Figure 3-38 (a) shows the output power of the laser as a function of the current. The current used for the direct injection is 400 mA which corresponds to a power of about 100 mW. As seen in Figure 3-38 (b), the photocurrent introduced on the QWIP by the laser, at 80 K and 300 K are around 6 orders of magnitude higher than the background

current at 80 K. According to the responsivity calculated previously for 80 and 300 K at -5 V, they, fortunately, both correspond to power on the detector of about 4 mW for a 20 μm square device. The superposition of the QCL and QWIP spectrum is shown in Figure 3-27 (b).

The RF signal injected in the laser will modulate the IV curve of the QCL according to the power of the injection as seen in Figure 3-38 (a), it will be translated by amplitude modulation of the laser intensity. The amplitude of the signal modulated detected by the QWIP correspond to the high-speed photocurrent shown in the inset of Figure 3-38 (a) as a function of the bias voltage. It follows the behaviour of the photocurrent measured using the blackbody radiation. The high-speed detection will be dependent on the transmission of the electrical circuit of the QWIP but also on the quality of the injection in the laser. Therefore, the high-speed measurements will combine not only the losses due to the QWIP but also on the QCLs. We used the rectification method on the laser (seen in the black dashed curve in Figure 3-39 (b)) to have an idea of the injection quality of the laser.

Figure 3-39: (a) Detected power (solid line) and antenna noise (dashed line) as a function of the frequency, inset: 3D graph of the modulated signal versus the frequency and the span. (b) The modulated signal detected on the QWIP at 80 K and -8 V for different injection powers on the laser in solid line and the rectification curve of the QCL in dashed line.

To avoid a microwave pick-up, we separated the emitter and detector by more than a meter, and the power of the injected microwaves are as low as possible. Moreover, to remove the remaining antenna effects, we have measured the background by doing the

same measurement with the laser turned off as seen in the inset of [Figure 3-39 \(a\)](#). [Figure 3-39 \(b\)](#) displays the high-speed measurements with a bias voltage of -8 V on the 4-7C QWIP at 80 K with several injection powers in the QCL. [Figure 3-40 \(a\)](#) exposes the superposition of the measurements at cryogenic and room temperature. The level of the signal detected is related to the responsivity of the QWIP, as observed previously the photocurrent is about three times higher at RT than 80 K. Consequently, the high-speed curve at room temperature is shifted of about 5 dBm compare to the cryogenic temperature measurement.

[Figure 3-40](#): Comparison of the modulated signal detected on the QWIP for 80 and 300 K at -4 V. The detections of the beatnotes signals are shown in green dashed line.

Moreover, as one of the leading applications for this detector is to characterise frequency combs laser optically, we also detected the beatnote signal of the QCL. This approach is complementary with the previous high-speed measurements; the detection of the beatnote

signal, arising from the frequency mixing of longitudinal modes of our multimode laser, is done by mixing the modes of the spectrum in a high-speed mixer which here is the QWIP. The frequency of this signal is directly related to the length of the cavity but also to the dispersion. Moreover, thanks to the high-speed of our devices we can analyse the intermode beat spectroscopy where the autocorrelation of the beatnote at the comb frequency is measured using an FTIR and allow to know which optical modes are involving in the beating signal [61], [73]. It is, therefore, a crucial tool to study frequency combs lasers. As the length of the laser is 3 mm we detected the first and second harmonic of the beating signal corresponding respectively to about 11.5 and 23 GHz shown in [Figure 3-41](#) (a). With our detector at 300 K, we were able to observe the injection-locking of the beatnote by shifting the frequency injection of the laser around the free-spectral-range, as seen in [Figure 3-41](#) (b) and the power injection of the laser in [Figure 3-41](#) (c) [161].

Figure 3-41: (a) Detection of the first and second harmonic of the beating signal of a 3 mm QCL. Injection locking of the beatnote, (b) by shifting the frequency around the beatnote and (c) by increasing the injection power of the laser.

The same measurement has been performed on a 2 mm QCL corresponding to a beatnote of 22.43 GHz. Figure 3-42 shows (a) the detection of the beating signal for different bias voltage and (b) the injection-locking of the beatnote by shifting the frequency injection of the laser around the free-spectral-range both at 300 K. We can observe in both cases a locking range of about 2 MHz around the beatnote.

Figure 3-42: (a) Beatnote detection at 22.2 GHz for different bias voltage, (b) injection locking of the beatnote by shifting the frequency around the beatnote.

By these measurements, we demonstrated the abilities to detect the higher-order harmonics of the beating signal up to 24 GHz which is limited by our spectrum analyser. It opens a new field on the characterisation of frequency comb lasers in this wavelength domain.

3.5.4. Indirect modulation using rectification method

Due to the inherent nonlinear I-V characteristics of QWIPs, microwave rectification technique is a way to study the frequency response of the QWIP by applying an RF signal to the detector and measuring the variation of its DC biasing current. These values reflect the frequency roll-off behaviour of the intrinsic photoconductive lifetime and the electrical circuit limited by the device capacitance and differential resistance, along with other parasitic effects [38], [27].

The rectified DC current is given by:

$$I_{rect} = \frac{1}{4} I'' V_{\mu}^2 \quad (3-21)$$

With V_{μ}^2 the amplitude of the microwave signal applied on the QWIP and I'' the second derivate of the I-V curve. The dependences of ω are on these two factors:

- $I'' = I_0'' \alpha(\omega)$ with as described earlier $\alpha(\omega)$ the frequency dependence due too the intrinsic-transport mechanism such as $\alpha(\omega) = \frac{1}{(1+(\omega\tau)^2)}$, with τ the photoexcited electrons lifetime.
- $V_{\mu}^2 = V_{\mu 0}^2 \beta(\omega)$, with $\beta(\omega)$ the electrical circuit dependence which, for an RC circuit is: $\beta(\omega) = \frac{1}{(1+(\omega R_L C)^2)}$.

Therefore, the rectification current can be written as:

$$I_{rect} = \frac{1}{4} I_0'' \alpha(\omega) V_{\mu 0}^2 \beta(\omega) \quad (3-22)$$

The power injected must verify the small-signal condition; it means that the rectified current must be smaller than the current flowing in the device. Due to the tiny current (below than nA) of our devices at low temperature, this condition is not verified, and the rectified current is in the current noise. For 300 K the dark current is the order of mA, the small-signal condition is confirmed.

We used two different configurations to perform the microwave rectification technique, one utilising a launcher in the cryostat and a second using an RF tip both at 300 K respectively shown in the inset of [Figure 3-44 \(b\)](#) and [Figure 3-45 \(b\)](#).

[Figure 3-43](#): Setup for rectification measurement of photoconductive detectors.

Using this technique, we are no longer limited by the spectrum analyser, but only by the synthesiser RF generator and we can extend the measurement up to 30 GHz. The setup is illustrated in [Figure 3-43](#); we used the lock-in amplifier technique with an electrical modulation of the RF signal using a pulse generator. Thus, the RF injection is

overmodulated by a 10 kHz rectangular pulses with a duty cycle of 50 % and sent to the QWIP through the bias-T which also provides the bias voltage of the QWIP. The variation of the voltage is directly recovered at the terminal of the generator using a BNC-T and sent to the AC input of the lock-in amplifier synchronised with the pulse generator. Figure 3-44 (a) shows the variation of the rectified current as a function of the frequency injection for a bias voltage of -0.5 V on the QWIP while Figure 3-44 (b) compare two different bias voltages for the same injection power.

Figure 3-44: (a) Rectification current at a bias voltage of -0.5 V on the QWIP for several modulated powers injected.

The second rectification measurements of our devices have been obtained using a radio frequency ground-signal-ground (GSG) coplanar tip as seen in the inset of [Figure 3-45](#) (b).

Figure 3-45: (a) Rectification current versus of the frequency for different bias voltages with an injection power of 10 dBm. The dashed lines represent the noise ground and the transmission of the circuit, (b) superposition of the rectification measurements for three different samples, inset: (first bottom left) global view the setup, (second bottom left) zoom-in the coplanar tips applied on the sample.

The curves presented in [Figure 3-45](#) (a) a strong resonance at about 21 GHz which is due to a parasite effect seen in the transmission line of this setup and which is not present in

the direct and indirect modulation using the configuration with the launcher. The relative amplitudes of the rectification current as a function of the voltage follows the same behaviour than the variation of the photocurrent with the bias voltage (not showing here). Figure 3-45 (b) shows the same high-frequency behaviour for three different samples. For all samples, the rectification methods show modulation up to 30 GHz which is the limitation of our electrical system.

In order to demonstrate that all the resonances appearing in the direct and indirect methods come from the electrical circuit, we performed a new rectification method on a third setup using an RF molex with a cut-off frequency at about 26 GHz. Figure 3-46 displays the comparison between the modulation of all the different system.

Figure 3-46: Comparison between rectification methods using three different setups.

3.5.5. High-speed results

In order to superpose the rectification measurements with the optical one, we need to transform the values from the spectrum analyser in the equivalent current. Therefore, we

converted the direct modulation given by the spectrum analyser in photocurrent and normalised it. These results at 300 K are shown in [Figure 3-47](#).

Figure 3-47: Photoresponse of the QWIP with the QCL modulated at -10 dBm at 300 K (red line + symbol) at a bias voltage of -4 V. The rectification measurements of QWIP using the cryostat setup (blue line + symbol) and using the coplanar tip configuration (green line + symbol) with in dashed grey line the resonances due to parasitic effects of its electrical circuit.

The rectification measurements, displayed in [Figure 3-47](#), exhibit good agreement with the direct modulation using the QCL modulated at -10 dBm and show a frequency response up to 30 GHz. At 22 GHz the signal of the direct modulation is attenuated by approximately 8 dBm while at 30 GHz the rectification signal shows a diminution of approximately 10 dBm. We do not identify a clear change of the slope in the bode diagram of the rectification measurements or direct modulations which would correspond to the

cut-off frequency. The resonances of our system around 10 GHz are due to some parasitic effects in the electrical circuit using the launcher in the cryostat. We conducted the same measurements using a simpler electrical circuit composed of an RF coplanar tip, where these resonances were not shown as seen in the rectification methods in [Figure 3-45](#) and green in [Figure 3-47](#). However, this setup suffers from another resonance at higher frequencies represented in grey dashed line, also visible in the transmission line of the electrical circuit not displayed here.

In conclusion, in this chapter, we have demonstrated that QWIP at $4.9\text{ }\mu\text{m}$ with very wide frequency response and operating at room temperature. The increase of the photocurrent with the temperature is due to the height of the contact barrier which does not allow a sufficient supply of electrons from the contact to the QWs. Higher thermal energy allows a better supply due to an increase of the carriers travelling to the structure by tunnelling effect and thermal activation. Our high-speed QWIPs have a specific design based on relatively small MESAs using air-bridges which allows the propagation of high-frequency signals. It has been tested by direct and indirect method up to, respectively, 26 GHz and 30 GHz both limited by the electrical systems. Even though the signal to noise ratio decrease, the higher responsivity at room temperature improves the high-speed behaviour of our devices. We demonstrated that all the resonances in the high-speed measurements come from the electrical circuit and by clearing them, the devices show a flat modulation up to 30 GHz.

Beyond the high-speed behaviour of our devices, their responsivity and detectivity are close to the state-of-the-art of the research which makes them very promising for many applications in the first atmospheric window. However, they show a possible

improvement of the overall performances by implementing a new structure which would allow the release of electrons from the contact at a lower temperature such as using an optically active ohmic contact device as already proposed in [155]. The good results of these devices have motivated a start-up called Lytid (in collaboration with Paris Diderot) to launch a new product based on this samples processed at MPQ.

Chapter 4 Conclusion

4.1. Overall description and conclusion

The work covered in this thesis will be summarised in this final chapter. We will then conclude on these four years of my PhD candidature and finally open the subject to new horizons.

The objective of this PhD project was to develop and study the high-frequency domain of quantum devices based on intersubband transitions for emission and detection of coherent light in the two atmospheric windows included in the MIR region. Due to their very short intrinsic lifetime, ISBT can be modulated at very high-frequency, which make them suitable for a large panel of applications. Moreover, for frequency comb lasers, the potential detection and action on the beating signals of the longitudinal modes at the round-trip frequency allow a new degree of freedom as which will be discussed later.

Firstly, we draw an overview of the quantum devices based on ISBT and their main advantages such as a simple tunability of the optical transitions by engineering the thickness of the structure layer or/and the Aluminium factor of the alloys. We studied the quantum cascade lasers as quantum emitter devices and their high-performance in the MIR region. We explained the different designs of the active region based on high-extraction of the lower state to improve the population inversion. Regarding the high-frequency behaviour, QCLs cannot be classified with other devices. Indeed, due to their significant short upper state lifetime, they have specific parameters which distinguish them from other lasers. Due to these intrinsic parameters, nonlinear effects are enhanced

allowing self-phase-locking mechanism through the generation of optical modes using four-wave mixing. In low dispersion area, this can lead to a passive mode-locking which are intensively studied for their strong improvement in many applications. Frequency comb lasers, for example, are well known for their creation of ultra-short pulses, which can be done with QCLs under certain circumstances. However, due to the ultra-short lifetime of the electrons, longitudinal modes exhibit a new phase dependence which can lead to other features such as the emission of FM-modulated output and therefore, can bring in new applications. Considering the specific parameters of ISBT described earlier, we terminated this chapter by expressing the history, the state-of-the-art, the theory and the figure of merits of Quantum Well Infrared Photodetectors.

Chapter 2 covered the high-speed behaviour and the AM modulation of QCLs at the round-trip cavity which permits to achieve injection-locking. After a brief presentation of the process and characterisation setup we investigated the theoretical transient time applied to our devices and described the high-speed setups, we also studied the optical characterisation and the high-speed behaviour of our QCLs emitting at $4.7\text{ }\mu\text{m}$ and $9\text{ }\mu\text{m}$ using the rectification method. We described the effect of the RF injection on the spectra and the beatnote considering the pulling and thermal effect. The pulling phenomenon is illustrated by the interaction between a slave and a master oscillator. We have studied the spectra for both, free-running and injected-locked QCLs, for the injection-locking we observed a remarkable generation of optical modes in the spectrum corresponding to the side-band generation at a round trip frequency. The key measurements in this chapter are the investigation of the spectrum under RF injection in and out of the locking range of the beating signal with a high-resolution FTIR. The FSR is constant over the spectrum and under the precision of the FTIR which corresponds to no-dispersion under 10 MHz.

Regardless of the thin beating signal of the free running modes, the analysis of the modes spacing shows a comb-like regime for the free-running and injected-locked QCL. Furthermore, it allows us to see the apparent effect of the injection in the locking range on a large part of the spectrum with the variation of the free-spectral range to meet the frequency injected. However, an injection out of the locking range leads to an effect on only a part of the spectrum. The tunability of the FSR is a precious advantage in dual-combs spectroscopies [65].

The second important part of this PhD project was the development of high-speed detectors. These tools are essential for many applications, including high-rate free space communication on the atmospheric windows. Quantum well infrared photodetectors are also based on intersubband transitions and show high performances. The same advantages as described earlier are also possible on these quantum devices. We first explained the physics of these detectors and the calculation of the figure of merits and then details the process required for high-frequency comportment. A study has been done to understand the temperature and electric field dependence of these devices, especially in the MWIR where the effect of the contact are dominants at low temperature. Our QWIP show performances at the state-of-the-art for these devices and perform very well at RT. Indeed, even though the signal to noise ratio is degraded with the temperature, RT operation is possible due to its high-responsivity at 300 K. This is explained by a better electron extraction from the contact which can refill the structure more efficiently at a higher temperature due to the more homogeneous distribution of the electric field across the quantum wells. The last part of this chapter is concentrated on the high-speed behaviour of the QWIPs; direct and indirect methods have been used and demonstrate a high-speed behaviour up to room-temperature limited to 30 GHz by the electrical system used for the

experiment. We concluded these experiments by the detection of the beatnote of the QCLs. Different lengths of the lasers were used and showed an injection-locking of the beating signal at room temperature and up to 22 GHz (for 2mm QCLs).

As a conclusion, the innovation created by high-speed operations and frequency comb lasers in the MIR has led to a considerable expectation in this field. Contrary to conventional materials, quantum devices based on intersubband transitions exhibits good efficiency in this domain and high-speed behaviour which make them suitable for a large panel of applications. The benefits of our quantum devices have been explored and used to develop high-speed and high-performance QCLs and QWIP in the MIR. Thin beating signal and no dispersion measured over the spectrum launch us on the track of frequency comb operation thanks to the coexistence of strong nonlinear process and low-dispersion region. Moreover, we performed injection-locking under RF injection at the round-trip frequency and in the locking-range. This powerful pair of quantum devices opens the field for innovation in the MIR region.

4.2. Recommendation

As it is always the case, a study permitting to solve problems raise more questions and investigations. Thus, our work in this PhD project has led us to a new exploration for a better understanding of some phenomena and improving the performances of our structures.

4.2.1. Passive mode-locking

With regards to the passive mode-locking in QCL devices, with the critical parameter being the dispersion, further works can be done by decreasing the one introduced by the

waveguide and by the gain curve. To reduce the dispersion and widen the effect of the FWM, a design of several active regions in cascade can lead to flatter gain curve and thus, reducing its impact. *Figure 4-1* (b) represents a superposition of 3 different active regions which have already shown good results in the literature. Moreover, to optimise the dispersion due to the confinement factor, the various active regions are grown in sandwiched as displayed in *Figure 4-1* (a).

Figure 4-1: (a) Illustration of the cross-section of 3 different active regions and their position to decrease the waveguide dispersion and (b) gain curves of each wavelength and the sum of the different active regions.

Moreover, dispersive mirror or grating can be designed to introduce an opposite dispersion compensating the actual one. These chirped mirrors have been successfully released for THz QCLs as shown in *Figure 4-2* (a) and (b) and for MIR QCLs in *Figure 4-2* (c).

Figure 4-2: Double-chirped mirror architectures for THz QCLs, (a) the refractive index change due to the width of the waveguide and the period for the wavelength reflected [162], (b) the high refractive index contrast is obtained by alternating layer of benzocyclobutene and the waveguide to ensure a compensation of the dispersion [73] and (c) different layer of Al_2O_3 and SiO_2 with a chirped frequency allowing a compensation of the dispersion [163].

A complete analyse of the mode-lock laser need to be done in order to have the information on the phase relation of each longitudinal modes under the comb regime. Due to the short lifetime of the carrier, it is necessary to use a no-conventional method using ultrafast detector as the one developed in Chapter 3. This method has been developed by A. Hugi et al., the relative coherence of QCL combs can be analysed by intermode beat spectroscopy where the autocorrelation of the beatnote at the comb repetition frequency is measured using a FTIR [61], [73].

Another interesting study would be to examine the chaos of these lasers. Indeed, due to their properties, QCLs are calcified as Class-A lasers. Under certain circumstances, we can observe, what seems to be, chaotic effects in the spectrum. It would be especially interesting to study this effect, in the light of a high-speed detector to see the infect include in the repetition rate of the laser. In such circumstances, we can observe many spikes in the RF domain which would disappear in normal operation. These phenomena are

observable for some current density; however, they could also be coming from the poor isolation of the back reflection in the laser cavity.

By using the high-speed abilities of our QWIPs, we could study the variation of the total intensity of the laser under such circumstances and repeat the same study under RF injection to show the beneficial effect of the injection on the stability of the laser.

4.2.2. Injection-locking to active mode-locking

Due to the poor overlapping of the RF signal with the active region, the efficiency of the RF injection is in the order of few percents. A better design can be done to enhance the injection efficiency, for example, a QCL embedded into a micro-strip line shown an improvement in the frequency response [161]. This idea, coupled with an intense investigation of the phase-sensitive mechanism such as the four-wave mixing, can lead to an overall improvement of the injection-locking. Other methods to achieve active mode-locking should be investigated, for example, by modulating only a short section of the QCL at the round-trip frequency [51]. Regarding the conventional mode-locking for the generation of pulse QCLs, the upper state lifetime must be engineered to be higher than the round-trip frequency and therefore, allow the propagation of an ultra-short pulse in the cavity. Active region using photon-assisted tunnelling transitions exhibits can fulfil such requirement [75].

4.2.3. Performance of the QWIP

The effect of the contact barrier does not allow a homogeneous repartition of the electric field. Therefore, at low temperature, we show a concentration of the electric field in the first barriers which does not allow the rest of the structure to perform correctly. An

improvement can be made by implementing a new structure enabling a more efficient carrier refill to the structure. Such layer can be imagined as an optically active ohmic contact.

4.2.4. High-speed behaviour

Due to the dimension of the wave, RF region is a complex domain where most of the part of the devices can act as an antenna effect and introduce resonances. A more robust system could lead to fewer resonances on the system as we saw by using coplanar tips. As for all RF experiments, a robust electrical circuit, along with a better impedance matching, can decrease the resonance and improve the amplitude of the modulation collected. Moreover, our operations were limited to 30 GHz by the electrical system. Pushing these limits would allow us to explore the intrinsic behaviour and the cut-off frequency of our system at a higher frequency.

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