Photonic engineering of terahertz quantum cascade lasers

Liang, Guozhen

2015

Liang, G. (2015). Photonic engineering of terahertz quantum cascade lasers. Doctoral thesis, Nanyang Technological University, Singapore.

<https://hdl.handle.net/10356/65315>

<https://doi.org/10.32657/10356/65315>

**NANYANG
TECHNOLOGICAL
UNIVERSITY**

**Photonic engineering of terahertz quantum
cascade lasers**

Liang Guozhen

School of Electrical & Electronic Engineering

2015

Photonic engineering of terahertz quantum cascade lasers

Liang Guozhen

School of Electrical & Electronic Engineering

**A thesis submitted to the Nanyang Technological University
in partial fulfillment of the requirements for the degree of
Doctor of Philosophy**

SUMMARY

The terahertz (THz) frequency range, lying between microwave and mid-infrared frequencies (~ 0.3 THz to 10 THz), remains one of the least developed regions in the electromagnetic spectrum. THz radiation has, however, proven potential widespread and important applications in, for example, spectroscopy, heterodyne detection, imaging and communications. In many of these applications, high power coherent THz sources with low beam divergence, single-mode operation, tunable intensity and controllable polarization are highly desirable. Therefore, much effort has been placed on the development of new and appropriate THz sources, and in particular with a focus on the development of electrically pumped, semiconductor-based light sources that can be mass produced.

A breakthrough occurred in 2002, when the first THz quantum cascade lasers (QCLs) was demonstrated. QCLs are semiconductor lasers comprising multiple strongly coupled quantum wells, exploiting electron transitions between subbands of the conduction band. By adjusting the widths of the quantum wells/barriers, the emission frequency and performance of the laser can be tailored, adding additional flexibility in the laser design. Without the assistance of an external magnetic field, THz QCLs have covered the frequency range from 1.2 to 5 THz. With respect to the optical power, THz QCLs can provide over 1W peak power in pulse mode, and 130 mW in continuous-wave (CW), at 10 K heatsink temperature, in devices using a single-plasmon waveguide. Although relatively good performance of THz QCLs has been achieved, there is still sufficient room to further improve the performance of THz QCLs, for achieving arbitrary beam control, spectral emission, intensity modulation, and polarization control, so on and so forth. Photonic engineering through the design of e.g. Bragg gratings, photonic crystals, plasmonic/metasurface/metamaterial structures, and hybrid material systems, provides an excellent platform to achieve full manipulation of THz waves.

In this thesis, we aim to explore photonic engineering techniques to obtain high performance THz QCLs. First, we report the design, fabrication and experimental characterization of surface-emitting THz frequency QCLs with distributed feedback concentric-circular-gratings (CCG) for beam engineering to achieve a narrow beam divergence. Single-mode operation is achieved at 3.73 THz with a side-mode suppression ratio as high as ~ 30 dB. The device emits ~ 5 times the power of a ridge

laser of similar dimensions, with little degradation in the maximum operation temperature. Two lobes are observed in the far-field emission pattern, each of which has a divergence angle as narrow as $\sim 13^\circ \times 7^\circ$.

In addition, we also report on the planar integration of tapered THz QCLs with surface plasmonic waveguides, which are developed to provide a versatile platform for beam engineering and optical components integration. For example, by introducing periodically arranged surface scatterers, the whole structure functions as an efficient collimator, resulting in a tight THz surface-emitting beam with a divergence as narrow as $\sim 4^\circ \times 10^\circ$. As all the structures are in-plane, this scheme also provides a promising platform for the construction of an active integrated THz photonic circuit by incorporating other optoelectronic devices such as Schottky diode THz mixers, and graphene modulators or photodectors.

Furthermore, for practical applications, modulation of THz light is usually needed to encode information in THz waves or to perform lock-in amplification of the signal, etc. However, traditional THz modulators suffer from either slow modulation speed (mechanical optical chopper) or small modulation depth (electro-optical devices). Here, we demonstrate integrated THz graphene modulators on THz CCG QCLs, which enable the modulation of THz light intensity with a 100% modulation depth and a fast modulation speed.

Thesis Supervisor: Wang Qijie
Title: Nanyang Assistant Professor

ACKNOWLEDGEMENT

First and foremost, I'd like to gratefully acknowledge my supervisor Prof. Wang Qijie for his patience, kindness and insightful guidance along the way. Being a creative advisor with an optimistic and humorous personality, he has made the researches and life in our group an enjoyable experience. I am also sincerely grateful for his great confidence in my ability of handling the projects and continuous help and encouragement during the hard time both in research and life. I could not have finished the works in this thesis without all these support.

I also want to thank the members of our group. I am indebted to Dr. Liu Tao, Dr. Liang Houkun, Tao Jin, Meng Bo, Hu Xiaonan, Yan Zhiyu, Dr. Li Xiaohui, Dr. Zhang Yongzhe, Shen Youde, Yu Xuechao, *et al.* It has been a great time working and living with them. I also would like to express my gratitude to Prof. E. H. Linfield, Prof. A. G. Davies and Dr. Lianhe H. Li from Leed University for the growth of the active regions and their comments on the manuscripts.

I would like to thank Mr. Fauzi and Ms. Seet in the characterization lab, Dr. Chong Gang Yih in CR1, Mr. Shamsul and Mr. Mak in CR2 for their prompt help and support in the experiments.

Finally, I would like to thank my wife Wang Lulu, who provides no end of support from the very beginning. Her optimism, great sense of humor and consolations always keep me going through the hardships and make life more joyful. I'm also grateful to my parents, sister and brother for their always unconditional love and support.

CONTENTS

Summary	1
Acknowledgement	3
List of Figures	6
List of Tables	11
1. Introduction	12
1.1 Background of the terahertz technology.....	12
1.1.1 THz applications	13
1.1.2 THz radiation sources	16
1.2 THz Quantum cascade lasers.....	19
1.3 Motivations and challenges.....	25
1.4 Objectives and methodologies	26
1.5 Thesis overview and major contribution	26
2. Active Region Design Of Terahertz Quantum Cascade Lasers	28
2.1 Theory	28
2.1.1 Subbands in quantum wells.....	28
2.1.2 Intersubband optical gain.....	30
2.1.3 Intersubband relaxations	32
2.1.4 Resonant tunneling transport	36
2.2 Typical active region designs	38
2.3 A bound-to-continuum phonon-photon-phonon active region design	42
2.3.1 Band structure.....	43
2.3.2 Fabrication	45
2.3.3 Preliminary experimental results	50
2.4 Conclusion	51
3. Surface-Emitting Concentric-Circular-Grating Terahertz Quantum Cascade Lasers	53
3.1 Introduction	53
3.2 Grating Design	57
3.3 Fabrication	64
3.4 Experimental results and discussion	65
3.4.1 Light-current-voltage characterization	65
3.4.2 Effect of boundary deformation on the optical mode.....	66
3.4.3 Coherent superimposition of the optical modes.....	68

3.4.4	Frequency splitting caused by the boundary deformation.....	69
3.4.5	The whispering-gallery-like Modes.....	70
3.4.6	Lasing at third-order azimuthal mode	71
3.5	Conclusion	73
4.	Planar Integrated Metasurfaces For Highly-Collimated Terahertz Quantum Cascade Lasers	74
4.1	Introduction	74
4.2	Device design	76
4.2.1	Device overview	76
4.2.2	Design of the tapered laser cavity	77
4.2.3	Design of the metasurface collimator	80
4.3	Fabrication	83
4.4	Experimental results and discussion	84
4.4.1	Light-current-voltage and far-field characterization	84
4.4.2	Influence of the groove width on the far fields	86
4.4.3	Comparison between the devices with and without the metasurface	87
4.4.4	Advantages and disadvantages	88
4.5	Conclusion.....	89
5.	Integrated Terahertz Graphene Modulator With 100% Modulation Depth.....	90
5.1	Introduction	90
5.2	Device overview	92
5.3	Individual characterizations of the THz CCG QCL and the graphene	93
5.4	100% modulation depth of an integrated graphene modulation with CCG QCLs	96
5.5	Modulation speed of the integrated graphene modulator	98
5.6	Fabrication	101
5.7	Supplementary Information	102
5.7.1	Farfield and optical mode of the concentric-circular-grating (CCG) QCL.....	102
5.7.2	Raman Spectra of the transferred graphene	103
5.7.3	Model used to retrieve the graphene parameters	104
5.7.4	Comparison of the graphene response at 78 K and 300 K.....	105
5.7.5	High-frequency circuit model of the graphene modulator.....	106
5.8	Conclusion.....	110
6.	Conclusions and Future works	111
	Publications.....	113
	Bibliography.....	114

LIST OF FIGURES

Figure 1-1. The ‘Terahertz gap’ in the electromagnetic spectrum lurking between the microwave and mid-infrared regions.	12
Figure 1-2. Radiated energy versus wavelength showing 30-K blackbody, typical inter-stellar dust, and key molecular line emissions in the terahertz (reprinted from [30]).....	14
Figure 1-3. (a) Schematics of the THz microscope setup. QCL: a 2.9 THz quantum cascade laser; L1, L2, L4: Picarin lenses; S1, S2: Silicon lenses; TPX: TPX lens; M2. (b) A photograph of a fresh leaf. (c) Confocal THz image of the same leaf. The measurements covered an area of $1.8 \text{ mm} \times 3.2 \text{ mm}$ and 90×160 pixels were recorded. (d) The rectangular white frame in panel (c) is imaged again with higher resolution (200×200 pixels). (Adapted from [35])	15
Figure 1-4. A portion of the band diagram of an initially proposed intersubband laser, formed by cascading multiple transition modules. Radiative transitions take place between level 3 and level 2, the electrons then undergo fast relaxation to level 1 and are re-injected into next module.	20
Figure 1-5. Schematic band diagram of the first quantum cascade laser operated at $4.2 \mu\text{m}$, including the quantized electron states and the magnitude squared of the wavefunctions. Each period/module of the structure consists of an injector (the digitally graded alloy) and an active region. (Reprinted from [67]).....	21
Figure 1-6. (a) Conduction band diagram of a portion of the layer stack of the first terahertz quantum cascade laser. The magnitude squared of the wavefunctions are shown, with miniband regions represented by shaded areas. Optical transition takes place between level 2 and 1. (b) Optical mode profile along the growth direction of the device. The waveguide core of 104 repetitions of the active region shown in (a) is between the bottom contact and top metal layer. (Reprinted from [18]).....	23
Figure 1-7. Maximum operation temperatures of THz QCLs as a function of frequency in the literature. The inset shows the timeline of the maximum temperatures. (Reprinted from [85])	24
Figure 2-1. Schematic view of the intersubband relaxation paths. LO: LO-phonon scattering; e-e: electron-electron scattering; ii: ionized impurity scattering; ir: interface roughness scattering; ac: acoustic phonon scattering.	33
Figure 2-2. (a) Electron-LO-Phonon scattering between two subbands with in-plane momentum exchange. (b) Relationship between the wavevectors of the electron before and after transition and the in-plane phonon wavevector.	34
Figure 2-3. Schematic illustration of energy states and their wavefunctions in adjacent quantum wells before (a) and after (b) bringing into resonance. Resonance of the two states occurs when they are aligned in energy and split into doublets. The minimum energy splitting Δ_0 is known as the anticrossing gap.	37
Figure 2-4. Different models used to depict the resonant tunneling of the electrons through a potential barrier. (a) In a coherent transport model, the states in resonance split into energy doublets, whose wavefunctions are spatially extended across the quantum wells. (b) In a tight-binding model, the two resonant states are coupled with a strength of $\Delta_0/2$, but their wavefunctions are localized within individual quantum well.	38
Figure 2-5. Simplified conduction band diagrams and the moduli squared of wavefunctions for typical active region designs of THz QCLs. The upper laser state u and lower laser state l are highlighted in red and blue, respectively. (a) A chirped superlattice design; (b) A bound-to-continuum design; (c) A resonant phonon design; (d) A phonon-photon-phonon design. Two modules are shown.....	40

Figure 2-6. Current densities when the injection state is aligned with (a) the lower laser state at low voltage and (b) the upper laser state at higher voltage.....	42
Figure 2-7. Conduction band structure and the moduli squared of wavefunctions of the BTC-PPP THz QCL active region design at a bias of 11.7 kV/cm. The layer thickness in nanometer are 4.1 /8.3/ 2.4 /12.0/ 1.7 /10.6/ 3.4 /8.7/ 3.8 /6.9/ 4.2 /14.6, the barriers are indicated in bold fonts and the underlined well is doped.....	44
Figure 2-8. Schematic illustration of the fabrication steps for metal-metal waveguide ridge THz QCL.	48
Figure 2-9. SEM images showing the cross-sectional view of the bonding region. The absence of a visible Au-Au interface indicates a good bonding quality.....	49
Figure 2-10. SEM images showing the sidewall profiles of the ridges on (100) GaAs wafers etched by the H ₂ SO ₄ : H ₂ O ₂ : H ₂ O (1:7:80) solution. (a) and (b) are for ridges aligned to the <110> and <101> directions, respectively.	49
Figure 2-11. Cross-sectional-view SEM image of a fabricated device.....	49
Figure 2-12. (a) Light-Current-Voltage characterization of the BTC-PPP laser at 10 K. The threshold and rollover current densities are $J_{th} = 0.185 \text{ kA/cm}^2$ and $J_{max} = 0.22 \text{ kA/cm}^2$, respectively. (b) Spectra of the laser at different current density at 10 K.	51
Figure 3-1. Schematic draw of a ridge QCL employing metal-metal waveguide, where the active region (laser core) is sandwiched between two metal plates.....	54
Figure 3-2. Some typical wave engineering schemes for THz QCLs, involving edge-emitting devices (a)-(d) and surface-emitting devices (e)-(i). Edge emission: (a) Integrated horn antenna [98]; (b) lens coupling [97]; (c) Integrated surface plasmonic collimator on the facet [100]; (d) Linear third-order DFB grating [101]. Surface emission: (e) Linear second-order DFB grating [103]; (f) Second-order ring grating [105]; (h) Phase-locked second-order DFB grating array [111]; (i) 2D photonic crystal [112].	56
Figure 3-3. (a) Three-dimensional schematic representation of the THz concentric-circular-grating quantum cascade laser design. The semitransparent rectangle represents the two-dimensional (2D) simulation plane. (b) 2D simulated magnetic field distribution making use of the 360°rotational symmetry of the designed structure. The magnified views show the widths of the metal in each period and the boundary region without metal coverage, indicated as R1, R2, ... R15 and δ , respectively. The black lines represent metal.....	58
Figure 3-4. (a) Electric field distribution of the standard second-order DFB CCG THz QCL in the central region. (b) – (d) Mode spectra of the structures for different R15 with other parameters unchanged. (e) – (g) the corresponding electric-field (E_z) profiles of the expected modes, highlighted by the red arrows in (b) – (d), respectively, obtained at the middle height of the active region.....	60
Figure 3-5. (a), (b), (c) Simulation results after each adjustment of the slit locations. (i) Electric field ($ E $) distribution of the expected mode at the central part of the grating. The first five slits are numbered 1, 2, ...,5, and their locations are indicated by the white arrows. White dashed lines near the arrows indicate the original locations of the slits. The displacement distance of each slit is given above the arrow. (ii) Mode spectrum of the adjusted structure, a red arrow indicates the expected mode. (iii) The electric-field (E_z) profile.	62
Figure 3-6. (a) A false-color SEM image shows the fabricated concentric circular grating (CCG) terahertz quantum-cascade laser (QCL), in which a three-spoke bridge structure connects the concentric rings together to allow electrical pumping of the whole grating. (b) Optical microscope images of the device. Magnified views on the right show the dimensions of the center ring and the boundary slit without metal coverage.	63
Figure 3-7. (a) Mode spectrum of the device using 3D full-wave simulation to take the higher azimuthal modes into account. Only the first four lowest order azimuthal modes are plotted. The electric field distributions of the modes with lowest losses (the fundamental and second	

azimuthal modes, highlighted by a red circle) are shown in the inset. (b), (c) Comparison of the Bessel function $J_0(r)$ and $-J_2(r)$, and $Y_0(r)$ and $-Y_2(r)$, respectively. The fundamental and second-order Bessel functions coincide except in the first few peaks. 64

Figure 3-8. (a) Light-current-voltage characterization of the CCG device at different heatsink temperatures under pulsed mode operation. (b) Emission spectra of the device at 9 K heatsink temperature as a function of injected current, from threshold to the rollover. The inset shows a logarithmic scale plot of the spectrum, demonstrating a side-mode suppression ratio of around 30 dB. 66

Figure 3-9. (a) Measured far-field pattern of a typical THz CCG QCL. (b), (c), and (d) Electric field distributions and the corresponding far-field patterns for the fundamental, the first-order and second-order azimuthal modes, respectively. 67

Figure 3-10. (a) Scanning electron-beam microscope images of the anisotropic sidewall profiles caused by wet chemical etching. The sidewalls of the upper and lower boundaries are more vertical than those of the left and right boundaries. (b) The change of electric field distribution and far-field pattern caused by a slight (2 μm) deformation of the circular active region disk. 68

Figure 3-11. Coherent superimposition of the far fields of the fundamental and second-order azimuthal modes. (a) – (e) Resulting far-field patterns with the phase difference of the two modes $\Delta\phi$ varies from 0° to 120° . (f) Resulting far-field pattern with $\Delta\phi = 0^\circ$ but 1.5 times amplitude for the second-order azimuthal mode. 69

Figure 3-12. Splitting of the fundamental azimuthal mode under deformation. (a) Electric field distribution of the original fundamental azimuthal mode. (b), (c) Electric field distributions of the splitted deformed fundamental azimuthal modes, and their corresponding far-field patterns (d), (e), respectively. 70

Figure 3-13. Supported modes in the CCG DFB QCLs, and the corresponding momentum space intensity distributions. (a) Electric field intensity of the third-order azimuthal lasing mode and (c) a whispering-gallery-like (WGL) mode. (b) and (d) in-plane momentum-space intensity distributions for the modes in (a) and (c), respectively. The red dashed circle represents the light-cone – only radiation components inside the light-cone can be out-coupled. There are no intensity components in the light-cone for the WGL mode, in contrast to the situation for the third-order CCG mode. 71

Figure 3-14. Measured results of an early CCG DFB laser where the refractive index of the active region was not properly estimated. (a) Pulsed (200 ns pulses repeated at 10 kHz) light-current-voltage (LIV) characteristics of the laser. (b) Emission spectra of the laser at 78 K for different injected current densities, from the threshold to the rollover. The inset shows a logarithmic scale plot of the spectrum, indicating a side-mode suppression ratio of around 30 dB. 72

Figure 3-15. Two-dimensional far-field emission patterns of the CCG DFB QCL. (a) The experimentally measured emission, and (b) the simulated emission of the third-order azimuthal mode. 73

Figure 4-1. (a) Scanning electron microscope (SEM) image of a fabricated device. The tapered THz QCL consists of DBR, ridge and taper sections. The inset shows the details of the DBR structure, which is formed simply by gold patterning on top of the active region. The markers by the side of the laser are used for mask alignment during the fabrication. (b) 3D schematic cross-sectional view of the device along the white dashed line in (a). (c) Enlarged top view of the central region of the device. 77

Figure 4-2. (a) Electric field (E_z) distribution of the tapered THz QCL with a curved front facet; magnified views show the details of the DBR region, and the taper. The black regions represent the active regions without metal coverage, which are highly absorbing. (b) E_z distribution of a tapered structure with a flat facet. The electric field in the laser cavity is

distorted, resulting in uncollimated emission. (c) Ez distribution of a second-order lateral mode which is suppressed due to a larger overlap of the mode with the side absorbers.	79
Figure 4-3. Simulated reflectance spectrum of the DBR region. High reflection only happens in the frequency band ranging from 2.9 THz to 3.4 THz.	80
Figure 4-4. (a) Simulated 2D light intensity distribution of a metasurface collimator. The simulation was performed in the (y-z) plane along the symmetric line (white dashed line in Figure 1(a)) of the device. (b) Cross-section of the metasurface collimator design. The narrow grooves have a width of 5 μm and a period of 10 μm . The narrow grooves are 9 μm deep in the orange region and 7 μm deep in the yellow region (which is repeated 25 times). The narrow grooves in the orange region are deeper to enhance coupling from the laser into the SSPs. The 25 μm grooves, which scatter out the SSPs, are 9 μm in depth. (c) Dispersion diagrams of the metasurface with grooves of 5 μm width and 10 μm period, but different depths. The black dotted curve, red curve and the blue dashed curve correspond to $h = 0 \mu\text{m}$ (flat surface), 7 μm and 9 μm , respectively. The $h = 0 \mu\text{m}$ curve almost coincides with the light line in vacuum in the THz frequency range. The lower inset presents a zoom-in view of the region marked by a green box. (d) Dependence of the transmission through the laser facet on the width of the metasurface. (e) Calculated far-field intensity profile along the metasurface waveguide direction. The enlarged view of the central lobe in the inset shows that the beam divergence is as narrow as 2.7° . (f) Simulated 2D light intensity of a laser without the metasurface structure. The inset shows that the light emission is relatively uniform in all directions.	82
Figure 4-5. Propagation length and the length of the evanescent tail of the THz surface plasmonic wave on the metasurface as a function of the groove depth. The inset shows a zoom-in view of the same data. The simulated wavelength is 95 μm	85
Figure 4-6. Pulsed light-current-voltage (LIV) characteristics of device A at different heat sink temperatures. Inset: Lasing spectra of devices A and B at 4.3 A and 9 K – conditions under which their far-field emission profiles were measured.	85
Figure 4-7. (a) SEM images of device A and B with groove widths of 600 μm and 1000 μm , respectively. (b) ϕ and θ direction line scans through the peak in the measured far-field radiation pattern in (c) for device A. (c) and (d) Measured far-field patterns of devices A and B, respectively. (e) Simulated far-field pattern of device A at the wavelength of 94 μm . (f) Simulated far-field patterns of device B taking into consideration the multi-mode emission from the active region at wavelengths between 92 μm and 98 μm	86
Figure 4-8. Near field light intensity distributions of (a) device A and (b) device B on a logarithmic scale, taken on a plane 2 μm above the device surface. (c) and (d) Amplitude and phase distributions of the radiative electric field (E_y) along the white dashed line in (a) and (b), respectively.	87
Figure 4-9. (a) Measured farfield pattern of a device with only the second order grating and without the metastructure. (b) Measured farfield pattern of a device with metastructure.	87
Figure 5-1. Overview of the integrated graphene modulator with quantum cascade laser. (a) Schematic illustration of the device. Only the central several rings of the circular-concentric grating (CCG) (orange region) are connected together with the spoke bridges to allow electrical pumping of the quantum cascade laser (QCL) over a small active region. Light is emitted vertically from the surface and is modulated by the electrically gated graphene. (b) Optical microscope images showing the central part of a fabricated device. (c) An enlarged view shows details of the graphene and the QCL electrodes, which are insulated by a 450-nm-thick SiO_2 layer. A wrinkle, resulting from folding of transferred chemical vapor deposition (CVD) graphene, is visible at the lower right corner.	93

Figure 5-2. Individual characterization of the THz QCL and the graphene layer. (a) Light-current-voltage (LIV) characteristics of the THz CCG QCL under different heatsink temperatures. (b) Laser spectra as a function of pump current I , at 20 K. (c) Schematic representation of the structure used to characterize the graphene layer. (d) DC conductivity, carrier density and Fermi energy of the graphene as a function of gate voltage, measured at 78 K. (e) Gate voltage dependence of the modulation of the THz transmitted intensity (~ 3 THz) by the graphene, showing a modulation depth of $\sim 11\%$ over the applied bias range from -40 V to 60 V.	95
Figure 5-3. Modulation depth of the integrated graphene modulator on CCG QCL. (a) Enhancement factor of the amplitude of the electric field in the graphene region near the output aperture of the CCG QCL as a function of distance above the CCG. The inset shows a magnified portion of the curve. (b) Enhanced modulation of the THz wave by the integrated graphene modulator. The intensity of the lasing peak varies from nearly zero at $V_G = -26$ V to a maximum at $V_G = +40$ V. (c) V_G dependence of the output power of the CCG QCL with (circles) and without (rectangles) the graphene. (d) Light-current characteristics of the QCL as a function of V_G confirming the 100% modulation depth achieved by the integrated graphene modulator.	97
Figure 5-4. Calculated losses for the sixth-azimuthal modes with the graphene gated at two different Fermi levels. The loss of the lasing mode increases by 2.8 cm^{-1} when the Fermi level of graphene is tuned from Dirac Point to -220 meV	98
Figure 5-5. Measurement of the modulation speed of the integrated graphene modulator. (a) (i) Device bias scheme. The pulse duration for the QCL was fixed at $T_{\text{QCL}} = 1 \text{ } \mu\text{s}$ (corresponding to a frequency of $f_{\text{QCL}} = 1 \text{ MHz}$), while an AC rectangular signal was applied onto the graphene electrode with 50% duty cycle and various periods of $T_G = T_{\text{QCL}} / N$ ($f_G = N f_{\text{QCL}}$), where N is an integer. (ii) The predicted output optical signal of the device if the graphene modulator is able to follow the variation of the applied voltage. In this case, the average peak power $P_{\text{ave}} = (P(V_{\text{gr}}) + P(V_0))/2$, where $P(V_{\text{gr}})$ and $P(V_0)$ are the output peak powers when a constant V_{gr} or V_0 is applied to the graphene electrode, respectively. (iii) The predicted output optical signal of the device if the applied signal on graphene is much faster than the speed of the modulator, in which case P_{ave} deviates from $(P(V_{\text{gr}}) + P(V_0))/2$. (b) P_{ave} as a function of modulation frequency. The blue line (star symbols) corresponds to $V_0 = 0 \text{ V}$ and $V_{\text{gr}} = 10 \text{ V}$, while the red line (circle symbols) corresponds to $V_0 = 0 \text{ V}$ and $V_{\text{gr}} = 15 \text{ V}$. The grey, blue and red shaded ribbons indicate the output power when the graphene electrode is DC biased at 0 V , 10 V and 15 V , respectively. The widths of the ribbons manifest the instability of the laser power during measurement period. (c) Frequency response of the graphene modulator.	100
Figure 5-6. Farfield and optical mode of the CCG QCL. (a) Measured two-dimensional farfield emission pattern of the surface-emitting CCG QCL, where the $(0, 0)$ position represents the normal to the laser surface, and the corresponding electric field (E_z) distribution of the laser in top view (b) and cross-section view (c). The white dash line enclosed the pumped area.	103
Figure 5-7. Raman characterization of the transferred graphene. Raman Spectra of the transferred graphene on (a) $\text{SiO}_2/\text{p-Si}$ substrate and (b) in the slits of the CCG of the integrated device.	104
Figure 5-8. Square resistance of the CVD graphene transistor as a function of the gate voltage. Symbols: measured data; Curve: modeling result.	105
Figure 5-9. Comparison of the graphene response at 78 K and 300 K. (a) Calculated transmittance normalized to the value at the Dirac point and the electrical transport measurements of the graphene sheet at 78 K and 300 K. (b) Modulation of the THz radiation by the graphene at 78 K and 300 K with the effect of the Si substrate removed.	106

Figure 5-10. Equivalent circuit model of the integrated graphene modulator. (a) Schematic of the modulation scheme, the internal impedance of the function generator is R_0 ($50\ \Omega$). (b) Main parts that affect the high speed performance of the integrated modulator. (c) cross-section view of modulator in (b) and the equivalent circuit for a single slit. (d) equivalent circuit for the whole modulator. (e) simplified circuit model for the modulator.	109
Figure 5-11. Frequency response of the integrated graphene modulator measured by a RF network analyzer. The blue triangular symbol represents the S21 response of the device and the black circular symbol corresponds to the electrical modulation applied to the graphene sheet, whose 3-dB cutoff frequency was estimated to be 110 MHz.	110
Figure 6-1. Schematic of a lab-on-a-chip device for molecule sensing application.	112

LIST OF TABLES

Table 2-1. Comparison of some important parameters of THz QCL between the BTC-PPP and other two state-of-art designs [93, 98].

Table 2-2. Growth sheet of the BTC-PPP active region.

1. INTRODUCTION

In this chapter, the background of the terahertz technology will be introduced in the beginning, followed by a review of the development of a novel semiconductor laser – quantum cascade laser, where challenges and motivations will be clarified. The organization and major contribution of the thesis will be given at the end of the chapter.

1.1 BACKGROUND OF THE TERAHERTZ TECHNOLOGY

Lying between the microwave and mid-infrared frequencies, the electromagnetic spectrum band (~ 0.3 THz to 10 THz, Fig. 1-1) was often referred to as ‘terahertz (THz) gap’ due to the difficulties in efficiently generating, manipulating and detecting THz radiation. However, driven by the great potential for many diverse applications ranging from non-destructive imaging [1–5], spectroscopic sensing [6–8] to ultra-high bit rate wireless communication [9–11], to be detailed below, continuous efforts have been devoted to this field in the past few decades. Although the THz frequency range still remains underdeveloped as compared to its neighboring microwave and infrared spectral regions, research in this region of the spectrum has undergone significant development over the last two decades, and the introduction and application of new materials such as graphene [12–14] and metamaterials [15–17], together with the development of novel devices such as the quantum cascade laser (QCL) [18–20], and the THz quantum well infrared photodetector (THz QWIP) [21], are bringing THz technologies closer to more widespread application.

Figure 1-1. The ‘Terahertz gap’ in the electromagnetic spectrum lurking between the microwave and mid-infrared regions.

1.1.1 THz applications

Most of the diverse THz applications fall into the following three broad categories:

1. Spectroscopy

Spectroscopy is the primary and oldest application of the THz technologies. It has been realized early last century that many molecules have their spectral signatures in the THz region, corresponding to some molecular rotations, vibrations and atomic fine-structure transitions. This fact attracted particular interest from the astronomers because, with THz spectroscopy, they could be able to study the chemical composition of a distant stellar body or the interstellar dust, of which a representative spectrum content in the THz band is shown in Fig. 1-2. Here, the technical challenge is more related to the THz detection, but frequency-tunable narrow-linewidth THz sources also play an important role as they enable high spectral resolution heterodyne THz detection when used as local oscillators.

Besides the heterodyne detection, Fourier Transform Far-infrared Spectroscopy (FT-FIR) and THz Time-Domain Spectroscopy (THz-TDS) are two other widely adopted techniques. While the former was developed much earlier (1961) [22,23], the latter (1990s) is advantageous in that it is capable of measuring directly the oscillating electric waveform via electro-optic sampling. This enables a much higher accuracy in estimating the complex refractive index of the material, which gives various parameters as a function of frequency, such as the dielectric constant, conductivity and surface impedance [23]. However, THz-TDS is limited by the availability of broadband coherent THz sources. THz spectroscopy has been applied to numerous materials in different applications, such as environment monitoring, explosive and drug inspection, pharmaceutical sciences, material analysis and plasma fusion diagnostics, among many others. Specifically, the THz spectroscopic study of biomolecules, proteins and DNA reveals their intermolecular vibrations, which will elucidate the dynamic of large biomolecules, and thus the knowledge of the human body [24]. Characterization of semiconductor wafers in terms of carrier mobility, conductivity, carrier density and plasma oscillation is also a field of interest for THz spectroscopy [25,26]. Additionally, in basic science, the pump and probe THz-DTS has become a powerful and routine approach for studying the ultra-fast non-equilibrium carrier processes, which can be traced, by the THz-DTS, with a time resolution better than 10 fs [27–29].

Figure 1-2. Radiated energy versus wavelength showing 30-K blackbody, typical inter-stellar dust, and key molecular line emissions in the terahertz (reprinted from [30]).

2. Imaging

THz radiation can penetrate a variety of opaque materials like plastics, polymer, cloth, cardboard, ceramic, pristine or lightly doped semiconductors, etc. It is therefore quite simple to inspect through these materials the concealed features or contents using THz imaging. Microwave is also being used for such tasks, however, imaging with THz wave provides a better spatial resolution thanks to its shorter wavelength. While compared with the X-ray imaging, THz imaging is a desirable alternative simply because it is safer: X-ray is ionizing radiation and is destructive to biological tissues. These attractive aspects of the THz imaging have continuously boosted the development of this field since the first practical demonstration in 1970s [31,32]. Recent advancements include the demonstrations of THz real-time imaging [33,34], confocal microscopy [35], three-dimensional imaging [3,36], high resolution inverse synthetic aperture radar (ISAR) imaging [37], and multi-frequency imaging for spectroscopic mapping of materials [38], etc., all realized with the newly developed THz quantum cascade lasers (QCLs). Among these, one striking example of imaging

with THz QCL is the coherent imaging based on the self-mixing (SM) phenomenon in THz QCLs. In this case, the THz radiation reflected by the imaging target is re-injected back into the laser cavity, causing voltage change on the QCL electrode that depends on both the amplitude and phase of the returning field [39,40]. As such, a single THz QCL functions as both the radiation source and detector.

The spatial resolution of the imaging is dependent on how tightly the THz beam can be focused. For traditional far-field imaging, typical resolution is in the millimeter level. However, several THz microscope systems have been demonstrated with much better resolutions. Fig. 1-3 shows an example of the THz microscope and the taken THz image of a fresh leaf, which indicates excellent image contrast and abundant details. In near-field imaging, thanks to the nanoscale field concentration at sharp metallic tips, a THz near-field nanoscope has been realized with 40 nm resolution at 2.54 THz [41].

Figure 1-3. (a) Schematics of the THz microscope setup. QCL: a 2.9 THz quantum cascade laser; L1, L2, L4: Picarin lenses; S1, S2: Silicon lenses; TPX: TPX lens; M2. (b) A photograph of a fresh leaf. (c) Confocal THz image of the same leaf. The measurements covered an area of $1.8 \text{ mm} \times 3.2 \text{ mm}$ and 90×160 pixels were recorded. (d) The rectangular white frame in panel (c) is imaged again with higher resolution (200×200 pixels). (Adapted from [35])

3. Communication

High capacity data transmission is essential in this era of information. Over the past decade, communication system has undergone a dramatic increase in the data rate, eventually to today's tens of Gbit/s per channel and ~100 channel per fiber in the commercial optical communication system. Foreseeably, under the double drives of the market demand and research interests, the hunger for even higher capacity data transmission will only increase. However, while optical fibers enable ultra-fast information torrent running between cities and continents, the transmission of the data often becomes problematic in the 'last mile' from the fiber backbone to the client premises (the so-called 'last mile gap' problem in the communication system). For example, sometimes it is not possible or practical to lay down optical fibers, which is also costly and time-consuming, and the currently used radio frequency wireless communication has limited bandwidth (up to 150 Mbit/s for 5G WiFi), limited by the frequency of the carrier wave. Therefore, extending the operation frequencies of the wireless communication towards THz region will be inevitable for faster data transmission. In preparation for this, IEEE 802.15 has established a THz Interest Group in 2008 to explore the feasibility of the THz wireless communication and to develop standards [10,42,43]. To date, many prototypical THz links has been demonstrated at frequencies ranged from 300 GHz to 4 THz [11,44–49], although tremendous hurdles are still to be overcome in the way to popularizing the technology [10].

1.1.2 THz radiation sources

The development of THz areas has been hindered by the long lack of compact, powerful, easy-to-use and low cost sources. This is because that electronic devices can hardly operate at the THz range due to the limitation on carrier response time, and that appropriate materials with sufficiently small bandgaps are not available. Therefore, such a THz source has been a highly sought-after goal in the research community. Currently, the approaches to generate THz radiation can categorized as the following four kinds:

1. Frequency multiplication of microwave

In the lower end of the THz spectrum, i.e., 0.3 - 2 THz, THz radiation can be obtained by upconverting the microwave source (with frequency below 100 GHz) using solid-state frequency multipliers (typically a Schottky diode or diode array). THz wave generated in this way benefits from the fact that it inherits the advantageous properties of the very mature microwave seed source. For instance, the resultant THz wave is phase-lockable and frequency tunable. Moreover, after more than 60 years development, multiplier chains are robust, relatively compact (as compared to the free electron based sources) and capable of room-temperature operation, albeit the typically less than 1% conversion efficiency. The current state-of-art devices can produce microwatt-level output power at 2.7 THz [50] and higher at lower frequencies. Although typical power is measured in microwatts, the above-mentioned advantages make these multiplier sources the devices of choice as local oscillators of heterodyne receivers for many years, especially for applications in astronomy [51]. However, pushing the oscillator frequency up above 2 THz is still very challenging, intrinsically limited by the transit time of the carriers (electron or hole) in the electronic solid-state devices.

2. Nonlinear optical process

Two types of nonlinear optical processes involve in the THz generation: optical rectification and difference frequency generation (DFG). The former is usually employed to generate ultra-fast THz pulses, where intense femtosecond (fs) pulses are incident on a photoconductive antenna or a crystal. In the photoconductive antenna case, when the gap between the closely spaced electrodes is illuminated by the fs pulses, carrier will be generated in the photoconductor (e.g. LT-GaAs, onto which the antenna is printed), and then accelerated by an applied bias field ($\sim 100\text{V}$). The current transient thus generated features a frequency comb with peaks separated by the repetition rate of the fs pulses, which is widespread into the THz region. Coupling to the antenna, this current radiates nanosecond broadband THz pulses into free space. Enabling coherent detection and easy implementation, the THz photoconductive antennas are widely used as the source in THz Time Domain Spectroscopy. However, limited by transit time of the carrier through the gap, the frequency can hardly go up above 3 THz and the power conversion efficiency is typically $\sim 10^{-4}$. Optical crystal has no such frequency limitation due to the absence of any electronic component. Utilizing the large second-order susceptibility in crystals like ZnTe, DAST, ultra-short

(a few optical cycles) THz pulses can be produced due to a wider spectral range upon fs laser illumination. The conversion efficiency is also much higher (~20%). Nevertheless, the meticulous optical configuration that is inevitable to achieve phase matching somewhat limits its practical applications.

In contrast to the broadband spectrum of the THz pulse generated by optical rectification, the DFG method can produce narrow band THz radiation, either continuous wave (CW) or pulses. In this case, two infrared laser beams (instead of a single fs laser in previous case), whose frequencies are slightly offset with the difference falling into the THz range, propagate in a nonlinear material (e.g. DAST, GaP, LiNbO₃, GaSe, etc.). THz radiation is generated by difference-frequency mixing of the two lasers resulted from the crystal's second-order susceptibility. Large wavelength tunability is obtainable by tuning the frequency of one or both of the pump lasers. Again, phase match condition should be satisfied and the THz power (milliwatt) is limited by the relatively low conversion efficiency ($10^{-4} - 10^{-9} \text{ W}^{-1}$) [52–54]. Recently, photoconductive antennas were also used for DFG in THz region to produce CW or quasi-CW THz radiation [55–57]. This kind of THz source is promising given the wide frequency tunability and room-temperature operation. However, low conversion efficiency is still a major limitation.

3. Optically pumped molecular-gas lasers

Before the recent boom of THz quantum cascade lasers, optically pumped molecular-gas lasers were the only commercialized and viable CW terahertz lasers for many practical use. The low-pressure gas medium is usually pumped by a CO₂ mid-infrared laser, population inversion can be achieved between certain rotational/vibrational states at room temperature. Depending on the gas used, discrete lasing lines can be obtained between 0.1 and 8 THz [58]. CW power is typically 1-50 mW below 3 THz but becomes much lower at higher frequencies. Perhaps the most successful example of this type of lasers has been the ones with methanol gas. It can emit tens of mW power at 2.5 THz and has been used on a long-duration space mission on NASA's AURA satellite that monitors the chemistry and dynamics of the earth's atmosphere [59]. Despite all these, such gas lasers are still expensive, bulky and power consuming, making them far less than ideal THz sources.

4. Semiconductor THz lasers

Electrically pumped semiconductor THz lasers, ideally of room-temperature operation, high optical output power and reasonable electrical power consumption, have been long pursued by the researchers. Although people are still on the road to this goal, the p-Ge hot-hole laser and quantum cascade laser mark two milestones.

The p-Ge lasers was invented in 1984. Population inversion could be obtained between the light and heavy hole bands [60,61] or between two light hole Landau levels [62], when subjected to crossed electric and magnetic fields (0.3 – 2 T) at low temperature (4 – 40 K). More recent developed strained p-Ge lasers eliminate the need of magnetic field, but a pressure of several kbar is necessary [63]. In these devices, the applied strain breaks the degeneracy of the light hole and heavy hold band and brings the 1s impurity state of the heavy hole into resonance with the light hole band such that a population inversion could be achieved between the heavy hole 1s state and the light hole impurity states, which are depopulated by electric field ionization. Peak power up to 10 W could be obtained. However, the low efficiency of the laser, as compared with the high electrical power input, usually renders the laser of pulse operation, with a duty cycle up to 5% (typical values are less than 10^{-4}) [64]. Continuous-wave lasing was also reported in small-volume devices at liquid helium temperature. Nevertheless, this was accompanied by a low emitting power (up to several tens of μW) [65]. Due to the stringent requirements for achieving lasing, i.e. high voltage (hundreds of volt), very low operation temperature (< 40 K), magnetic field or strain, the p-Ge laser is of limited research interests. The research for new THz semiconductor laser continues.

A breakthrough occurs in 2002, when Köhler et al. reported the first THz quantum cascade laser (QCL) [66]. A brief review will be given bellow.

1.2 THz QUANTUM CASCADE LASERS

In conventional laser diodes, radiative transition take place between the conduction band and the valence band across the material bandgap. However, this interband transition scheme cannot be extended to the far-infrared region as suitable materials with narrow enough bandgaps are not available. Therefore, researchers had been seeking the possibility of achieving radiative transition between the quantized states (subbands) within the semiconductor conduction/valence band in quantum well

structures, typically superlattices. The process was termed as intersubband transition, based on which intersubband light amplification in superlattice were proposed as early as 1971 [67], followed by several improved proposals in 1980s [68,69]. However, the initial schemes (Fig.1-4) are not suitable for practical implementation because: 1. The structure is unlikely to obtain a population inversion due to the difficulty in selectively extract the electrons from level 2 without depopulating the upper laser state 3, and the fast nonradiative transition paths from level 3 to 2, 1 (mainly via the sub-picosecond LO-phonon scattering); 2. The device tend to operate in the negative-differential resistance (NDR) region, and its associated electrical instability prevents the device from being biased at the correct point.

Figure 1-4. A portion of the band diagram of an initially proposed intersubband laser, formed by cascading multiple transition modules. Radiative transitions take place between level 3 and level 2, the electrons then undergo fast relaxation to level 1 and are re-injected into next module.

It was not until 1994 that the first intersubband laser, named quantum cascade laser (QCL), was demonstrated by Faist *et al* at Bell Labs [70]. Fig. 1-5 shows a portion of the conduction band diagram of the device. 25 periods/modules were cascaded, each of which consists of an injector (the ‘digitally graded alloy’ in the figure) and an active region, formed by $\text{Al}_{0.48}\text{In}_{0.52}\text{As}/\text{Ga}_{0.47}\text{In}_{0.53}\text{As}$ heterostructures. The injector is actually a superlattice with constant period but increasing $\text{Al}_{0.48}\text{In}_{0.52}\text{As}$ duty cycle so that a flat effective band profile could be obtained when the required voltage is applied. Electrons are injected, through resonant tunneling, from the injector into the upper laser state (level 3), and then undergo the radiative, as well as non-radiative, transitions to level 2, which is fast depopulated by an underneath level 1 via LO-phonon scattering. The electrons are then cooled down in the injector and re-used in the next module. In this way, one electron is able to generate multiple

photons thanks to the cascade scheme. It is worth mentioning that another important function of the injector is that it eliminates the NDR region before laser threshold so that the device could be homogeneously and stably biased.

This first QCL operated in the Mid-infrared region at a wavelength of 4.2 μm . The relatively low optical gain resulted in a high threshold current density of $\sim 14 \text{ kA/cm}^2$ and restricted the laser to pulsed operation with a maximum operation temperature of $\sim 90 \text{ K}$. However, rapid development ever since has made the MIR QCLs practical sources for real-world applications, which are now commercially available. Through band engineering, the MIR QCLs have covered the MIR spectral range from 3 – 24 μm . Most of them now operate at room temperature and watt level cw output power can be routinely produced for 3-6 μm .

Figure 1-5. Schematic band diagram of the first quantum cascade laser operated at 4.2 μm , including the quantized electron states and the magnitude squared of the wavefunctions. Each period/module of the structure consists of an injector (the digitally graded alloy) and an active region. (Reprinted from [70])

Despite the success in MIR region, the development of such a device in the THz region turned out to be much more difficult. The first problem is related to the difficulties in selectively injecting the electrons into the upper laser state and selectively extracting them from the lower laser state as the two involved states are very closely spaced in energy ($\sim 10 \text{ meV}$). Another problem is due to the high loss in conventional laser waveguide because the free carrier absorption scales with

wavelength as λ^2 . In addition, the traditional optical mode confinement scheme, where the active core medium is sandwiched by two cladding layers with lower refractive index, is no longer suitable for the THz waves as it would require unreasonably thick core and cladding layers due to the long wavelength, which are not compatible with the growth techniques (MBE or MOCVD). Therefore, novel waveguide structure, that can tightly confine the optical in the active region, is an additional necessity for THz QCL. These problems explain why the THz QCL (2002) came almost 8 years after the demonstration of the first MIR QCL [18], although intrasubband light emission from quantum cascade structure was firstly observed in the THz region in 1989 [71]. In the first THz QCL, ~ 2.5 mW peak power was observed at 4.4 THz in pulsed mode at 8 K, and device worked up to 50 K. The band structure of this THz QCL is shown in Fig. 1-6(a). Optical transition takes place between level 2 and 1. The adoption of the chirped superlattice active region ('Active SL') makes the lower laser miniband flat at the operation voltage, which allows efficient electron depletion of the lower laser state 1. At the same time, electrons are fast funneled into the upper laser state 2 from the lower miniband through the ground states g . Population inversion can thus be obtained. Laser action was further facilitated by the novel waveguide design, what was later called 'semi-insulating surface-plasmon waveguide'. As shown in Fig. 1-6(b), they used a semi-insulating GaAs substrate to reduce the free carrier absorption in the substrate. A thin n^+ GaAs layer, with carefully chosen doping level, was inserted between the waveguide core and the substrate, resulting in a remarkable 47% modal overlap with the waveguide core while maintaining a low loss. This semi-insulating surface-plasmon waveguide and the later developed metal-metal waveguide are the two dominant waveguide used today in THz QCLs. In metal-metal waveguide, the active region is sandwiched by two metal plates, leading to near 100% modal confinement and reduced loss. It therefore has a better temperature performance (higher maximum operation temperature), but suffers from relatively low output power and extremely large beam divergence ($>180^\circ$).

Figure 1-6. (a) Conduction band diagram of a portion of the layer stack of the first terahertz quantum cascade laser. The magnitude squared of the wavefunctions are shown, with miniband regions represented by shaded areas. Optical transition takes place between level 2 and 1. (b) Optical mode profile along the growth direction of the device. The waveguide core of 104 repetitions of the active region shown in (a) is between the bottom contact and top metal layer. (Reprinted from [18])

The advent of the first THz QCL spurred intensive research interests in this field. To date, THz QCLs have covered the spectral range 1 – 5 THz. Maximum operation temperature has been pushed to 200 K [72], and efforts are under way to lift it further, at least to the temperature achievable by thermoelectric cooling (240 K). Fig. 1-7 summarizes maximum operation temperatures of THz QCLs in the literature as a function of frequency. With regard to output power, a single THz QCL could provide 1 W peak power in pulsed mode [20] and over 130 mW in cw [73] at 10 K heatsink temperature. In addition, various resonator designs (e.g. DFB grating [74], photonic crystal [75], disk [76], etc.) and beam engineering techniques [77] have been

implemented with THz QCL, enhancing the laser performance in terms of spectral purity or beam quality. Frequency tunable devices have also been demonstrated [78–81], among which the most efficient tuning scheme is to manipulate the transverse wave vector of a wire laser, achieving a tuning range of 330 GHz (8.6% of the center frequency) [81]. Moreover, to control the THz QCL frequency with extremely high accuracy for metrological applications, researchers have phase locked THz QCLs to ultra-stable microwave references [82–84]. In addition, active mode locking of THz QCLs was demonstrated by direct modulating the applied voltage at the round-trip frequency (10 – 30 GHz) [85,86]. More recently, frequency combs were generated directly from THz QCLs that employed a chirped distributed-feedback reflector (DBR) to cancel out the cavity dispersion [87]. Such THz QCLs are ideal candidates for making compact spectrometers. In addition to these THz QCLs that directly generate THz radiation, THz sources based on intracavity DFG in mid-infrared QCLs demonstrated more recently are also very appealing as they are capable of room-temperature operation and delivering up to 1.5 mW peak power at 3.5 THz [19,88]. Moreover, extremely wide tuning range from 1.2 THz to 5.9 THz has been demonstrated in an external-cavity device [89]. Nevertheless, the work of this thesis will be focused on the THz QCLs that directly produce THz radiation.

Figure 1-7. Maximum operation temperatures of THz QCLs as a function of frequency in the literature. The inset shows the timeline of the maximum temperatures. (Reprinted from [90])

1.3 MOTIVATIONS AND CHALLENGES

THz QCLs have emerged as a compact semiconductor source of coherent THz radiation since their first demonstration in 2002, and have promise for exploitation in THz applications such as spectroscopy, imaging and communication that have been detailed in Section 1.1.1.

In spectroscopy, a broad gain width of the THz QCL is highly desirable. However, active regions that feature a broader gain width, such as the bound-to-continuum design, typically have worse temperature performance compared to the resonant-phonon design. The challenges of achieving high temperature performance in these designs lie in the difficulty in selectively injecting the electrons into the upper laser state that are very close in energy to the lower laser state, and the thermally activated LO-phonon scattering of the electrons from the upper laser state, as well as the thermal backfilling of the lower laser state.

For many of the THz applications, high power single-mode lasers accompanied by low beam divergence are highly desirable. However, owing to the lack of a mode-selection mechanism, and the sub-wavelength mode confinement inherent particularly to metal-metal waveguides, conventional edge-emitting ridge-waveguide THz QCLs suffer from multi-mode operation and an extremely wide beam divergence ($>180^\circ$). Moreover, the strong mode confinement in the laser cavity leads to a large impedance mismatch between the modes inside and outside the cavity, resulting in a low output power. Furthermore, for some applications, more complex radiation beams in terms of polarization and beam shape may be required. For example, circular polarization is important for detecting molecules that exhibits circular dichroism, radially polarized light source is desirable for near-field imaging, and hollow beam is beneficial for high-resolution microscopy. Therefore, flexible beam engineering techniques are required.

In addition, modulation of THz wave is often needed, e.g., to encode information in the THz wave, to performance lock-in amplification, and to achieve active mode locking, etc. However, the development of fast and efficient THz modulators is in its infancy. Traditional two-dimensional electron gas (2DEG) can only modulate the THz light intensity by only a few percent, although through the incorporation of metamaterials or plasmonic structures that enhanced the interaction between the THz

radiation and the 2DEG, a modulation depth of 30% was achieved. This small modulation depth is limited by the achievable tenability in carrier concentration, which is up to $\sim 1 \times 10^{12} \text{ cm}^{-2}$ for a 2DEG in conventional semiconductor. Recently, graphene, a monolayer of carbon atoms arranged in a honeycomb lattice, was found to be more efficient in modulating THz light, and a 64% modulation depth has been obtained. However, further increase of the modulation depth has proven difficult. Moreover, all these THz modulators are suffering from a slow operation speed, up to $\sim 13 \text{ MHz}$.

1.4 OBJECTIVES AND METHODOLOGIES

Aiming at achieving a broad gain width of the THz QCL with better temperature performance, we analyze the limitations of the existing active region designs of THz QCL, and proposed a novel bound-to-continuum phonon-photon-phonon design that combines the advantages of the indirectly pumped scheme and the phonon-photon-phonon design.

To overcome the drawbacks associated with the ridge-waveguide THz QCLs, we design and investigate a novel concentric-circular grating based resonator. Single-mode surface-emitting THz radiation with low beam divergence is expected.

Moreover, to control the THz wave in a more flexible way, a metasurface based surface-plasmonic waveguide is studied and integrated with THz QCL to provide a versatile platform for THz beam engineering and active THz photonic circuit.

Finally, to efficiently and fast modulate the THz wave from QCL, we develop an integrated graphene modulator, improved performance in terms of modulation depth, modulation speed is expected as a result of a greatly enhanced interaction of the graphene with the THz laser field, enabled by the intimate integration scheme.

1.5 THESIS OVERVIEW AND MAJOR CONTRIBUTION

This thesis summarizes the main works that have been done in the course of my PhD project. Chapter 2 discusses the fundamental working principles of the THz QCL, followed by a comparison between different kinds of the existing active region designs, after which a novel design is proposed with some preliminary experimental results. In this part, I designed the band structure of the active region, which was grown by Leed University in UK, fabricated and characterized the devices independently. Chapter 3 reports a surface-emitting resonator design for THz QCLs, based on distributed

feedback concentric-circular grating. Robust single-mode operation has been achieved with directional optical emission. The detailed design, fabrication and experimental characterization will be presented. In this part, I performed the design and numerical simulations of the structures, as well as all the fabrication, characterization and result analysis under the guidance of my supervisor. The Leed University provided the active region. In chapter 4, metasurfaces are integrated with THz QCLs in a planar way, highly collimated THz beam can thus be obtained with a divergence as narrow as $\sim 4^\circ \times 10^\circ$. The scheme also provides a promising platform for constructing active THz photonic circuit by incorporating other optoelectronic devices such as Schottky diode THz mixers or graphene modulators and photodetectors. In this part, I designed the structure, performed the electric field simulations, fabricated the devices and conducted the characterization. The active region used was grown by University of Waterloo in Canada. Chapter 5 describes an integrated graphene modulator with THz QCLs. A 100% modulation depth has been achieved as a result of strong interaction between the laser and the graphene. In this part, I designed and fabricated the devices except the active region growth, and carried out all the experimental measurements and result analysis. Finally, Chapter 6 summarizes the work with proposals for future researches.

2. ACTIVE REGION DESIGN OF TERAHERTZ QUANTUM CASCADE LASERS

In this chapter, the fundamental theories behind the THz QCL will be discussed as the first part, followed by a comparison of the major active region designs. Finally, a design based on indirectly-pumped bound-to-continuum scheme is proposed, and some preliminary experimental results are presented.

2.1 THEORY

2.1.1 Subbands in quantum wells

Quantum wells are formed when nanoscale thin semiconductor layers are sandwiched by barrier materials with higher bandgaps. Electron motion is then restricted in the growth (z-) direction, and energy quantization occurs when the restriction is on the order of the electron's DeBroglie wavelength. Specific to the THz QCL in this thesis, the materials involved are AlGaAs and GaAs for barrier and well, respectively. To calculate the electron wavefunctions and energy states in such a multiple quantum well (MQW) system, the Schrödinger equation is solved in the framework of the envelope function approximation and effective mass equation, where the electron wavefunction is given by [91]

$$\psi(\vec{r}) = \psi_n(\vec{r}) u_{n,\vec{k}}(\vec{r}) \quad (2-1)$$

where $u_{n,\vec{k}}(\vec{r})$ is the periodic Bloch state wavefunction associated with the periodic crystal atoms, and $\psi_n(\vec{r})$ is the slowly varying envelope function. As the electrons are free in the in-plane (x-y) directions, $\psi_n(\vec{r})$ can be written as

$$\psi_n(\vec{r}) = \frac{1}{\sqrt{S_{xy}}} e^{i\vec{k}_{\parallel} \cdot \vec{r}_{\parallel}} \psi_n(z) \quad (2-2)$$

where $\vec{k}_{\parallel}$ is the in-plane wavevector, S_{xy} is the cross-sectional area of the quantum-well layer included for normalization, and $\psi_n(z)$ satisfies the following effective mass equation

$$\left[-\frac{\hbar^2}{2} \frac{d}{dz} \frac{1}{m^*(z)} \frac{d}{dz} + V_c(z) \right] \psi_n(z) = E_n \psi_n(z) \quad (2-3)$$

where $m^*(z)$ is position-dependent effective mass, $V_c(z)$ is the conduction band profile, and E_n is the energy of the n -th subband. Including the in-plane kinetic energy, the total energy of an electron in the n -th subband is $E_n(\vec{k}_\parallel) = E_n + \frac{\hbar^2 k_\parallel^2}{2m^*}$. In this Eq. (2-3), we have assumed that the Bloch wavefunction $u_{n,\vec{k}}(\vec{r})$ is the same for the well and barrier materials. This is acceptable for the lattice-match heterostructure (e.g. AlGaAs/GaAs) as the materials involved have the same crystal structure and lattice parameters.

To take into account the Coulomb effect induced by the accumulated electrons, it is often necessary to solve the Poisson equation

$$\frac{d}{dz} \left[\varepsilon(z) \frac{d}{dz} \Phi(z) \right] = -\rho(z) \quad (2-4)$$

where $\Phi(z)$ is the electrostatic potential, $\varepsilon(z)$ is the position-dependent permittivity and $\rho(z)$ is the space charge density, given by

$$\rho(z) = e \left(N_D(z) - \sum_n N_n |\psi_n(z)|^2 \right) \quad (2-5)$$

with $N_D(z)$ being the doping of the material, and N_n the electron density for the n -th subband, determined by

$$N_n = \frac{m^*}{\pi \hbar^2} k_B T \ln \left(1 + e^{\frac{E_f - E_n}{k_B T}} \right) \quad (2-6)$$

where E_f is Fermi energy. The conduction band profile in Eq. (2-3) is then given by

$$V_c(z) = V_{c,0}(z) - e\Phi(z) \quad (2-7)$$

with $V_{c,0}(z)$ being the intrinsic conduction band profile. Eq. (2-4) and (2-5) indicate that the Schrödinger equation and Poisson equation are coupled. In practice, the solution is obtained by solving the above equations iteratively. The validity of this effective mass model has been extensively studied, and it is found that the model

provides excellent approximation as long as the envelope functions are slowly varying as compared with the Bloch state functions [92].

2.1.2 Intersubband optical gain

Optical gain that resulted from radiative transition between two energy states is essential for laser action. To calculate the optical gain, we will have to investigate the two kinds of radiative transitions in QCL, i.e., transitions via spontaneous emission and stimulated emission of photons.

1. Spontaneous emission

Considering two laser subbands i and j with an energy separation of $\hbar\omega$, the spontaneous emission rate can be derived from the Fermi's golden rule

$$W_{ij} = \frac{2\pi}{\hbar} \left| \langle \psi_i | \hat{H} | \psi_j \rangle \right|^2 \delta(E_i - E_j - \hbar\omega), \text{ which yields [93]}$$

$$W_{ij}^{sp} = \frac{e^2 \omega_0^3 n^3}{3\pi c^3 \hbar \epsilon_0} |z_{ij}|^2 \quad (2-8)$$

where n is the refractive index, ω is the angular frequency of the photon, c is the speed of light in vacuum, ϵ_0 is the permittivity of vacuum, and z_{ij} is the dipole matrix element, defined as $z_{ij} = \langle \psi_i | z | \psi_j \rangle$.

For intersubband transition at THz frequency, the spontaneous lifetime τ_{ij}^{sp} , which is the inversion of the spontaneous emission rate, is much longer ($>1 \mu s$) than that of shorter wavelength as a result of the scaling factor ω_0^3 to the emission rate. The very long spontaneous lifetime, as compared to the picosecond non-radiative lifetime, renders the spontaneous radiation very inefficient in THz QCL, with an efficiency of $10^{-7} - 10^{-6}$. Therefore, spontaneous emission plays neglecting role in carrier transport in THz QCL, and it is difficult to make a THz light emitting diode based on intersubband transition.

2. Stimulated emission

The stimulated emission rate is related to the light intensity $I(\nu)$ in the medium, expressed as [93]

$$W_{ij}^{st} = W_{ij}^{sp} \frac{\eta \lambda^2 I(\nu)}{8\pi h \nu n^2} L(\nu) \quad (2-9)$$

where ν is the frequency and

$$L(\nu) = \frac{\Delta\nu / 2\pi}{(\nu - \nu_0)^2 + (\Delta\nu / 2)^2} \quad (2-10)$$

is the Lorentzian lineshape with

$$\Delta\nu = \frac{1}{\pi T_2} = \frac{1}{\pi} \left(\frac{1}{2\tau_i} + \frac{1}{2\tau_j} + \frac{1}{T_2^*} \right) \quad (2-11)$$

being the linewidth of the transition centered at ν_0 . Here, T_2 is the total phase breaking time, τ_i and τ_j are the lifetimes of the initial and final states, and T_2^* is the pure dephasing time. The parameter $\eta = 3|z_{ij}|^2 / |\vec{r}_{ij}|^2$ describes the interaction of the incident electric field with the dipole. As the all the intersubband dipoles are oriented in the z- direction, in contrast to the randomly oriented interband dipoles, $\eta = 3$.

3. Optical gain

The induced optical power density due to the radiative transition between subbands i and j can be written as

$$P = (N_i W_{ij}^{st} - N_j W_{ji}^{st}) h\nu = h\nu W_{ij}^{st} (N_i - N_j) \quad (2-12)$$

where we have used $W_{ji}^{st} = W_{ij}^{st}$. If there is a population inversion between the two laser states, i.e., $\Delta N = N_i - N_j > 0$, the intensity of the light will get amplified as it propagates in the active region

$$\frac{dI(\nu)}{dz} = g(\nu) I(\nu). \quad (2-13)$$

Derived from the above two equations, the bulk gain per unit length can be written as

$$g(\nu) = \frac{\Delta N e^2 \pi \nu_0 |z_{ij}|^2}{\hbar c n \varepsilon_0} L(\nu) \quad (2-14)$$

where ΔN is the bulk population inversion density in the unit of cm^{-3} , and it is related to the sheet population inversion density ΔN_{sh} as $\Delta N = \Delta N_{sh} / L_{\text{mod}}$, where L_{mod} is the length of an active region module. From Eq. (2-14), we can get the gain peak for the transition:

$$g_c = g(\nu_0) = \frac{2e^2\nu_0}{\hbar cn\epsilon_0} \frac{\Delta N |z_{ij}|^2}{\Delta\nu} = \frac{e^2}{2\pi m^* cn\epsilon_0} \frac{\Delta N f_{ij}}{\Delta\nu} \quad (2-15)$$

$$\approx 70 \times \left(\frac{\Delta N}{10^{15} \text{cm}^{-3}} \right) \left(\frac{f_{ij}}{\Delta\nu / \text{THz}} \right) \text{cm}^{-1} \quad (\text{for GaAs})$$

where f_{ij} is the oscillator strength, a unitless parameter defined as

$$f_{ij} = \frac{2m^*(E_j - E_i)|z_{ij}|^2}{\hbar^2}.$$

2.1.3 Intersubband relaxations

Based on the rate equations model of the QCL, the sheet population inversion density can be expressed as

$$\Delta N_{sh} = \frac{J_{iu}}{e} \tau_u \left(1 - \frac{\tau_l}{\tau_{ul}} \right) \quad (2-16)$$

where J_{iu} is the injected current density to the upper laser state, τ_u and τ_l are the total lifetimes of the upper and lower laser states, respectively, and τ_{ul} is the scattering time from upper laser state to lower laser state. Obviously, in order to achieve a large population inversion for laser action, it is important to let τ_u and τ_{ul} be as long as possible and τ_l as short as possible. This requires knowledge on the nonradiative relaxation mechanisms of the electrons in the system. As shown in Fig. 2-1, the intersubband electron relaxation paths include scattering with longitudinal optical (LO) phonon, another electron (e-e scattering), ionized impurity (ii), interface roughness (ir) and acoustic phonon (ac), etc.

1. Electron-LO-phonon scattering

When the energy separation of the two involved subband ΔE is close to the LO phonon energy ($\hbar\omega_{LO} \sim 36$ meV in AlGaAs/GaAs material system), the LO phonon scattering process is ultrafast, happens within subpicosecond. Therefore, most of the new active region designs of QCLs employ LO phonon scattering as an efficient mechanism to deplete the lower laser state, i.e., to obtain an short τ_u . LO phonon scattering also play important role even when the two subband are closely spaced with $\Delta E < \hbar\omega_{LO}$, e.g., between the upper and the lower laser state. In this case, LO phonon scattering takes place from the hot tail of the upper laser state to the bottom of the lower laser state. This process is named thermally activated LO phonon scattering, and it limits the highest operation temperature of THz QCLs.

Figure 2-1. Schematic view of the intersubband relaxation paths. LO: LO-phonon scattering; e-e: electron-electron scattering; ii: ionized impurity scattering; ir: interface roughness scattering; ac: acoustic phonon scattering.

Between subbands i and j , the electron-LO-phonon scattering rate can be expressed as [94]

$$W_{ij}^{LO} = \frac{m^* e^2 \omega_{LO}}{8\pi \hbar^2} \left(\frac{1}{\epsilon_{\infty}} - \frac{1}{\epsilon_{dc}} \right) n_{\omega_{LO}} \int_0^{2\pi} B_{ij}(q_{\parallel}) d\theta, \quad (2-17)$$

where ε_∞ and ε_{dc} are, respectively, the high-frequency and static permittivity of the material, $n_{\omega_{LO}}$ is Bose-Einstein occupation, and $B_{ij}(q_\parallel)$ is defined as

$$B_{ij}(q_\parallel) = \int_{-\infty}^{\infty} dz \int_{-\infty}^{\infty} dz' \psi_j^*(z) \psi_i(z) \psi_i^*(z') \psi_j(z') \frac{1}{q_\parallel} e^{-q_\parallel |z-z'|} \quad (2-18)$$

where

$$q_\parallel = |\vec{k}_i - \vec{k}_j| = \sqrt{k_i^2 + k_j^2 - 2k_i k_j \cos \theta} \quad (2-19)$$

is the in-plane phonon wavevector, with θ being the angle between the in-plane wavevector $\vec{k}_i$ and $\vec{k}_j$ (Fig. 2-2).

Figure 2-2. (a) Electron-LO-Phonon scattering between two subbands with in-plane momentum exchange. (b) Relationship between the wavevectors of the electron before and after transition and the in-plane phonon wavevector.

The total scattering time from subband i to j is then given by averaging over all possible initial states in the subband

$$\frac{1}{\tau_{ij}^{LO}} = \frac{\int f[k_i] W_{ij}^{LO}(k_i) dk_i}{\int f[k_i] dk_i} \quad (2-20)$$

where $f[k_i]$ is the distribution function of the in-plane wavevector.

2. Electron-electron scattering

Unlike the scattering with LO phonon, the electron-electron scattering is an elastic process. The electron energy is conserved as a whole, but the energy distribution of the electron is thermalized. The electron-electron scattering process can be very fast when

energy states are closely spaced, especially in the miniband. The first THz QCL design used this property to depopulate the lower laser state to achieve the population inversion. Electron-electron scattering is a sophisticated many-body problem and a complete description is difficult. Nevertheless, a simplified static single-band-approximation model has been developed by Smet et al [95]. Considering an electron in subband i scattering with another electron in subband j , after which they fall into subbands f and g , respectively, the interaction Hamiltonian reads

$$H'_{ij,fg}(\vec{k}_i, \vec{k}_j, \vec{k}_f, \vec{k}_g) = \frac{2\pi e^2}{S\varepsilon} A_{ij,fg}(q_{\parallel}) \delta(\vec{k}_f + \vec{k}_g - \vec{k}_i - \vec{k}_j) \quad (2-21)$$

where

$$A_{ij,fg}(q_{\parallel}) = \int_{-\infty}^{\infty} dz \int_{-\infty}^{\infty} dz' \psi_i(z) \psi_f^*(z) \psi_j(z') \psi_g^*(z') e^{-q|z-z'|} \quad (2-22)$$

$$q_{\parallel} = |\vec{k}_i - \vec{k}_f| \quad (2-23)$$

The scattering rate of the electron in subband i can be obtained by substitute Eq. (2-21) into the Fermi golden rule, which gives

$$\begin{aligned} W_{ij,fg}(\vec{k}_i) = & \frac{e^4}{2\pi\hbar\varepsilon^2} \int d^2\vec{k}_j \int d^2\vec{k}_f \int d^2\vec{k}_g \frac{|A_{ij,fg}(q_{\parallel})|^2}{\varepsilon_{sc}^2(q_{\parallel}, T) q_{\parallel}^2} f_{j, \vec{k}_j} (1 - f_{g, \vec{k}_g}) (1 - f_{f, \vec{k}_f}) \\ & \times \delta(E_f(\vec{k}_f) + E_g(\vec{k}_g) - E_i(\vec{k}_i) - E_j(\vec{k}_j)) \delta(\vec{k}_f + \vec{k}_g - \vec{k}_i - \vec{k}_j) \end{aligned} \quad (2-24)$$

where $\varepsilon_{sc}^2(q_{\parallel}, T)$ is the effective dielectric constant and the function f_j represents the possibility of occupation of the state j by an electron. Among various kinds of the electron-electron scattering, it is found that the 22,11 and 22,21 processes (Fig. 2-1) are the most efficient, with typical scattering time ~ 100 ps for an individual scattering event. However, in present of a large phase space provided by, e.g., a miniband, the total scattering time of an electron can be subpicosecond. Apart from this complex model, a much simplified model has been proposed by Hyldgaard *et al* [96], which gives

$$W_{e-e} \propto \frac{N_2}{E_{21}} |U(q_{\parallel})|^2 \quad (2-25)$$

where N_2 is the upper state population, and $U(q_{\parallel})$ is a form factor similar to Eq. (2-22). While one cannot rely on this equation to accurately calculate the electron-electron scattering rate, it provides an intuitive understanding of the scattering behavior: the scattering rate is proportional to the electron density and inversely proportional to the subband energy separation.

Other types of scattering (e.g. ionized impurity, interface roughness and acoustic phonon) are much less sufficient and hard to control, they are therefore not considered during the active region design but are used for result analysis.

2.1.4 Resonant tunneling transport

Quantum cascade lasers rely heavily on the resonant tunneling of electrons through potential barriers. Besides the radiative and nonradiative transitions described above, resonant tunneling is another important electron transport in QCL. Two energy states are in resonance when they are brought into aligned in energy. The interaction between the two states will split the aligned states into doublets, with a minimum energy splitting of Δ_0 , which represents the interaction strength between the two states and is known as ‘anticrossing gap’. The resulting wavefunctions of the doublets are spatially extended across the quantum wells (Fig. 2-3(b)). Carrier transport is then described as electrons scattered into and out of the doublets from the preceding state (2") and into the subsequent state (1"), as illustrated in Fig. 2-4(a). In this picture, the electron transport is considered to be coherent as they preserve their phases across the barrier. The current density can be written as [97]

$$J = \frac{eN_A}{\tau_A} + \frac{eN_S}{\tau_S} \approx \frac{eN}{2\tau} \quad (2-26)$$

where $\tau^{-1} = \tau_A^{-1} + \tau_S^{-1}$ and the last term is valid if $N_A \approx N_S = N/2$ with N being the total electron sheet density of the two states, which is true for strong coupling (e.g., $\Delta_0 > 3\text{meV}$).

However, this coherent-tunneling picture fails when the coupling is weak (small Δ_0) through a thick barrier. In this case, the tunneling process is easily dephased by the interface roughness scattering and ionized impurity scattering, etc. The tight-bonding model is more suitable for this case (Fig. 2-4(b)). In this picture, the wavefunctions of

the two resonant states are localized within their individual quantum well and no splitting of energy state occurs, and the coupling strength is represented by $\Delta_0/2$. The electrons will take half of the so-called Rabi period $\hbar/\Delta_0$ to transport through the barrier from one well to the other. The current density can be estimated by

$$J \approx \frac{eN}{2\tau} \left(1 - \frac{1}{1 + \left(\frac{\Delta_0}{\hbar} \right)^2 \tau \tau_{\parallel}} \right) \quad (2-27)$$

where $\tau_{\parallel}$ is the dephasing time. As we can see, if the barrier is made thin enough so that Δ_0 is large and $(\Delta_0/\hbar)^2 \tau \tau_{\parallel} \gg 1$, we regain the coherent tunneling.

Figure 2-3. Schematic illustration of energy states and their wavefunctions in adjacent quantum wells before (a) and after (b) bringing into resonance. Resonance of the two states occurs when they are aligned in energy and split into doublets. The minimum energy splitting Δ_0 is known as the anticrossing gap.

Figure 2-4. Different models used to depict the resonant tunneling of the electrons through a potential barrier. (a) In a coherent transport model, the states in resonance split into energy doublets, whose wavefunctions are spatially extended across the quantum wells. (b) In a tight-binding model, the two resonant states are coupled with a strength of $\Delta_0/2$, but their wavefunctions are localized within individual quantum well.

2.2 TYPICAL ACTIVE REGION DESIGNS

From Eq. (2-15) and Eq. (2-16), the optical gain between the upper laser state u and the lower laser state l can be written as

$$\begin{aligned}
 g_c &= \frac{e^2}{2\pi m^* c n \varepsilon_0} \frac{\Delta N f_{ul}}{\Delta \nu} \\
 &= \frac{e}{2\pi m^* c n \varepsilon_0 L_{\text{mod}}} \frac{f_{ul} \tau_u}{\Delta \nu} \left(1 - \frac{\tau_l}{\tau_{ul}} \right) J_{iu} \\
 &= \tilde{g}_c J_{iu}
 \end{aligned} \tag{2-28}$$

where $\tilde{g}_c$ is defined as the gain coefficient and J_{iu} is the current injected to the upper laser state. Obtaining an as-large-as-possible gain coefficient is the goal of any laser active region design. From Eq. (2-28), it is obvious that the gain coefficient $\tilde{g}_c$ is proportional to the oscillator strength f_{ul} and the upper laser state lifetime τ_u while inversely proportional to the emission linewidth $\Delta \nu$. The former two factors are coupled: a larger f_{ul} requires larger spatial overlap of the two wavefunctions, which, however, tends to increase the nonradiative scattering, leading to a reduced upper states lifetime τ_u . In addition to achieving a large product of f_{ul} and τ_u , a good

design should also have a small τ_l/τ_{ul} , i.e., make τ_l as short as possible and τ_{ul} as long as possible.

To these ends, active regions employing different schemes have been reported, and most of them can be categorized into the following four classes (Fig. 2-5): chirped superlattice (CSL), bound to continuum (BTC), resonant phonon (RP) and phonon photon phonon (PPP).

1. Chirped superlattice (CSL)

The CSL design is the very first generation of the THz QCL active region design. An exemplary band diagram is shown in Fig. 2-5(a), where the energy states of the quantum wells that constitutes the superlattice are tightly coupled. Due to the subband broadening the individual energy state cannot be clearly discriminated. Therefore, the closely spaced energy states form minibands. Thanks to the large k space provided by the miniband for electron-electron scattering, electrons in each of the minibands will be fast thermalized and relax to the bottom of the miniband, leaving the top states relatively empty. A population inversion can thus be achieved at the edges of the bandgap (the red and blue states in Fig. 2-5(a)). The design features a vertical optical transition and a large oscillator strength owing to the large spatial overlap between the radiative states. However, this is accompanied by a poor injection efficiency as a relatively large portion of the electrons will be scattered into the lower laser state l , rather than being injected into the upper laser state u . Reduction in τ_u is also expected, especially that the LO phonon scattering is allowed in energy from state u to the lower states of the lower miniband. Additionally, electron thermal backfilling of the state l from the bottom of the lower miniband is another practical issue. These are the major limitations of the CSL design, making the maximum operation temperature of the CSL device below 100 K.

2. Bound to continuum (BTC)

The BTC design solves part of the problems in the CSL design by inserting an additional well before the original superlattice (Fig. 2-5(b)). This quantum well acts as a defect of the superlattice so that a bound ‘defect’ state is presented in the minigap. Optical transition then takes place between this bound state and the top edge of the lower miniband. As the bound state is localized to the left of the superlattice, the optical transition is diagonal, leading to a longer upper state lifetime, albeit a slightly

reduced oscillator strength. The diagonal nature also helps to increase the injection efficiency because the injection states now have less spatial overlap with the lower laser states. As a result, improved performance has been achieved in the BTC design, with a maximum operation temperature up to 160 K [98].

Figure 2-5. Simplified conduction band diagrams and the moduli squared of wavefunctions for typical active region designs of THz QCLs. The upper laser state u and lower laser state l are highlighted in red and blue, respectively. (a) A chirped superlattice design [66]; (b) A bound-to-continuum design [99]; (c) A resonant phonon design [100]; (d) A phonon-photon-phonon design [98]. Two modules are shown.

3. Resonant phonon (RP)

RP is perhaps the most successful design for THz QCL so far. In contrast to the previous designs, it does not rely on electron-electron scattering in a miniband to deplete the lower laser state, but rather, it employs the subpicosecond LO phonon scattering (Fig. 2-5(c)). In this design, the lower laser state l is in resonance with the excited state of an adjacent wide quantum well, whose fundamental state inj is placed

a LO-phonon energy (~ 36 meV) bellow. In this way, the wavefunction of the lower laser state extends into the wide well so that the electrons will be quickly evacuated by the state *inj* through ultrafast LO phonon emission. As the upper laser state remains localized, it has very little overlap with the state *inj* and the parasitic nonradiative relaxation is minimized. The large energy separation between the injection state *inj* and the lower laser state *l* is another boon, because it reduces the thermal backfilling of the electron from the state *inj* to the lower laser state *l*. Unlike the CSL or BTC designs where electrons in the upper laser state can transit radiatively to multiple states of the lower miniband, very few states are involved in the optical transition in the RP design. Therefore, the RP design typically have a narrower gain width $\Delta\nu$ than the other two designs. The effect is twofold, while a smaller $\Delta\nu$ implies a larger peak gain, a wider gain width is desirable for some applications such as broadband tunable laser [78,80], or frequency comb generation in THz QCL [87,101]. Until now, the RP design enables the record working temperature of THz QCL (200 K) [72].

4. Phonon Photon Phonon (PPP)

The bottleneck of the electron transport in the RP design lies in the resonant tunneling from the injection state into the upper laser state through a thick injection barrier. The thick injection barrier is necessary to prevent a large leakage current at low voltage prior to laser threshold when the injection state aligns with the lower laser state. Figs. 2-6(a) and (b) shows the situations when the injection state is aligned with the lower laser state at low voltage and the upper laser state at higher voltage, respectively. The corresponding current densities are also given below the band diagrams. As the lower laser state lifetime τ_l is much shorter than that of the upper laser state τ_u , careful engineering of the two coupling strength (Δ_{il} and Δ_{iu}) is needed to avoid $J_{il} > J_{iu}$, because it will make the device work in the electrically unstable negative-differential resistance (NDR) region, which will make the device fail to lase in reality. To this end, a thick injection barrier is introduced to let Δ_{iu} be ~ 1 meV. However, the resultant weak coupling, i.e., $(\Delta_{il}/\hbar)^2 \tau_l \tau_{\parallel} \sim 1$, makes the current densities sensitive to the dephasing time $\tau_{\parallel}$, which decreases significantly with temperature. This effect limits the high temperature performance of the RP THz QCLs and bound their maximum operation temperature around $\hbar\omega/k_B$.

Nevertheless, this limitation was recently surpassed by Kumar *et al* using a novel indirect pumping (IDP) scheme (Fig. 2-5(d)) [102]. In this design, electrons are not injected directly into the upper laser state u , but to an overhead state i_2 from where the electrons will then be scattered by LO phonons into the upper laser state. As now the injection state i_1 is located more far away from the lower laser state l , $\Delta_{i_1 l}$ is reduced. Therefore, one can use a large $\Delta_{i_2 i_1}$ so that $(\Delta_{i_2 i_1}/\hbar)^2 \tau_{i_2} \tau_{i_1} \gg 1$ and $J_{i_2 i_1} \approx eN/2\tau_{i_2}$, which becomes temperature-insensitive. Moreover, the IDP design enables a higher injection efficiency due to a reduce overlap between the injection state and the lower laser state upon i_1 - i_2 alignment. With the combination of a coherent tunneling and a higher injection efficiency, Kumar's PPP device works up to $\sim 1.9 \hbar\omega/k_B$.

Figure 2-6. Current densities when the injection state is aligned with (a) the lower laser state at low voltage and (b) the upper laser state at higher voltage.

2.3 A BOUND-TO-CONTINUUM PHONON-PHOTON-PHONON ACTIVE REGION DESIGN

As mentioned above, the BTC design is beneficial for applications that require a broad gain width, however, the thermal backfilling and the weak i - u coupling, as in the RP design, limit the performance of the BTC devices. Here, we propose a design that combine the advantages of the BTC and PPP schemes, named bound-to-continuum phonon-photon-phonon (BTC-PPP).

2.3.1 Band structure

The band diagram of the BTC-PPP design is plotted in Fig.2-7, which consists 6 wells labeled as W1, W2, ..., W6. The fundamental states of W3 – W5 and the excited state of W6 are coupled together to constitute the miniband, the fundamental state of W6 (state i_l) couples to that of W1 of the next module (state i_2) with a coupling strength of $\Delta_{inj} = 3.4$ meV, and they together form the injection states. With this large Δ_{inj} , the electron transport bottle neck in both the RP and BTC designs is eliminated. The upper laser state u is actually the fundamental state of W2, whose energy could be adjusted by changing the width of W2. The energy separation between state i_2 and u is $E_{iu} \approx 26$ meV. We make E_{iu} smaller than the phonon energy (36 meV) so that the electrons will be cooled down in the upper laser state after being scattered from the injection doublet by LO phonon. Assuming an electron temperature of 200 K for the injection states i_l and i_2 , the LO phonon scattering times from the injection states are:

$$\tau_{i_l \rightarrow u} = 1.99 \text{ ps}, \quad \tau_{i_2 \rightarrow u} = 2.44 \text{ ps}, \quad \tau_{i_l \rightarrow l,a,b,e} = 5.55 \text{ ps}, \quad \tau_{i_2 \rightarrow l,a,b,e} = 4.01 \text{ ps},$$

leading to injection time $\tau_{i \rightarrow u} = 1.1$ ps, and parasitic leakage time $\tau_{i \rightarrow l,a,b,e} = 2.33$ ps.

Based on these the injection efficiency is calculated to be $\eta_{inj} = (1/\tau_{i \rightarrow u}) / (1/\tau_{i \rightarrow u} + 1/\tau_{i \rightarrow l,a,b,e}) = 68\%$. Here the electron leakage from the injection states to the upper miniband is neglected. Because of the relatively large energy separation between them (15.6 meV), it is not easy for electrons to leak into the upper miniband. Even if the leakage somehow happens, approximated 50% of the escaped electrons will relax back to state u from the upper miniband by fast LO phonon emission (0.5 ps). As a comparison, in Kumar's design (Fig. 2-5(d)) [102], the same calculation yields $\eta_{inj} = 56\%$. The slightly better injection efficiency of our design is due to a more diagonal LO phonon emission.

Figure 2-7. Conduction band structure and the moduli squared of wavefunctions of the BTC-PPP THz QCL active region design at a bias of 11.7 kV/cm. The layer thickness in nanometer are **4.1/8.3/2.4/12.0/1.7/10.6/3.4/8.7/3.8/6.9/4.2/14.6**, the barriers are indicated in bold fonts and the underlined well is doped.

The photon energy is designed to be 12.6 meV (3.1 THz). Population inversion is insured by a relatively long τ_u for the upper laser state and an ultrashort τ_l for the lower laser state, which is depopulated by the combination of electron-electron scattering and the LO phonon. The related parameters are given in the table below, where three different designs are compared. The BTC design in the table is the one with best temperature performance (160 K) [93]. Assuming the same current flow J_{total} for the three cases, the BTC-PPP design gives a lightly larger product of gain (g_c) and bandwidth ($\Delta\nu$).

Table 2-1. Comparison of some important parameters of THz QCL between the BTC-PPP and other two state-of-art designs [93, 98].

Parameter	Kumar's PPP [98]	BTC [93]	BTC-PPP
E_{ul} (meV)	7.7	12.4	12.6
Z_{ul} (nm)	6.1	4.49	5.27
f_{ul}	0.487	0.454	0.626
τ_{ul} (ps)	2.51	4.66	2.67
τ_u (ps)	1.62	2.44	1.65
τ_l (ps)	0.2	0.3	0.1
η_{inj}	0.56	0.49	0.68
L_{mod} (nm)	54.6	65.6	80.7
Δn (a.u.)	1	1.1	0.87
Δv (a.u.)	1	1.3	1.4
g (a.u.)	0.49	0.38	0.39
$g \cdot \Delta v$ (a.u.)	0.49	0.50	0.54

$$f_{ul} = \frac{2m^* E_{ul} Z_{ul}^2}{\hbar^2}$$

$$\Delta n = \frac{J_{iu}}{eL_{mod}} \tau_u \left(1 - \frac{\tau_l}{\tau_{ul}}\right)$$

$$= \frac{\eta_{inj} J_{total}}{eL_{mod}} \tau_u \left(1 - \frac{\tau_l}{\tau_{ul}}\right)$$

$$g_c = 70 \frac{[\Delta n / (10^{15} \text{ cm}^{-3})] f_{ul}}{[\Delta v / (1 \text{ THz})]}$$

2.3.2 Fabrication

The heterogeneous GaAs/AlGaAs structure was grown by molecular beam epitaxy (MBE) onto a GaAs substrate (see the growth sheet below), 130 modules were stacked, resulting in a total thickness of $\sim 10.5 \mu\text{m}$. The 300nm $\text{Al}_{0.5}\text{Ga}_{0.5}\text{As}$ serves as an etch stop layer for substrate removal. The subsequent fabrication process of the metal-metal waveguide ridge laser can be summarized in Fig. 2-8. The epitaxial wafer, together with an n^+ GaAs receptor wafer, was cleaved into $\sim 6 \text{ mm} \times 8 \text{ mm}$ pieces. A Ti/Au (15/550 nm) layer was then deposited on both the epitaxial wafer and the receptor wafer by electron-beam evaporation. The epitaxial wafer pieces were put upside down on the receptor wafer pieces, crystal orientations of the wafer pair were careful aligned to facilitate cleaving. Wafer bonding was undertaken in a commercial bonding machine, heat (300°) and pressure ($\sim 5.4 \text{ Mpa}$) were applied to the wafer pairs for 1

hour under vacuum environment. After cooling down to room temperature, the wafer pairs were bonded. Fig. 2-9 shows the scanning electron microscopic (SEM) images of the bonding region, the absence of a visible Au-Au interface indicates a satisfactory bonding quality.

After wafer bonding, the original substrate of the epitaxial wafer was mechanically lapped down to ~ 100 μm using fine alumina powders. The remained substrate was then etched away using the $\text{NH}_4\text{OH}:\text{H}_2\text{O}_2$ (1:19) etchant, which stopped at the $\text{Al}_{0.50}\text{Ga}_{0.50}\text{As}$ stop layer. When the stop layer has been reached, the surface becomes mirror-like. After that, the stop layer was removed by immersing the sample in 49% HF acid for ~ 30 seconds.

The top metal strips (Ti/Au 15/300 nm) were defined by standard optical lithography and lift-off processes, following which a second lithography was used to cap the metal strips with wider photoresist strips. The photoresist is used as the mask for the subsequent wet etching, and it is 30 μm wider than the metal layer to account for lithography misalignments and undercut during the wet etching. The metal strips themselves cannot be used as self-aligned masks for wet etching because the galvanic effect in the presence of metal will cause severe undercut at the metal-semiconductor interface and tend to lift off the metal layer. The active region was then etched to form the laser ridges using a $\text{H}_2\text{SO}_4:\text{H}_2\text{O}_2:\text{H}_2\text{O}$ (1:7:80) solution (etch rate ~ 0.4 $\mu\text{m}/\text{min}$). Depending on the orientation of the ridge, the etchant results in different sidewall profiles of the etched ridges (Fig. 2-10 and 2-11).

Following the defining of the laser ridge, the receptor substrate was thinned down to ~ 150 μm by polishing with the alumina powder. The thinner substrate improves heat removal from the working devices. A Ti/Au (20/300 nm) was then employed for the bottom contact. The samples were cleaved, indium soldered onto copper chip carriers, wired, and finally mounted on the cold finger of a cryostat for measurement.

Table 2-2. Growth sheet of the BTC-PPP active region.

Layer	Material	[nm]		Ratio	Doping (cm ⁻³)	Remark
1	GaAs	250				
2	AlGaAs	300		Al(50)Ga(50)As		etch stop layer
3	GaAs	75			5E+18	contact layer
4	AlGaAs	4.1		Al(15)Ga(85)As		
5	GaAs	14.6			2.00E+16	QCL doping
Start of 130 repeat periods						
6S1	AlGaAs	4.2		Al(15)Ga(85)As		
7S1	GaAs	6.9				
8S1	AlGaAs	3.8		Al(15)Ga(85)As		
9S1	GaAs	8.7				
10S1	AlGaAs	3.4		Al(15)Ga(85)As		
11S1	GaAs	10.6				
12S1	AlGaAs	1.7		Al(15)Ga(85)As		
13S1	GaAs	12				
14S1	AlGaAs	2.4		Al(15)Ga(85)As		
15S1	GaAs	8.3				
16S1	AlGaAs	4.1		Al(15)Ga(85)As		
17S1	GaAs	14.6			2.00E+16	QCL doping
End of repeat periods						
18	AlGaAs	4.2		Al(15)Ga(85)As		
19	GaAs	50			5.00E+18	contact layer

Figure 2-8. Schematic illustration of the fabrication steps for metal-metal waveguide ridge THz QCL.

Figure 2-9. SEM images showing the cross-sectional view of the bonding region. The absence of a visible Au-Au interface indicates a good bonding quality.

Figure 2-10. SEM images showing the sidewall profiles of the ridges on (100) GaAs wafers etched by the H_2SO_4 : H_2O_2 : H_2O (1:7:80) solution. (a) and (b) are for ridges aligned to the $\langle 110 \rangle$ and $\langle 101 \rangle$ directions, respectively.

Figure 2-11. Cross-sectional-view SEM image of a fabricated device.

2.3.3 Preliminary experimental results

The fabricated devices were characterized using a Bruker Vertex 70x Fourier transform infrared spectrometer (FTIR) with a room-temperature DTGS detector, optical power was obtained by integrating the spectra over the emission peaks. This enables a relatively high sensitivity in power measurement, as compared to the case where the power is measured directly with a DTGS detector. The results are shown in Fig. 2-12. The devices exhibited a very low threshold current density of $J_{th} = 0.185 \text{ kA/cm}^2$ even in the quasi-CW operation mode (20% – 50% duty cycle). Considering the high duty cycle operation and the relatively large noise equivalent power (NEP) of the detector used (compared to the average output power of our device), the low threshold current density of 0.185 kA/cm^2 is already an overestimated value. As comparison, typical THz QCLs in the literature are with threshold current densities ranged from $0.4 - 1.2 \text{ kA/cm}^2$ for pulsed operation, detected by highly sensitive liquid-helium-cooled bolometers. A low J_{th} is a sign of a high gain coefficient or a low parasitic leakage current before threshold or both [103]. However, considering the low output power of our device, we attributed the low J_{th} to a low parasitic leakage current before the threshold bias, which is also what we expected from the indirect pumping scheme.

However, the small dynamic range (defined as $J_{max} - J_{th}$) of 35 A/cm^2 limits the temperature performance of the device. Detected by the room-temperature DTGS detector, and with 50% duty cycle, our device worked only up to 40 K. We did not characterize the device using a low duty cycle because the very small dynamic range may have made itself inaccessible by short pulses. This is because the short pulses typically have overshoots at the beginning, which can push the device into the NDR region [103]. The small J_{max} is likely due to an early decoupling of injection state i_1 and i_2 , which causes an early NDR and ceases the lasing. Therefore, further optimization may include: 1, push state i_2 higher by slightly narrowing the width of W1; 2, adjust the anticrossing gap between state i_1 and i_2 ; 3, increase the doping level in the structure.

Figure 2-12. (a) Light-Current-Voltage characterization of the BTC-PPP laser at 10 K. The threshold and rollover current densities are $J_{\text{th}} = 0.185 \text{ kA/cm}^2$ and $J_{\text{max}} = 0.22 \text{ kA/cm}^2$, respectively. (b) Spectra of the laser at different current density at 10 K.

2.4 CONCLUSION

This chapter reviewed some theoretical basics of the THz QCL, followed by a comparison of the major design schemes, after which a BTC-PPP active region design was proposed aiming at combining the advantages of the BTC and PPP designs, that is,

broad gain width and high temperature performance. Preliminary experimental result shows a very low threshold current due to the suppression of the parasitic current before the threshold bias. However, the temperature performance of the device is limited by the smaller dynamic range, which is believed to be caused by an early decoupling of the injection states i_1 and i_2 . This problem may be solved by pushing up state i_2 , adjusting the coupling strength between states i_1 and i_2 , or increasing the doping level of the structure.

3. SURFACE-EMITTING CONCENTRIC-CIRCULAR-GRATING TERAHERTZ QUANTUM CASCADE LASERS

In this chapter, we report the design, fabrication and experimental characterization of surface-emitting terahertz (THz) frequency quantum cascade lasers (QCLs) with distributed feedback concentric-circular-gratings. Single-mode operation is achieved at 3.73 THz with a side-mode suppression ratio as high as ~ 30 dB. The device emits ~ 5 times the power of a ridge laser of similar dimensions, with little degradation in the maximum operation temperature. Two lobes are observed in the far-field emission pattern, each of which has a divergence angle as narrow as $\sim 13^\circ \times 7^\circ$. We demonstrate that deformation of the device boundary, caused by anisotropic wet chemical etching is the cause of this double-lobed profile, rather than the expected ring-shaped pattern.

3.1 INTRODUCTION

Lasing action in THz QCL was initially enabled by the novel semi-insulating surface-plasmon waveguide. However, the latter developed metal-metal waveguide (Figure 3-1) has a low waveguide loss and a near 100% modal confinement in the active region. It is, therefore, superior in temperature performance, making THz QCL operate up to ~ 200 K [72]. Nevertheless, owing to the subwavelength mode confinement of the metal-metal waveguide, conventional edge-emitting ridge THz QCLs are characterized by an extremely wide beam divergence ($> 180^\circ$), and a low output power, which is a result of a large impedance mismatch between the modes inside the cavity and in free space. Moreover, without a mechanism for mode-selection, ridge-waveguide lasers suffer from multiple-mode operation. These problems impede the practical applications of THz QCLs. For example, for spectroscopy and heterodyne detection, single-mode operation is often essential, and, for most of the applications, a low beam divergence is highly desirable because it improves the collection efficiency of the optical power.

Figure 3-1. Schematic draw of a ridge QCL employing metal-metal waveguide, where the active region (laser core) is sandwiched between two metal plates.

Various approaches have been developed to address the above-mentioned challenges, involving both edge-emitting and surface-emitting devices (Fig. 3-2). For the former, early works employed additional optical components, such as silicon lenses [104] and horn antennas [105,106] to collimate the divergent output beam. These solutions are not easily integrated with the laser and require careful alignment, but Yu *et al.* subsequently developed an integrated spoof surface plasmon collimator for THz QCLs, and successfully reduced the beam divergence to $\sim 10^\circ$ [77,107]. None of these techniques provide optical mode selection, only optical collimation and beam engineering. However, in 2009, Amanti *et al.* showed that a third-order distributed feedback (DFB) grating could simultaneously enable single-mode operation and collimate the edge-emitting beam tightly ($\sim 10^\circ$) [108]. Compared with edge-emitting devices, surface-emitting counterparts would benefit from the capability of 2D on-chip integration, i.e., a 2D array of the devices can be fabricated on a single chip. In this category, linear second-order DFB grating THz QCLs have been demonstrated to couple the optical power out of a laser cavity [74,109,110]. However, although the beam divergence is narrow along the direction parallel to the ridge axis, it is still highly divergent perpendicular to the ridge axis. Therefore, to achieve a narrow and symmetric far-field profile in two dimensions, some researchers tried bending the laser ridges into rings, resulting in second-order DFB ring-grating THz QCLs [111,112]. However, the reported far-field patterns of these structures remained distorted. As an alternative approach, two-dimensional (2D) photonic crystals were investigated for controlling the surface emission from THz QCLs [113–115]. This allows the far-field profile to be engineered and provides controllable emission properties, and a large emission area. Indeed, single-mode devices with a range of far-field patterns have been reported, and beam divergences as low as $\sim 12^\circ \times 8^\circ$ have been obtained [115]. Nevertheless, it is not simple to achieve single-mode operation with these photonic

crystal cavities because the design usually relies on the band structure of a unit cell of the photonic crystal with periodic boundary conditions, without considering the actual finite scale of the device. Moreover, it is not easy to predict and control the radiation loss (power) of the device without 3D full-wave simulations of the whole structure, which are often computationally intensive. Recently, we have demonstrated that through the use of second-order concentric-circular gratings (CCGs), one can realize surface-emitting THz QCLs with a side-mode-suppression-ratio of 30 dB and efficient power output, with little degradation in the maximum temperature performance [116,117]. The design of such CCGs is much easier than photonic crystal cavity structures, as it is possible to take advantage of the circular symmetry.

Figure 3-2. Some typical wave engineering schemes for THz QCLs, involving edge-emitting devices (a)-(d) and surface-emitting devices (e)-(i). Edge emission: (a) Integrated horn antenna [105]; (b) lens coupling [104]; (c) Integrated surface plasmonic collimator on the facet [107]; (d) Linear third-order DFB grating [108]. Surface emission: (e) Linear second-order DFB grating [110]; (f) Second-order ring grating [112]; (g) Phase-locked second-order DFB grating array [118]; (i) 2D photonic crystal [119].

In this chapter, we present the detailed design, fabrication and experimental results of THz frequency second-order CCG DFB QCLs. Robust single-mode operation is realized at all bias currents, and the side-mode suppression ratios were as high as ~30 dB, indicating efficient single-mode selection. Moreover, the maximum operation temperature of 110 K is comparable with that of a ridge laser of similar size (130K), despite the output power of the CCG QCLs being ~5 times higher. The measured far-field pattern comprises two small lobes, both of which are within a $13.5^\circ \times 7^\circ$ angular

range. The deviation of the far-field pattern from the expected ring shape is attributed to deformation of the active region boundary due to the anisotropic wet chemical etching during fabrication.

3.2 GRATING DESIGN

Our scheme for implementing second-order CCG QCLs is shown in Fig. 3-3(a), where the 10- μm -thick active region is sandwiched between the metallic CCGs on top and a metal plate on the bottom. Inside the active region, the electric-field profile can be approximately expressed as a superposition of Bessel functions of the first and second kind, J_m and Y_m , respectively [120]:

$$E_z(r) = [A(r)J_m(kr) + B(r)Y_m(kr)]e^{im\varphi} \quad (3.1)$$

where $A(r)$ and $B(r)$ are the slowly varying mode amplitudes, k is the wavevector in the active region, φ is the azimuthal coordinate, and $m = 0, 1, 2, \dots$, denotes the order of the Bessel functions and also the order of the azimuthal modes.

The design of the structure was undertaken through numerical simulations using a commercial finite element method solver: *Comsol Multiphysics*. This solver is very effective in finding the eigenfrequencies and mode distributions of a resonator, although three-dimensional (3D) full-wave simulations of the proposed structure are computationally intensive and thus it is impractical to perform 3D simulations throughout the design. However, we can take advantage of the mode's known azimuthal dependence ($e^{im\varphi}$) and simplify the problem to a 2D cross-section simulation, as shown in Fig. 3-3(b); the magnified views show the widths of the metal in each period and the boundary region without metal coverage, denoted as R_1 , R_2 , ... and δ , respectively. Using the 2D "PDE (partial differential equation) Mode" of the *Comsol Multiphysics*, which allow the simulation based on user's own equations, one is able to calculate the optical modes with arbitrary azimuthal order (m) by solving the following equations:

$$\nabla \times (\epsilon^{-1} \nabla \times \vec{H}) - \omega^2 \epsilon^{-2} \vec{H} = 0 \quad (3.2)$$

$$\vec{H}(r, z, \varphi) = \vec{H}(r, z) e^{-im\varphi} \quad (3.3)$$

where $\vec{H}$ is the magnetic field, ϵ the relative permittivity, c the speed of light in vacuum and ω the angular frequency of the light. Without loss of generality, we restrict our design to $m=0$ (the fundamental azimuthal mode) because the surface-emitting beams of these modes give radial polarization, which is highly desirable when coupling light into a THz metal-wire waveguide or the metallic tip of a near-field imaging system [41].

The structure was designed to operate at ~ 3.75 THz. At this frequency, the real part of the refractive index of the active region was calculated to be ~ 3.6 , from the emission spectra of a ridge laser with the same active region, using $n_{\text{active}} = c / (2 L \Delta f)$, with L being the length of the laser ridge and Δf the Fabry-Pérot mode spacing in the spectra. To calculate the emission loss only, the imaginary part of the refractive index of the active region was set to zero and the metal was considered to be perfect. The 2D structure was then surrounded by an absorbing boundary that can isotropically absorb the emitted radiation without reflection.

Figure 3-3. (a) Three-dimensional schematic representation of the THz concentric-circular-grating quantum cascade laser design. The semitransparent rectangle represents the two-dimensional (2D) simulation plane. (b) 2D simulated magnetic field distribution making use of the 360° rotational symmetry of the designed structure. The magnified views show the widths of the metal in each period and the boundary region without metal coverage, indicated as R_1 , R_2 , ... R_{15} and δ , respectively. The black lines represent metal.

The design began by determining the grating period and the widths of the open slits. For large area electric pumping via one or a few connected wires, it is important to guarantee uniform current injection over the pumping area, otherwise the mode behavior of the QCL may be disturbed. Therefore, the width of each open slits on the grating was predetermined as $2 \mu\text{m}$. The effective refractive index of the CCG

structure was then estimated to be $n_{\text{eff}} = 3.57$, and so for a target wavelength of $80\text{ }\mu\text{m}$ (3.75 THz), a second-order grating period of $22.7\text{ }\mu\text{m}$ is required, which makes the mark-to-space ratio of the open slits less than 10%. We restricted the radius of the semiconductor active region R_{total} to be ~ 16 grating periods, taking into account the maximum current output of commercial power supplies and the heat extraction capacity of a typical cryostat. To suppress unwanted whispering-gallery-like modes, an annular boundary region of width $\delta = 22.7\text{ }\mu\text{m}$ was left uncovered by metal [116].

The grating was initially designed as a standard second-order DFB grating. The whole grating structure starting from the center to the boundary is as follows: 20.7/2/20.7/2/20.7/2/20.7/2/20.7/2/20.7/2/20.7/2/20.7/2/20.7/2/20.7/2/20.7/2/20.7/2/22.7/**22.7** μm , where the bold number indicates the open region without metal coverage. The corresponding electromagnetic field distributions and spectra are shown in Figs 3-4(a) and (b), respectively. A red arrow highlights the mode with the lowest loss, which is expected to be the lasing mode. However, since neighbouring modes have similar predicted losses, this structure may not achieve single-mode operation. In linear DFB gratings, the width of the grating boundary plays a critical role in determining the mode behavior [74], and this also holds for our CCG design here. As shown in Figs 3-4(c) and (d), by varying R_{15} whilst keeping all other parameters unchanged, the corresponding mode spectrum changes significantly. The best result was achieved when $R_{15}=16.2 \mu\text{m}$ and correspondingly, $R_{\text{total}}=356.7 \mu\text{m}$, as the expected lasing mode was most distinct from neighbouring ones. In the modifications that will be discussed below, δ , R_{15} and R_{total} are left unchanged.

To exploit the gain of the active region fully, one should consider the electric field profile inside the cavity, with an evenly-distributed electric field profile being desirable. A uniform electric field distribution also helps improve the beam divergence. We thus investigated the E_z (z-polarized electric field) profile inside the cavity, as the QCL active region can only provide gain for the TM mode (Figs 3-4(e)-(g)). For our initial designs, it can be seen that E_z is concentrated in the central region (also shown in Fig. 3-4(a)). This is because the first few slits in the central region provide too strong feedback. In fact, the strength of feedback and the emission property of a slit depend critically on the location of the slit relative to the electric field: the slit provides the strongest feedback when placed at a null of the electric field, albeit with high emission loss from this slit. In contrast, a slit placed at an extremum of

the electric field provides the weakest feedback and the lowest emitting loss. From Fig. 3-4(a) one can see that the first few slits are located near the nulls of the electric field, which explains their strong feedback and hence the high field concentration in the central region. To mitigate this, the positions of the first few slits were systematically adjusted to reduce their feedback.

Figure 3-4. (a) Electric field distribution of the standard second-order DFB CCG THz QCL in the central region. (b) – (d) Mode spectra of the structures for different R_{15} with other parameters unchanged. (e) – (g) the corresponding electric-field (E_z) profiles of the expected modes, highlighted by the red arrows in (b) – (d), respectively, obtained at the middle height of the active region.

Fig. 3-5 shows the simulated results following adjustment of slit locations. The slits are numbered outwards from the center as slit 1, 2, 3... . First, slits 1 and 2 were moved outwards by 3.4 μm ; this aligned them with the maximum of the electric field (Fig. 3-5(a)(i)). E_z thus becomes more evenly distributed along the radius (Fig. 3-5(a)(iii)), compared with Fig. 3-4(g), and the expected lasing mode is also more distinct from others (Fig. 3-5(a)(ii)) because of the reduction in emission loss after this adjustment, as a result of the suppressed light emission from slits 1 and 2. To optimize the structure further, slit 3 was moved outwards by 1.9 μm (Fig. 3-5(b)), and then slits 4 and 5 were moved outwards by 0.7 μm (Fig. 3-5(c)). Fig. 3-5(c) shows the final

design. It has the greatest mode distinction, and most evenly-distributed E_z field. Furthermore, the calculated emission loss is 3.9 cm^{-1} , comparable to that of a typical ridge THz QCL. The grating parameters starting from the center are: 24.1/2/20.7/2/19.2/2/19.5/2/20.7/2/20.2/20.7/2/20.7/2/20.7/2/20.7/2/20.7/2/20.7/2/20.7/2/20.7/2/20.7/2/16.2/22.7 μm , where the bold number indicates the open regions (slit or boundary region).

Since the CCG is designed for an electrically-pumped QCL, a three-spoke bridge structure was employed to connect the concentric rings, as illustrated in Fig. 3-6, and to enable electric pumping of the whole grating.

The design discussed above considers only the fundamental azimuthal modes. However, higher azimuthal modes ($m \geq 1$) are also eigen-solutions of the circular structure, similar to the higher lateral modes in a ridge laser. To take into account these higher azimuthal modes and, more importantly, the influence of the three-spoke bridge, we performed a 3D full-wave simulation of the final structure. The computation took over 35 hours and 180 gigabytes RAM, running parallelly on a computing cluster node with 16 processes. Fig. 3-7(a) presents the mode spectra of the first four azimuthal modes; modes $m > 4$ are not shown because they have higher loss. It can be seen that there usually exists a second-order azimuthal mode accompanying each fundamental mode with a similar loss. As the electric field in the active region can be described as a superposition of the Bessel functions of the first and second kind (Eq. (3.1)), we compare the fundamental ($J_0(r)$, $Y_0(r)$) and second-order ($J_2(r)$, $Y_2(r)$) Bessel functions in Fig. 3-7(b) and (c). From Fig. 3-7(b), we can see that the $-J_2(r)$ almost coincides with $J_0(r)$ except at the first two or three peaks, and the same is true for $-Y_2(r)$ and $Y_0(r)$ (Fig. 3-7(c)). Therefore, the CCG provides similar feedback and emission loss for the fundamental and second-order azimuthal modes, and hence the second-order azimuthal modes accompany the fundamental azimuthal modes, with similar emission losses. Nevertheless, the CCG is specifically optimized for the expected fundamental mode, and it consistently has a lower loss than the corresponding second-order mode. It should also be noted, from the inset of Fig. 3-7(a), that the perturbation of the three-spoke structure on the optical modes in the active region is minor.

Figure 3-5. (a), (b), (c) Simulation results after each adjustment of the slit locations. (i) Electric field ($|E|$) distribution of the expected mode at the central part of the grating. The first five slits are numbered 1, 2, ..., 5, and their locations are indicated by the white arrows. White dashed lines near the arrows indicate the original locations of the slits. The displacement distance of each slit is given above the arrow. (ii) Mode spectrum of the adjusted structure, a red arrow indicates the expected mode. (iii) The electric-field (E_z) profile.

Figure 3-6. (a) A false-color SEM image shows the fabricated concentric circular grating (CCG) terahertz quantum-cascade laser (QCL), in which a three-spoke bridge structure connects the concentric rings together to allow electrical pumping of the whole grating. (b) Optical microscope images of the device. Magnified views on the right show the dimensions of the center ring and the boundary slit without metal coverage.

Figure 3-7. (a) Mode spectrum of the device using 3D full-wave simulation to take the higher azimuthal modes into account. Only the first four lowest order azimuthal modes are plotted. The electric field distributions of the modes with lowest losses (the fundamental and second azimuthal modes, highlighted by a red circle) are shown in the inset. (b), (c) Comparison of the Bessel function $J_0(r)$ and $-J_2(r)$, and $Y_0(r)$ and $-Y_2(r)$, respectively. The fundamental and second-order Bessel functions coincide except in the first few peaks.

3.3 FABRICATION

The active region of the THz QCLs used for this work is a re-growth of that reported in Ref. [100] but with a slightly higher doping, whose heterogeneous GaAs/Al_{0.15}Ga_{0.85}As structure was grown on a semi-insulating GaAs substrate by molecular beam epitaxy. A Ti/Au (15/550 nm) layer was deposited on both the active wafer and a receptor n⁺ GaAs wafer by electron-beam evaporation. The active wafer was inverted onto the receptor wafer, carefully aligning the crystal orientation of the

wafer pair to facilitate cleaving. The wafer pairs were bonded together under application of pressure (~ 5.4 Mpa) at 300°C for 1 hour in a vacuum environment. The original substrate of the active wafer was then removed by lapping and selective chemical etching; this was followed by the removal of the highly absorptive contact layer of the active region using a H_2SO_4 : H_2O_2 : H_2O (1:7:80) solution, to prevent significant loss for the THz radiation coupled out through the grating slits. Top metal gratings (Ti/Au 15/200 nm) were defined by standard optical lithography and lift-off, after which the active region was wet-etched to form a disk structure using a H_3PO_4 : H_2O_2 : H_2O (1:1:10) solution with photoresist (AZ5214) used as the mask. The backside substrate was thinned down to ~ 150 μm to improve heat dissipation, and a Ti/Au (20/300 nm) layer employed for the bottom contact. The sample was then cleaved, mounted onto Cu submounts, wire bonded, and finally put on the cold finger of a cryostat for measurement.

3.4 EXPERIMENTAL RESULTS AND DISCUSSION

3.4.1 Light-current-voltage characterization

Light-current-voltage (LIV) characterization was carried out at different heatsink temperatures under pulsed mode operation (200 ns pulses at 10 kHz repetition), and results are shown in Fig. 3-8(a). The devices operate up to 110 K. For comparison, the maximum operation temperature, under the same operation conditions, of double-metal waveguide, ridge lasers of similar size fabricated from the same epitaxial wafer is around 130 K. Fig. 3-8(b) shows the emission spectra of the device at a 9 K heatsink temperature for various injection currents, ranging from threshold to the rollover. The detected side-mode suppression ratio (SMSR) is around 30 dB, limited by the noise floor of the room-temperature pyro-electronic detector, as shown in the inset to Fig. 3-8(b). This, together with the robust single-mode operation at 3.73 THz under all injected currents, reflects the strong single mode selectivity of the structure. Moreover, the collected power of the device is ~ 5 times higher than that of its ridge laser counterpart, indicating efficient light coupling out of the cavity by the grating.

Figure 3-8. (a) Light-current-voltage characterization of the CCG device at different heatsink temperatures under pulsed mode operation. (b) Emission spectra of the device at 9 K heatsink temperature as a function of injected current, from threshold to the rollover. The inset shows a logarithmic scale plot of the spectrum, demonstrating a side-mode suppression ratio of around 30 dB.

3.4.2 Effect of boundary deformation on the optical mode

To investigate the electric field distribution in the grating cavity, the two-dimensional far-field emission pattern was measured. The radiation emitted by the device was sampled through a small aperture in front of a parabolic mirror, which was scanned over part of a spherical surface centered on the device. The parabolic mirror reflected the radiation into a liquid-helium cooled bolometer for detection. The small aperture was used to increase the measurement angular resolution. The measured far-field pattern (Fig. 3-9(a)) exhibits double lobes, with full-width-at-half-maximum (FWHM) values of $12.5^\circ \times 6.5^\circ$ and $13.5^\circ \times 7^\circ$. Similar results were obtained in several devices. This implies that the mode in the CCG resonator is not the expected fundamental mode, which should have a ring-shaped far-field pattern (Fig. 3-9(b)). Based on the method described in Ref. [121], the calculated far-field profiles of the fundamental, the first-order and second-order azimuthal modes are shown in Figs 3-9(b) – (d), respectively; none has a double-lobe far-field profile.

Figure 3-9. (a) Measured far-field pattern of a typical THz CCG QCL. (b), (c), and (d) Electric field distributions and the corresponding far-field patterns for the fundamental, the first-order and second-order azimuthal modes, respectively.

It was found that the deviation of the far-field pattern from the expected ring shape is due to an anisotropic sidewall profile caused by wet chemical etch. As shown in Fig. 3-10(a), the wet-etched sidewall is more vertical for the upper and lower boundaries than the left and right boundaries, which makes the shape of the disk slightly elliptical (the conjugate radius along the y-axis is $356.7 \mu\text{m}$ and the transverse radius along the x-axis is $358.7 \mu\text{m}$). It should be noted that this deformation actually stems from the anisotropic crystal structure of the active region semiconductor, which makes the wet-etched sidewall profiles and the undercuts different along different crystallographic direction. Therefore, all the devices have similar, if not the same, boundary deformation. Although slight, this deformation breaks the circular symmetry of the fundamental mode (Fig. 3-10(b)) and consequently the simulated far-field pattern is as shown in Fig. 3-10(b), which agrees well with the measured result. It is worth mentioning that although the deformation of the boundary has such an effect on the fundamental azimuthal modes, the simulations show that there is negligible effect on the higher azimuthal modes. This is because higher order azimuthal modes have a lower degree of circular symmetry and are less sensitive to this deformation.

Figure 3-10. (a) Scanning electron-beam microscope images of the anisotropic sidewall profiles caused by wet chemical etching. The sidewalls of the upper and lower boundaries are more vertical than those of the left and right boundaries. (b) The change of electric field distribution and far-field pattern caused by a slight (2 μm) deformation of the circular active region disk.

3.4.3 Coherent superimposition of the optical modes

We have also considered the possibility that this far-field pattern might be due to the coherent superimposition of the fundamental and second-order azimuthal modes (Fig. 3-9 (b) and (d)) since they're very close in the spectrum (Fig. 3-7(a)) and may not be discriminated in the measured laser spectra. If the phase difference of these two modes $\Delta\phi$ is constant from pulse to pulse so that their far fields could be coherently superimposed during the measurement, the resulting far-field pattern may be double-lobed when their far-field electric fields have proper amplitudes and phases (Fig. 3-11(a)). Note that our devices worked in pulse mode operation, if $\Delta\phi$ varies from one pulse to another, the measured far-field pattern of these two modes, if there are, will be resulted from their incoherent superimposition, which then is not double-lobed. However, even the best calculated result shown in Fig. 3-11(a) doesn't match well with the measured result (Fig. 3-9(a)). Furthermore, the mechanism that locks the phase difference of different modes is usually due to the nonlinear effects in the laser cavities, which requires high electric field intensity and, thus, may happen in high-power (watt level) lasers [122–124]. Since our devices are of relatively low power, it's unlikely that this nonlinear effect would play a role here. Moreover, when the injected current changes, this nonlinear effect will change the value of $\Delta\phi$ and the

amplitudes of these two modes significantly [122–124], leading to different far-field patterns, as shown in Fig. 3-11. However, in our measurement, the double-lobed far-field pattern did not change when we changed the injected current. Therefore, with all these considered, it is unlikely that the double-lobed far-field patterns we measured are due to the superimposition of different azimuthal modes.

Figure 3-11. Coherent superimposition of the far fields of the fundamental and second-order azimuthal modes. (a) – (e) Resulting far-field patterns with the phase difference of the two modes $\Delta\phi$ varies from 0° to 120° . (f) Resulting far-field pattern with $\Delta\phi = 0^\circ$ but 1.5 times amplitude for the second-order azimuthal mode.

3.4.4 Frequency splitting caused by the boundary deformation

Another effect of the deformation is that it leads to frequency splitting of the original modes. As shown in Fig. 3-12, for the fundamental azimuthal mode, it will split into two deformed modes (Fig. 3-12 (b) and (c)) under the deformation, whose corresponding far-field patterns are shown in Fig. 3-12 (d) and (e), respectively. This is because, due to the deformation, the effective cavity length seen by these two modes are different, resulting in different eigen-frequencies and losses. However, in experiments, all the measured far-field patterns are similar as the one shown in Fig. 3-

12(d), where the two lobes align in the perpendicular direction, indicating that only the mode shown in Fig. 3-12(b) is excited due to lower loss.

Figure 3-12. Splitting of the fundamental azimuthal mode under deformation. (a) Electric field distribution of the original fundamental azimuthal mode. (b), (c) Electric field distributions of the splitted deformed fundamental azimuthal modes, and their corresponding far-field patterns (d), (e), respectively.

3.4.5 The whispering-gallery-like Modes

It is worth mentioning that the device could also display whispering-gallery-like (WGL) modes, similar to those shown in Fig. 3-13(c), since the device has a disk structure. These WGL modes typically have high-Q factors, and therefore can be excited at lower drive currents, before the appearance of the low-order azimuthal mode, e.g., third-order as shown in Fig. 3-13(a). However, these WGL modes are well confined in the cavity with very low out-coupling efficiencies, which prevents them from being observed experimentally. To gain insight into the behavior of these WGL modes, we calculated the momentum-space intensity distributions by using the Fourier transform of the in-plane fields (Figs. 3-13(b) and (d)) [121]. The red dashed circle in each figure represents the light-cone defined as $k_x^2 + k_y^2 = (\omega/c)^2$, where only the radiation components inside the light-cone can be out-coupled. As there is no observable component in the light-cone (Fig. 3-13(d)) for the WGL modes, such modes cannot contribute to the far-field radiation. This contrasts with CCG modes, where efficient out-coupling can be expected.

Figure 3-13. Supported modes in the CCG DFB QCLs, and the corresponding momentum space intensity distributions. (a) Electric field intensity of the third-order azimuthal lasing mode and (c) a whispering-gallery-like (WGL) mode. (b) and (d) in-plane momentum-space intensity distributions for the modes in (a) and (c), respectively. The red dashed circle represents the light-cone – only radiation components inside the light-cone can be out-coupled. There are no intensity components in the light-cone for the WGL mode, in contrast to the situation for the third-order CCG mode.

3.4.6 Lasing at third-order azimuthal mode

As the same as in the photonic crystal design, the validity of the design approach described above relies on the accurate estimation of the refractive index of the active region n_{active} . In an early device where n_{active} was not determined properly, lasing was achieved at the third-order azimuthal mode instead of the fundamental azimuthal mode. There, the whole designed grating structure starting from the center of disk to the boundary is as follows: 23.4/2/20.1/2/18.7/2/18.9/2/20.1/2/19.4/2/20.1/2/20.1/2/20.1/2/19.9/2/20.1/2/20.1/2/20.5/2/16.9/**24.2** μm , where bold number indicates the

open slits. As a result of the deviation in the azimuthal mode, the device emitted less optical power (~ 3 times as the ridge laser) at a slightly higher frequency of ~ 3.8 THz. Nevertheless, single mode operation was achieved for all driving conditions with a SMSR around 30 dB. Fig. 3-14 presents the related experimental results, and the measured and simulated far-field emission patterns are shown in Fig. 3-15.

Figure 3-14. Measured results of an early CCG DFB laser where the refractive index of the active region was not properly estimated. (a) Pulsed (200 ns pulses repeated at 10 kHz) light-current-voltage (LIV) characteristics of the laser. (b) Emission spectra of the laser at 78 K for different injected current densities, from the threshold to the rollover. The inset shows a logarithmic scale plot of the spectrum, indicating a side-mode suppression ratio of around 30 dB.

Figure 3-15. Two-dimensional far-field emission patterns of the CCG DFB QCL. (a) The experimentally measured emission, and (b) the simulated emission of the third-order azimuthal mode.

3.5 CONCLUSION

We have presented the design, fabrication and experimental measurements of surface emitting THz QCLs with second-order CCGs. Compared with the photonic crystal cavity structures, the design of the CCG cavity is much simpler thanks to its circular symmetry. Robust single-mode operation has been achieved with a SMSR as high as ~ 30 dB. Furthermore, the device emits ~ 5 times the power of a ridge laser of similar size, with little compromise in the maximum operation temperature. The beam divergence of the emitted radiation is greatly reduced, showing two small lobes in the far-field, each of which has a divergence angle within $13.5^{\circ} \times 7^{\circ}$, corresponding to the deformed fundamental azimuthal mode. The deviation of the far-field pattern from the expected ring shape is attributed to the boundary deformation of the active region, caused by the anisotropic wet chemical etching during fabrication.

4. PLANAR INTEGRATED METASURFACES FOR HIGHLY-COLLIMATED TERAHERTZ QUANTUM CASCADE LASERS

In this chapter, we report planar integration of tapered terahertz (THz) frequency quantum cascade lasers (QCLs) with metasurface waveguides that are designed to be spoof surface plasmon (SSP) out-couplers by introducing periodically arranged SSP scatterers. The resulting surface-emitting THz beam profile is highly collimated with a divergence as narrow as $\sim 4^\circ \times 10^\circ$, which indicates a good waveguiding property of the metasurface waveguide as the beam divergence is inversely proportional to the emission area. In addition, the low background THz power implies a high coupling efficiency for the THz radiation from the laser cavity to the metasurface structure. Furthermore, since all the structures are in-plane, this scheme provides a promising platform where well-established surface plasmon/metasurface techniques can be employed to engineer the emitted beam of THz QCLs controllably and flexibly. More importantly, an integrated active THz photonic circuit for sensing and communication applications could be constructed by incorporating other optoelectronic devices such as Schottky diode THz mixers, and graphene modulators and photodetectors.

4.1 INTRODUCTION

Surface plasmons (SPs) are surface electromagnetic waves bound at the interface between metallic and dielectric materials. A flexible means of confining or manipulating optical waves at a subwavelength level can thus be achieved by patterning the metallic surface, and as such SPs have underpinned numerous studies on compact photonic circuits [125], enhanced light-matter interactions [126], near-field imaging systems [41,127], and beam shaping [77,128,129], inter alia. Conventionally, operation of these plasmonic devices relies on external illumination of a properly polarized laser beam on an input coupler (such as a prism or grating), where the SPs are generated. However, these methods often require meticulous alignment of the external optical components, which are bulky and inconvenient for compact integration. Therefore, efforts have been made to integrate plasmonic structures monolithically with semiconductor lasers, resulting in integrated active plasmonic devices. As a straightforward implementation, various kinds of plasmonic structures have been integrated directly onto semiconductor laser facets for beam shaping, such

as generating deep-subwavelength laser spots [130,131], reducing beam divergence [129,132], producing multi-beam emissions [133,134], and controlling the polarization state [135]. Moreover, integration of a passive SP waveguide into a semiconductor laser has also been recently demonstrated [136,137].

Although the use of SPs in these types of devices has proven successful in the visible to mid-infrared regions of the spectrum, the SP concept cannot be easily translated into the terahertz (THz) region. This is because the metal behaves more like a perfect conductor at such frequencies, and hence the penetration depth of the THz field into the metal is negligible (three orders of magnitude shorter than the wavelength in free space); this leaves the electromagnetic field only loosely bound to the flat surface. However, it was found that artificial metallic structures at a deep-subwavelength scale can mimic the optical response of the metal atoms to visible and near-infrared light in the longer wavelength region. Metasurface made from such ‘artificial atoms’ (meta-atoms) can thus been designed to support tightly confined THz surface waves [138–141] in a way just like the SP behavior of metal at shorter wavelengths. Moreover, the dispersion relation of these THz surface waves, usually referred to as ‘spoof’ surface plasmons (SSPs), can be geometrically tailored, providing additional freedom in the design of THz plasmonic devices.

The invention of the THz frequency quantum cascade laser (QCL) [66] in 2002 opened up a number of possibilities for THz photonics [107,142,143]. THz QCLs are electrically pumped compact semiconductor lasers based on the electronic transitions between subbands in the conduction band, which can be flexibly engineered. As such, the light in the active region is intrinsically TM-polarized, matching the polarization of SPs. Therefore, QCLs are potentially ideal sources for integrated THz active plasmonic system. For example, using a planar-integrated metasurface waveguide, it is possible to couple the THz radiation directly and efficiently out of the laser cavity as SSP waves, which can then be fed into a SSP device or circuit. Although an integrated THz metasurface collimator has been demonstrated on a QCL facet itself [107], the fabrication was difficult, requiring use of focused ion beam technology, and the small facet area also limits its adoption for practical applications. Planar integration of metasurface components to a THz source for active plasmonic systems has yet to be demonstrated.

In this chapter, we report the planar integration of tapered THz QCLs with metasurface structures, which are processed into SSP out-couplers by introducing

periodically arranged scatterers. The resulting surface-emitting THz beam is highly collimated with a beam divergence as narrow as $\sim 4^\circ \times 10^\circ$. This low divergence indicates a good waveguiding property of the metasurface waveguide, and the low background THz power, away from the surface-emitting beam, implies a high coupling efficiency of the THz radiation from the laser cavity to the metasurface. Moreover, since the whole structure is in-plane, this scheme provides a promising platform where well-established surface plasmon techniques can be employed to engineer an emitted THz QCL beam controllably and flexibly. Furthermore, an integrated active THz photonic circuit may be constructed by guiding and coupling the SSP to other optoelectronic devices such as a Schottky diode THz mixer [142,143], or graphene modulators [12] and photodetectors [144].

4.2 DEVICE DESIGN

4.2.1 Device overview

Fig. 4-1(a) shows a scanning electron microscope image of a fabricated device, where the tapered THz QCL is shown on the left and the metasurface structure on the right. The laser cavity is formed between the curved facet of the tapered structure and a back distributed Bragg reflector (DBR) towards the left of the ridge region (inset of Fig. 4-1(a)). The metasurface structure consists of a SSP waveguide (periodic narrow and shallow gold-coated grooves, each of which functions as a simple one-dimensional meta-atom) and SSP scatterers (wide and deep gold-coated grooves), as illustrated in Fig. 4-1(b), which is a 3D schematic cross-sectional view of the device along the white dashed line in Fig. 4-1(a). Fig. 4-1(c) then presents an enlarged top view of the central region of Fig. 4-1(a), showing details of the fabricated device.

Figure 4-1. (a) Scanning electron microscope (SEM) image of a fabricated device. The tapered THz QCL consists of DBR, ridge and taper sections. The inset shows the details of the DBR structure, which is formed simply by gold patterning on top of the active region. The markers by the side of the laser are used for mask alignment during the fabrication. (b) 3D schematic cross-sectional view of the device along the white dashed line in (a). (c) Enlarged top view of the central region of the device.

4.2.2 Design of the tapered laser cavity

The tapered THz QCL used in this work comprises a 10- μm -thick active region design, labeled as V775 with a gain peak at ~ 3 THz (~ 100 μm) [145]; this is confined by a double-metal waveguide structure in the vertical (z -) direction. The QCL is patterned with a tapered structure, first to collimate the emission in the lateral direction as the ideal input for the one-dimensional SSP waveguide is a parallel beam (the beam divergence in the lateral direction is inversely proportional to the facet width) and, second, to increase the THz emission amplification in the laser cavity. Fig. 4-2(a) shows a numerical simulation of the electric field (E_z) distribution of the tapered THz QCL calculated using COMSOL Multiphysics, a commercial finite-element-method solver. The taper section has a tapering angle of $\sim 36^\circ$ and a length of 800 μm , which increases the width of the output facet from 50 μm to 500 μm . The shape of the taper facet is designed to be an arc centered 20 μm to the left of the ridge-taper interface to collimate the output beam (Fig. 4-2(a)). A flat taper facet [146] is not appropriate for such a large tapering angle because it significantly distorts the optical field owing to

the large mismatch between the shape of the facet and the wavefront of the radiation emitted from the laser ridge, which would lead to an uncollimated emitted beam (Fig. 4-2(b)). The rear DBR (Fig. 4-2(a)) is simply formed by a metal grating patterned on top of the active region, whose reflectance spectrum is plotted in Fig. 4-3. Finally, to suppress higher-order lateral modes, side-absorbers (black regions in Fig. 4-2(a) and (b)) are created by leaving the active region uncovered by the metal; the exposed upper n^+ -GaAs contact layer of the active region acts as an efficient absorber with which the higher-order lateral modes have larger overlaps than the fundamental lateral mode [147]. Fig. 4-2(c) shows the electric field distribution of a second-order lateral mode of the structure.

Figure 4-2. (a) Electric field (E_z) distribution of the tapered THz QCL with a curved front facet; magnified views show the details of the DBR region, and the taper. The black regions represent the active regions without metal coverage, which are highly absorbing. (b) E_z distribution of a tapered structure with a flat facet. The electric field in the laser cavity is distorted, resulting in uncollimated emission. (c) E_z distribution of a second-order lateral mode which is suppressed due to a larger overlap of the mode with the side absorbers.

Figure 4-3. Simulated reflectance spectrum of the DBR region. High reflection only happens in the frequency band ranging from 2.9 THz to 3.4 THz.

4.2.3 Design of the metasurface collimator

Fig. 4-4(a) shows a 2D simulation of the electric field distribution of the whole device performed in the y-z plane along the symmetric line of the structure (white dashed line in Fig. 4-1(a)), with the metasurface collimator geometry presented in Fig. 4-4(b). The excitation and scattering of the SSPs can be clearly observed. The SSP wavelength varies from 89.4 μm to 92.7 μm as the wave propagates away from the laser facet, compared to a 96 μm value in free space. The shorter SSP wavelength in the vicinity of the laser facet is explained by a larger wavevector of the orange region than that of the yellow region in Fig. 4-4(b), as reflected in the dispersion diagram (Fig. 4-4(c)), which was obtained by a finite-difference time-domain (FDTD) method with the commercial Lumerical software. The SSP dispersions show strong similarities with the dispersion of the SP mode on a flat metal surface. However, while the asymptotic frequency f_c of the SP mode is fixed as $\omega_p / 2\sqrt{2}\pi$ (ω_p is the bulk plasma frequency of the metal), f_c of this groove-type metasurface and thus the behavior of the SSP mode, can be controlled by the groove depth h : $f_c \approx c_0 / 4h$ [141], where c_0 is the velocity of light in vacuum. For a given frequency, a smaller asymptotic frequency means a larger SSP wavevector, which implies a better confinement of the surface

wave. Nevertheless, a stronger confinement of wave on metal surface comes with a larger ohmic loss in the metal. Figure 4-5 shows the propagation length of the SSP wave on the metasurface, as well as the length of the evanescent tail, as a function of the groove depth, calculated by FDTD simulations. For a groove depth of 7 μm , the propagation length is 30 mm while it is 12.5 mm for 9 μm groove depth. A groove depth of 0 μm corresponds to the bare gold surface, and the corresponding propagation length is 1020 mm. In this design, we chose the groove depth to be 7 μm as a tradeoff between the wave confinement and the scattering efficiency of the 25 scatterers: while a tighter confinement ensures a better coupling efficiency of the light from the curved laser facet, it will make the scatterers less efficient so that not all of the surface wave will be scattered into far field. After all, if the groove depth of the waveguide part is the same as that of the scatterer, the surface wave will see less difference between the waveguide part and the scatterers.

In Fig. 4-4(a), the 1/e evanescent tail of the SSP wave is around 50 μm , in agreement well with the value calculated by $1/\sqrt{k_{ssp}^2 - k_0^2}$, with k_{ssp} being the SSP wave vector and k_0 the free-space wave vector. The corresponding ohmic loss of the metal is 1-2 cm^{-1} . With respect to the emission power, it is dependent on the intensity transmission at the laser output facet, which can be expressed as $T = 1 - \left| \frac{Z_F - Z_{MM}}{Z_F + Z_{MM}} \right|^2$, where Z_{MM} and Z_F are the impedance of the QCL's metal-metal waveguide and that of the outer space seen from the laser facet, respectively. Usually, Z_F differs greatly from Z_{MM} (large impedance mismatch), resulting in an insufficient output efficiency of the THz power. However, Z_F is affected by the surrounding environment of the laser facet. In our case here, the metasurface varies Z_F in such a way as to reduce the mismatch between Z_F and Z_{MM} , leading to a higher output power. With this in mind, it is logical that the width of the metasurface might have effect on the facet transmission. However, if the grooves of the metasurface is wide enough to cover the entire width of the laser facet (500 μm), the transmissions through the laser facet are almost the same with an enhancement factor of ~ 1.2 compared to the case without metasurface (Fig. 4-4(d)). In terms of coupling efficiency, it is estimated that 40% of the laser output power is coupled into the SSPs, with the remaining radiated directly

into free space. Most of the power in the SSPs is then scattered out perpendicular to the device surface (z-direction) by the 25 SSP scatterers, which are grooved on the waveguide with a periodicity of $78\ \mu\text{m}$ so that the scattered SSP light and the uncoupled light interfere constructively, resulting in a narrow ($\sim 3^\circ$) single lobe in the far field (Fig. 4-4(d)). As a comparison, the laser without the metasurface structure emits uniformly in all directions, as shown in Fig. 4-4(e).

Figure 4-4. (a) Simulated 2D light intensity distribution of a metasurface collimator. The simulation was performed in the (y-z) plane along the symmetric line (white dashed line in Figure 1(a)) of the device. (b) Cross-section of the metasurface collimator design. The narrow grooves have a width of $5\ \mu\text{m}$ and a period of $10\ \mu\text{m}$. The narrow grooves are $9\ \mu\text{m}$ deep in the orange region and $7\ \mu\text{m}$ deep in the yellow region (which is repeated 25 times). The narrow grooves in the orange region are deeper to enhance coupling from the laser into the SSPs. The $25\ \mu\text{m}$ grooves, which scatter out the SSPs, are $9\ \mu\text{m}$ in depth. (c) Dispersion diagrams of the metasurface with grooves of $5\ \mu\text{m}$ width and $10\ \mu\text{m}$ period, but different depths. The black dotted curve, red curve and the blue dashed curve correspond to $h = 0\ \mu\text{m}$ (flat surface), $7\ \mu\text{m}$ and $9\ \mu\text{m}$, respectively. The $h = 0\ \mu\text{m}$ curve almost coincides with the light line in vacuum in the THz frequency range. The lower inset presents a zoom-in view of the

region marked by a green box. (d) Dependence of the transmission through the laser facet on the width of the metasurface. (e) Calculated far-field intensity profile along the metasurface waveguide direction. The enlarged view of the central lobe in the inset shows that the beam divergence is as narrow as 2.7° . (f) Simulated 2D light intensity of a laser without the metasurface structure. The inset shows that the light emission is relatively uniform in all directions.

Figure 4-5. Propagation length and the length of the evanescent tail of the THz surface plasmonic wave on the metasurface as a function of the groove depth. The inset shows a zoom-in view of the same data. The simulated wavelength is $95\ \mu\text{m}$.

4.3 FABRICATION

Fabrication of the tapered THz QCLs with metasurface waveguides began with Au-Au thermocompression bonding of the QCL active region to an n+ GaAs receptor wafer. The original QCL substrate was then removed by a combination of lapping and selective chemical etching. A top contact metal (Ti/Au 15/350nm) was next defined by conventional optical lithography and lift-off, the DBR section was also formed in the same process. The laser mesa and deep grooves (SPP scatterers) were first etched down $\sim 3\ \mu\text{m}$ by reactive ion etching (RIE) using a SiO_2 mask. Another lithography process was then performed to define additional patterns for the shallow grooves (SSP waveguide) on the same SiO_2 mask. The sample was subsequently etched again by

RIE until the depth of the shallow grooves reached 7 μm . At this point, the 10- μm active region around the laser mesa and outside the metasurface was total removed while there remained 1 μm on the bottoms of the deep grooves. This was followed by (and without removing the SiO_2 mask) multiple-angle Ti/Au electron-beam evaporation to ensure full coverage of metal on the whole structure. The Ti/Au layer on the laser top and sidewalls was then removed by a gold etchant and a dilute HF solution with a thick AZ4620 photoresist covering the metasurface region. The SiO_2 mask on top of the laser was then removed by RIE using a mixture of HF_4 and O_2 gases. The substrate was thinned to 120 μm and a 20/300 nm Ti/Au layer deposited to form the bottom contact. The samples were cleaved, indium-mounted on Cu submounts, wire-bonded, and finally attached to the cold finger of a cryostat for measurement.

4.4 EXPERIMENTAL RESULTS AND DISCUSSION

4.4.1 Light-current-voltage and far-field characterization

Devices with two different groove widths were fabricated, labeled as A and B (Fig. 4-7(a)). The groove widths of device A were 600 μm , whilst device B had a wider groove width of 1000 μm . Both devices show similar light-current-voltage (LIV) characteristics. Fig. 4-6 shows the LIV curves of a typical device as a function of temperature; the insets show the lasing spectra of devices A and B at 4.3 A and a 9 K heat sink temperature, the conditions at which their far field emission profiles were measured. The maximum operating temperature was found to be around 118 K under pulse mode operation with a 500 ns pulse width and 10 kHz repetition rate, comparable with that of a ridge laser (100 μm wide and 1500 μm long) fabricated from the same QCL active region, which operated up to 136 K under the same pulsing conditions.

Figure 4-6. Pulsed light-current-voltage (LIV) characteristics of device A at different heat sink temperatures. Inset: Lasing spectra of devices A and B at 4.3 A and 9 K – conditions under which their far-field emission profiles were measured.

2D far-field emission patterns of the devices were measured by scanning a pyroelectric detector on a spherical surface centered on the curved laser facet. 3D simulations were also performed using Lumerical FDTD. Fig. 4-7(c) shows the measured far-field pattern of device A, with Fig. 4-7(b) showing the line scans through the peak value. This reveals that the beam divergence in the ϕ direction is as low as $\sim 4^\circ$, while it is $\sim 10^\circ$ in the θ direction. Moreover, the background intensity is less than 10% of the peak value, indicating a high coupling efficiency of the THz radiation into the SSPs. Note that the measured beam divergence in the ϕ direction is larger than the simulated results at a wavelength of $94\ \mu\text{m}$ (Fig. 4-7(e)). This is due to the multi-mode emission of the devices (inset of Fig. 4-6) – the position of the radiation lobe shifts in the ϕ direction in the far-field as the laser wavelength changes. Simulations show that a change of $1\ \mu\text{m}$ in the wavelength leads to $\sim 1^\circ$ shift in the far-field pattern. Therefore, the measured far-field pattern is actually the superimposition of several slightly shifted far-field patterns if the device is not operating single-mode. This effect is more

apparent for device B, which has a wider emission spectrum (inset of Fig. 4-6). The measured and simulated far-field patterns of device B are shown in Figs 4-7(d) and (f), respectively. Here, the simulation considered the emission bandwidth from 92 μm to 98 μm , and closer agreement is obtained.

Figure 4-7. (a) SEM images of device A and B with groove widths of 600 μm and 1000 μm , respectively. (b) ϕ and θ direction line scans through the peak in the measured far-field radiation pattern in (c) for device A. (c) and (d) Measured far-field patterns of devices A and B, respectively. (e) Simulated far-field pattern of device A at the wavelength of 94 μm . (f) Simulated far-field patterns of device B taking into consideration the multi-mode emission from the active region at wavelengths between 92 μm and 98 μm .

4.4.2 Influence of the groove width on the far fields

In contrast to our original expectation that device B would give a narrower beam divergence in the θ direction as it is with a wider groove width, it actually has a larger beam divergence in the θ direction than device A. To understand this phenomenon, we investigated the near-field distributions of the two devices (Figs 4-8(a) and (b)). The SP is found to spread in the lateral direction so that device B indeed provides a broader laser emission width compared with device A. However, the spread of the SSP in the lateral direction induces phase delays of the electric field. Figs 4-8(c) and (d) plot the amplitude and phase distributions of the radiative electric field (E_y) along the white dashed lines in Figs 4-8(a) and (b). As shown in Figs 4-8(c) and (d), the phase of E_y of

device B varies greatly in the lateral (x -) direction, whereas it is almost uniform for device A. The large phase difference ($>180^\circ$) of the electric field between the central region and the lateral boundary region in device B leads to destructive interference in the far field, thereby giving a larger beam divergence.

Figure 4-8. Near field light intensity distributions of (a) device A and (b) device B on a logarithmic scale, taken on a plane 2 μm above the device surface. (c) and (d) Amplitude and phase distributions of the radiative electric field (E_y) along the white dashed line in (a) and (b), respectively.

4.4.3 Comparison between the devices with and without the metasurface

We have also fabricated several devices with just the second-order grating and without the metastructure. The farfield measurement results show that the coupling efficiency of such devices is lower due to a looser SSP confinement, resulting in a relatively stronger background and a slightly larger beam divergence, as expected (Fig. 4-9).

Figure 4-9. (a) Measured farfield pattern of a device with only the second order grating and without the metastructure. (b) Measured farfield pattern of a device with metastructure.

4.4.4 Advantages and disadvantages

Compared to a grating coupler built right into the metal-metal waveguide, the advantage of our structure is that it can provide a much larger emission area for collimation purpose (the beam divergence is inversely proportional to the emission area), or a much longer waveguide for photonic circuit applications. These are enabled by the low loss of our waveguide structure (recall that for groove depth of $7\text{ }\mu\text{m}$, the propagation length is 30 mm). If we build such a large coupler right into the metal-metal waveguide, we will have to supply a very large current ($>10\text{ A}$) to make the otherwise rather lossy waveguide transparent, and even larger current is needed should gain is desired, which is not very practical in reality. Another advantage of our structure is that our SSP structure has little influence on the optical mode in the laser cavity, allowing separated design of the laser cavity and the SSP structure. However, if one is to build a grating coupler into the metal-metal waveguide, careful care must be taken to avoid the undesired effects of the structure to the laser mode because the reflected wave can be quite easily re-injected into the laser cavity and distort the optical mode. On the other hand, the disadvantage of our structure is that the coupling efficiency of our structure may not be as high as the metal-metal waveguide.

4.5 CONCLUSION

In conclusion, we have demonstrated in-plane integration of THz tapered QCLs with metasurface waveguides. As an illustrative example, the metasurface waveguides were made into a plasmonic directional out-coupler for beam collimation of the generated THz wave from QCLs. The output beam is highly collimated with a beam divergence as narrow as $\sim 4^\circ \times 10^\circ$. Moreover, low background THz power (less than 10% of the peak value) indicates a high coupling efficiency of the light from the laser facet to the metasurface. We note that not only can the metasurface waveguides be processed into various geometries to shape the output emission profile, but they can also serve as a platform for integrated THz circuits for applications in sensing, spectroscopy, and communications.

5. INTEGRATED TERAHERTZ GRAPHENE MODULATOR WITH 100% MODULATION DEPTH

Terahertz (THz) frequency technology has the potential for important application in non-destructive imaging, spectroscopic sensing, and high bit-rate free space communications. An optical modulator is a key component for many of these applications. However, it has proved challenging to achieve high-speed modulation with a high modulation depth across a broad bandwidth of THz frequencies. Here, we demonstrate that through the combination of two advanced technologies in contemporary THz research, graphene and quantum cascade lasers, a fast modulator with a 100% modulation depth can be achieved. In the design, the graphene material interacts strongly with the laser field through a compact monolithic vertical integration. The small area of the device in comparison to existing THz modulators enables a faster modulation speed, greater than 100 MHz, which can be further improved through the laser cavity and modulator architecture designs. Moreover, as the graphene absorption spectrum is broadband in nature, our integration scheme can be readily scaled to other wavelength regions, such as the important but under-investigated mid-infrared regime, and applied to other optoelectronic devices.

5.1 INTRODUCTION

An optical modulator is a key component widely used for beam manipulation, imaging, optical communication, as well as active mode locking and others. However, the development of fast and efficient THz modulators is in its infancy, compared with the significant advances made in THz frequency lasers and detectors, where compact THz radiation source with 1 Watt output power [20] and several kinds of fast and sensitive THz detectors have been demonstrated [21,148,149].

Attempts to fabricate THz amplitude modulators have, for example, exploited semiconductor heterostructures containing a two-dimensional electron gas (2DEG) in which electrons can be accumulated or depleted by an applied gate voltage [150]. The modulation depth of such devices is ultimately limited by the achievable tunability in electron density, which is up to $\sim 1 \times 10^{12} \text{ cm}^{-2}$ for a 2DEG in conventional semiconductor [14]. These devices were initially found to modulate the THz amplitude by only a few percent, although through the incorporation of metamaterials or

plasmonic structures that enhanced the interaction between the THz radiation and the 2DEG [15,151], a modulation depth of 30% was achieved. Another promising material for efficient THz radiation modulation is graphene, a monolayer of carbon atoms with extraordinary electronic and optical properties. Its carrier concentration can be electrically tuned to as high as $1 \times 10^{14} \text{ cm}^{-2}$, and its natural bidimensionality and flexibility allows easy incorporation with other materials and structures. Recently a modulation depth of $\sim 15\%$ was achieved through electrical gating of a graphene sheet [12], which was increased to 64% through use of plasmonic or metamaterial structures, albeit with a narrow bandwidth [14,152,153]. However, further increase of the modulation depth has proven difficult. In addition, all the graphene modulators to date were studied as isolated components, with typically large active areas (tens of mm^2) required to facilitate optical alignment. The consequent large time constant of the effective RC circuit of the device package limited the modulation speed of such graphene modulators to only $\sim 13 \text{ MHz}$ [14]. Although this is similar to that found in semiconductor modulators [151], a much higher modulation speed ought be possible in graphene-based modulators owing to the material's ultra-high intrinsic carrier mobility ($> 20,000 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ for graphene on SiO_2) [154].

To explore the full potential of graphene, the monolithic integration of a graphene modulator with a THz radiation source is a promising solution. The benefits are three-fold: first, miniaturization will increase the modulation speed owing to reduced parasitic capacitance and resistance of the device; second, a larger modulation depth may be achieved as a stronger interaction of the THz radiation with the graphene layer is possible; and, finally, the integration eliminates one stage of optical alignment and the associated bulky mirrors or lenses.

Here, we report, for the first time, the realization of graphene modulators integrated with surface-emitting concentric-circular-grating (CCG) THz quantum cascade lasers (QCLs) [116,117]. By employing the CCG as a feedback element [116,117], steady single-mode THz QCL operation can be obtained with relatively high output power. We demonstrate that a 100% modulation depth can be achieved as a consequence of a strong interaction of the graphene with the laser field, which is greatly enhanced at the output apertures of the laser cavity. Moreover, the small device area of the device allows fast modulation that could be further improved through improved design of the laser cavity and the modulator architecture.

Furthermore, since graphene has a broadband absorption spectrum [155], our integration scheme can be readily scaled to other wavelength regions. For example, it can be easily extended to the important but under-investigated mid-infrared (mid-IR) regime, by monolithically integrating mid-IR QCLs with graphene.

5.2 DEVICE OVERVIEW

Fig. 5-1(a) shows a schematic illustration of the integrated device, which consists of an underlying surface-emitting CCG THz QCL with a graphene modulator whose Fermi level can be dynamically tuned by a voltage V_{gr} applied to the gating electrode. The active region of QCL, with a peak gain at around 3 THz, is sandwiched between a bottom gold plate and the upper CCG metal grating. The CCG was designed as a second-order grating such that single-mode THz radiation will be emitted vertically through the grating apertures, which is modulated by the electrically gated graphene. To facilitate graphene transfer, the active region of QCL was kept flat without etching. Thus, the laser cavity is formed solely by the CCG itself. Additionally, only the central several rings of the CCG (the orange region in Fig. 5-1(a)) are connected together with spoke-like metal bridges to allow electrically pumping over a small active region beneath. We pumped only part of the CCG, rather than the whole, to maintain the otherwise high current to a few amperes. With the correct CCG design, the optical modes are confined in the pumped area so that the QCL maintains a sufficiently low enough threshold current for lasing action (see Section 5.7.1). On top of the CCG, a SiO_2 layer insulates the QCL electrode from the graphene gate electrode, followed by the graphene sheet; the device fabrication is assisted by patterning the graphene gate electrode before the introduction of the graphene sheet. Fig. 5-1(b) shows optical microscope images of the central part of a fabricated device. The enlarged view in Fig. 5-1(c) shows the details of the graphene and QCL electrodes, which are insulated by the SiO_2 layer. A wrinkle, resulting from folding of the transferred chemical-vapor-deposition-grown (CVD) graphene, is visible at the lower right corner.

Figure 5-1. Overview of the integrated graphene modulator with quantum cascade laser. (a) Schematic illustration of the device. Only the central several rings of the circular-concentric grating (CCG) (orange region) are connected together with the spoke bridges to allow electrical pumping of the quantum cascade laser (QCL) over a small active region. Light is emitted vertically from the surface and is modulated by the electrically gated graphene. (b) Optical microscope images showing the central part of a fabricated device. (c) An enlarged view shows details of the graphene and the QCL electrodes, which are insulated by a 450-nm-thick SiO₂ layer. A wrinkle, resulting from folding of transferred chemical vapor deposition (CVD) graphene, is visible at the lower right corner.

5.3 INDIVIDUAL CHARACTERIZATIONS OF THE THz CCG QCL AND THE GRAPHENE

We first characterized the THz CCG QCL with the graphene electrode floating. The QCL lased under pulsed operation (500 ns pulse width, 10 kHz repetition rate) up to 102 K heatsink temperature (Fig. 5-2(a)), compared with ~135 K for a typical ridge laser fabricated from the same QCL wafer. Surface emitted single mode lasing was

obtained at all temperatures and pump currents. Fig. 5-2(b) shows the laser spectra as a function of current at a heatsink temperature of 20 K.

To characterize the properties of the CVD-grown graphene, a separate graphene modulator was fabricated on a SiO₂/Si substrate, as shown schematically in Fig. 5-2(c), using the same CVD graphene sheet (see Section 5.6). The 450-nm-thick SiO₂ layer was formed by plasmon-enhanced CVD (PECVD) and identical to that used for the QCLs. Raman spectroscopy was used to confirm that the graphene sheet after transfer was both high quality and a monolayer (Fig. 5-6). In view of the fact that the QCL-integrated devices will be operated under cryogenic condition, we investigated the electrical and optical absorption properties of the graphene at 78 K (the QCL surface and active region are typically several tens of Kelvin higher than the heatsink temperature). Using a simple model to fit the gate-voltage-dependent sheet resistance of the graphene (see Section 5.7.3), we obtained the gate-voltage-dependent sheet conductivity, carrier density and Fermi energy of the graphene, as plotted in Fig. 5-2(d). The inherent p-type doping of the CVD graphene is confirmed by the reduction in conductivity with increasing gate bias, up to the Dirac point ($V_g \sim 50$ V). The graphene conductivity and carrier density were found to change from 0.16 mS to 0.65 mS, and from $0.1 \times 10^{13} \text{ cm}^{-2}$ to $0.45 \times 10^{13} \text{ cm}^{-2}$, respectively, as the gate bias was increased from -40 V to $+60$ V. Correspondingly, the Fermi energy was tuned from -230 meV to $+20$ meV. The non-zero conductivity at the Dirac point is due to a residual carrier concentration of $0.1 \times 10^{13} \text{ cm}^{-2}$, resulting from charged impurities [156].

Although the use of a back-gate limits the widest range of carrier density and Fermi energy tuning possible (a top gating scheme could realize a carrier density as high as 10^{13} – 10^{14} cm^{-2}) [157], the tunability of the carrier density in graphene is still much larger than that of the 2DEGs in bulk semiconductors or quantum wells. As the optical absorption of graphene in THz region is dominated by intraband optical transitions (i.e., free carrier absorption), this larger tunability in carrier density, along with the generally higher mobility of graphene ($\sim 900 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ in our case), makes graphene a much more efficient THz wave modulator over the alternative semiconductor devices. Normalized to the maximum light transmission at the Dirac point, T_{CNP} , a modulation depth of $\sim 11\%$ in transmitted power ($T(V_g)/T_{\text{CNP}}$) was observed for our graphene modulator upon sweeping the gate voltage from -40 V to $+60$ V, measured at 78 K, using a standard ridge 3.0 THz QCL as the radiation source

(fabricated from the same wafer as used for the CCG QCL of our integrated device) (Fig. 5-2(e)). The effect of the Si substrate on the modulation was removed using a transfer matrix method [12], and the measured result agrees well with theoretical analysis (Fig. 5-2(e), see Section 5.7.4 for details). It is worth noting that the optical absorption of graphene in the THz region of the spectrum differs fundamentally from that at visible and near infrared frequencies, where interband optical transitions dominate, resulting in only a $\sim 2.3\%$ intensity modulation.

Figure 5-2. Individual characterization of the THz QCL and the graphene layer. (a) Light-current-voltage (LIV) characteristics of the THz CCG QCL under different heatsink temperatures. (b) Laser spectra as a function of pump current I , at 20 K. (c) Schematic representation of the structure used to characterize the graphene layer. (d) DC conductivity, carrier density and Fermi energy of the graphene as a function of gate voltage, measured at 78 K. (e) Gate voltage dependence of the modulation of the THz transmitted intensity (~ 3 THz) by the graphene, showing a modulation depth of $\sim 11\%$ over the applied bias range from -40 V to 60 V.

5.4 100% MODULATION DEPTH OF AN INTEGRATED GRAPHENE MODULATOR WITH CCG QCLs

A larger modulation strength is expected once the graphene modulator is integrated with the THz CCG QCLs owing to the electric field enhancement in the graphene near the output aperture of the CCG QCLs. This is accompanied by a similar enhancement on the insertion loss due to the non-zero absorption of graphene at the Dirac point. Fig. 5-3(a) plots the electric field enhancement factor, defined as the ratio of the amplitude in the near field to that in the far field, as a function of the distance above the CCG surface. For a SiO₂ spacing thickness of 450 nm, the amplitude of the electric field is enhanced by a factor of ~13 in the graphene region, corresponding to an intensity enhancement factor of ~170. The resulting enhanced modulating effect of graphene on the CCG QCL is shown in Fig. 5-3(b), where the intensity of the lasing peak varies from nearly zero at $V_G = -26$ V to a maximum at $V_G = +40$ V. During the measurement, the QCL was biased at 14.8 V (corresponding to an injected current of 2.8 A) in pulsed mode operation (500 ns pulse width, 10 kHz repetition rate) at a 20 K heatsink temperature. The graphene was gated at a series of DC biases V_G defined by $V_G = V_{QCL} - V_{gr}$, with V_{QCL} the voltage applied to the QCL electrode and V_{gr} the voltage applied to the graphene electrode (see Fig. 5-1(a)). The circles in Fig. 5-3(c) show the average laser output as a function of V_G , which is consistent with the results in Fig. 5-2(d) and Fig. 5-2(e). As a comparison, a control device without the graphene sheet was fabricated, whose output power as a function of V_G is plotted as the rectangles in Fig. 5-3(c), and no obvious modulation effect was observed. Note that with presence of the graphene, the output power decreases significantly, the power of the integrated device at $V_G = +40$ V is around a quarter of that of the control device. To verify the effect of the graphene modulator further, we measured the light-current curves of the laser at a series of graphene gate voltages, as shown in Fig. 5-3(d); the integrated device allows a 100% modulation depth of the THz radiation amplitude.

Figure 5-3. Modulation depth of the integrated graphene modulator on CCG QCL. (a) Enhancement factor of the amplitude of the electric field in the graphene region near the output aperture of the CCG QCL as a function of distance above the CCG. The inset shows a magnified portion of the curve. (b) Enhanced modulation of the THz wave by the integrated graphene modulator. The intensity of the lasing peak varies from nearly zero at $V_G = -26$ V to a maximum at $V_G = +40$ V. (c) V_G dependence of the output power of the CCG QCL with (circles) and without (rectangles) the graphene. (d) Light-current characteristics of the QCL as a function of V_G confirming the 100% modulation depth achieved by the integrated graphene modulator.

It should be noted that as the graphene interacts with the cavity mode, it changes the loss of the cavity mode (Fig. 5-4), which is inevitable for this kind of intimate integration. In this sense, it is an internal modulator. Compared to the direct modulation case where the laser itself is switched on and off, our scheme allows the laser to operate more stably – the pumped current and, thus, the refractive index and working temperature are remained unchanged. In contrast, the varying refractive index resulting from the varying injected current and temperature in directly-modulated laser

makes its frequency more fluctuating, an effect called frequency chirping. Therefore, our scheme may be advantageous in applications where the chirping effect is a problem. By saying so, we make no pretension that our devices totally get rid of the chirping effect – after all, the electron density in the graphene varies during operation. However, it could be expected that our devices have a smaller chirping effect thanks to the unvarying current applied to the laser and the small power consumption of the graphene modulator.

Figure 5-4. Calculated losses for the sixth-azimuthal modes with the graphene gated at two different Fermi levels. The loss of the lasing mode increases by 2.8 cm^{-1} when the Fermi level of graphene is tuned from Dirac Point to -220 meV .

5.5 MODULATION SPEED OF THE INTEGRATED GRAPHENE MODULATOR

Due to the lack of a fast THz detector, we cannot observe the time-varying THz modulated signal directly to determine the operational speed of the modulator. Therefore, we estimate the modulation speed by monitoring the average output power of the device while an AC voltage signal is applied to the graphene electrode, as shown in Fig. 5-5(a)(i). The pulse duration for the QCL is fixed at $T_{\text{QCL}} = 1 \text{ } \mu\text{s}$ (corresponding to a frequency of $f_{\text{QCL}} = 1 \text{ MHz}$), while the AC rectangular signal on the graphene has a 50% duty cycle and a period of $T_{\text{G}} = T_{\text{QCL}}/N$ ($f_{\text{G}} = Nf_{\text{QCL}}$), where N is an integer. If the graphene modulator can follow the variation of the applied voltage, the output optical signal of the device will be as that shown in Fig. 5-5(a)(ii), and the average power P_{ave} detected experimentally should be $(P(V_{\text{gr}}) + P(V_0))/2$ regardless of

the modulation frequency, where $P(V_{\text{gr}})$ and $P(V_0)$ are the output peak powers when constant gating voltages V_{gr} and V_0 are applied to the graphene electrode, respectively. Otherwise, if the applied signal is much faster than the response speed of the graphene modulator, the optical output power will adopt the form shown in Fig. 5-5(a)(iii), and P_{ave} will deviate from $(P(V_{\text{gr}}) + P(V_0))/2$. Therefore, one could expect a flat P_{ave} vs f_G curve up to a cut-off point, where the corresponding f_G is approximately the modulation speed of the device. The measured P_{ave} vs f_G curve is shown in Fig. 5-5(b), in which the blue line (star symbols) corresponds to $V_0 = 0$ V and $V_{\text{gr}} = 10$ V, while the red line (circles) corresponds to $V_0 = 0$ V and $V_{\text{gr}} = 15$ V. The grey, blue and red shaded ribbons indicate the output powers when the graphene electrode is DC biased at 0 V, 10 V and 15 V, respectively. The widths of the ribbons indicate the instability of the laser power during the measurement period. A flat dependency was observed up to 12 MHz, the limit of our experimental facilities, with no cut-off point, which already makes it to be one of the fastest THz modulators. To extend the measurement range, we investigated the dynamic response (S_{21}) using a radio frequency (RF) network analyzer. Using this together with an equivalent circuit model, we obtained the frequency response of the integrated modulator (Fig. 5-5(c); see Section 5.7.5 for details). The modulation speed can therefore be estimated to be 110 MHz from the 3-dB cutoff point.

The electrical modulation on the graphene sheet is given by (Section 5.7.5)

$$\begin{aligned} \frac{V_G}{V_S} &= \frac{1}{1 + i\omega R_0 C_p^t + R_0 / (R_G^t + 1 / i\omega C_G^t)} \times \frac{1}{1 + i\omega R_G^t C_G^t} \\ &\approx \frac{1}{1 + i\omega R_0 (C_p^t + C_G^t)} \times \frac{1}{1 + i\omega R_G^t C_G^t} \end{aligned} \quad (5-1)$$

where V_S and V_G are the amplitude of the voltage on the graphene electrode and that applied to the graphene sheet, respectively, R_0 is the internal impedance of the RF source, R_G^t is the effective graphene resistance, C_G^t and C_p^t are the total capacitance of the graphene sheet and the metal contacts, respectively, and ω is the angular frequency of the modulation. In Eq. (5-1), we assumed $R_G^t \ll 1 / i\omega C_G^t$ for our case. Two time constants $\tau_1 = R_0 (C_p^t + C_G^t)$ and $\tau_2 = R_G^t C_G^t$ enter the equation and determine the cutoff frequency. The first time constant can be decreased by reducing the size of the graphene back gate (i.e., the QCL bias contact), which requires a smaller cavity

design. The second time constant can be decreased by reducing the widths of the graphene electrode slits (this makes the effective area of the graphene sheet smaller so that the graphene capacitance is smaller) and by reducing the graphene resistance. This is dominated by the contact resistance at the metal/graphene edges and can be reduced though several techniques [158–160].

Figure 5-5. Measurement of the modulation speed of the integrated graphene modulator. (a) (i) Device bias scheme. The pulse duration for the QCL was fixed at $T_{QCL} = 1 \mu s$ (corresponding to a frequency of $f_{QCL} = 1 \text{ MHz}$), while an AC rectangular signal was applied onto the graphene electrode with 50% duty cycle and various periods of $T_G = T_{QCL} / N$ ($f_G = N f_{QCL}$), where N is an integer. (ii) The predicted output optical signal of the device if the graphene modulator is able to follow the variation of the applied voltage. In this case, the average peak power $P_{ave} = (P(V_{gr}) + P(V_0))/2$, where $P(V_{gr})$ and $P(V_0)$ are the output peak powers when a constant V_{gr} or V_0 is applied to the graphene electrode, respectively. (iii) The predicted output optical signal of the device if the applied signal on graphene is much faster than the speed of the modulator, in which case P_{ave} deviates from $(P(V_{gr}) + P(V_0))/2$. (b) P_{ave} as a function of modulation frequency. The blue line (star symbols) corresponds to $V_0 = 0 \text{ V}$ and $V_{gr} = 10 \text{ V}$, while the red line (circle symbols) corresponds to $V_0 = 0 \text{ V}$ and $V_{gr} = 15 \text{ V}$. The grey, blue and red shaded ribbons indicate the output power when the graphene electrode is DC biased at 0 V , 10 V and 15 V , respectively. The widths of the ribbons manifest the instability of the laser power during measurement period. (c) Frequency response of the graphene modulator.

5.6 FABRICATION

Preparation of the graphene film.

The commercially-sourced CVD-grown graphene on copper foil was spin-coated with a polymethyl-methacrylate (PMMA) film, which was then fully cured in a 110°C oven for 5 min resulting in a PMMA thickness of $\sim 1 \mu\text{m}$. The copper substrate was etched by floating the PMMA/graphene/copper sheet on an ammonium persulfate (0.1 M, Sigma Aldrich) solution for >3 hours. After rinsing, the PMMA/graphene film was ready for transfer.

Fabrication of the isolated graphene modulator.

A 450-nm-thick layer of SiO_2 was blanket deposited on a lightly doped p-type Si substrate (resistivity $1 - 20 \Omega\cdot\text{cm}$) using PECVD. The source, drain and back electrodes were formed by electron-beam evaporation, after which the PMMA/graphene film ($1 \text{ cm} \times 1 \text{ cm}$) was transferred and contacted with source and drain electrodes. The sample was dried in air, followed by annealing in nitrogen at 160 °C for 3 hours. The PMMA layer was then removed in acetone.

Fabrication of the integrated devices with graphene modulator and CCG QCL.

The active region of the THz QCL was grown by molecular beam epitaxy on an undoped GaAs substrate. Fabrication of the devices commenced with the gold-gold thermocompression bonding of the active region to an n+ GaAs receptor wafer. The original QCL substrate was then removed through lapping and selective chemical etching. This was followed by the removal of the highly absorbing contact layer of the active region to prevent attenuation of the THz radiation that couples out of the grating slits. Top metal gratings (Ti/Au/Ti 15/300/10 nm) were defined by standard optical lithography and lift-off; the top 10 nm Ti layer was used as an adhesive between the Au and the subsequent SiO_2 insulation layer, which was grown by PECVD. After patterning the SiO_2 layer to allow electrical access to the QCL, another optical lithography and lift-off process was carried out to define the graphene electrode on top of the SiO_2 /CCG rings. A graphene sheet was then transferred onto the graphene electrode, followed by annealing in nitrogen at 160 °C for 3 hours to relax the PMMA/graphene film slowly and make full contact with the surface underneath. After

removing the PMMA layer with acetone, the graphene was patterned to remove unwanted material area and avoid contacting the other electrodes. The samples were then cut and indium-mounted onto Cu submounts, wire-bonded, and finally attached to the cold finger of a cryostat for measurement.

5.7 SUPPLEMENTARY INFORMATION

5.7.1 Farfield and optical mode of the concentric-circular-grating (CCG) QCL

The design of the concentric-circular grating follows the rule in Chapter 2, the whole grating structure starting from the center to the boundary is as follows: 59.8/3/27.4/3/26.3/3/26.6/3/26.5/3/26.4/3/27.4/3/27.4/3/27.4/3/27.4/3/27.4/3/27.9/3/27.9/3/27.9/3/150 in mm, where the bold number indicates the slit region, and the underlined parts are connected by spoke structures to allow electrically pumping their underneath active region (Fig. 5-1).

Two-dimensional far-field emission pattern of the device has been measured by scanning a pyroelectric detector on a spherical surface centered at the laser, as shown in Fig. 5-6(a), where the (0, 0) position represents the normal to the laser surface. According to the measured far-field pattern, we were able to identify the optical mode in the laser cavity as that in Fig. 5-6(b) and Fig. 5-6(c). As it is shown, the optical mode is confined in the pumping area (enclosed by a white dash circle) of the CCG QCL. Note that the actual pumped area is slightly larger than that enclosed by the white dash circle in Fig. 5-6(b) or line in Fig. 5-6(c) considering the lateral current spreading in the active region, which is around 20 μm .

According to the optical mode, the CCG QCL operates at the sixth-order azimuthal mode although the design was target on the first-order mode. This deviation is mainly due to the underestimation of the gain peak frequency of the active region.

Figure 5-6. Farfield and optical mode of the CCG QCL. (a) Measured two-dimensional farfield emission pattern of the surface-emitting CCG QCL, where the (0, 0) position represents the normal to the laser surface, and the corresponding electric field (E_z) distribution of the laser in top view (b) and cross-section view (c). The white dash line enclosed the pumped area.

5.7.2 Raman Spectra of the transferred graphene

Fig. 5-7(a) shows the Raman spectrum of the transferred graphene on the SiO_2/Si substrate, and Fig. 5-7(b) shows the Raman spectrum of the transferred graphene in different slits of the concentric-circular grating of the final device. Compared with Fig. 5-7(a), the additional small peaks in Fig. 5-7(b) around 2250cm^{-1} is likely from the chemical residue in the slits. Overall, the near absent D peak, single-Lorentzian-shape 2D peak and the 2D/G intensity ratio (~ 3) of each spectrum confirm the high quality of the graphene after transfer.

Figure 5-7. Raman characterization of the transferred graphene. Raman Spectra of the transferred graphene on (a) SiO₂/p-Si substrate and (b) in the slits of the CCG of the integrated device.

5.7.3 Model used to retrieve the graphene parameters

The carrier density in the graphene can be approximately expressed by [161]

$$n_{tot} = \sqrt{n_0^2 + n(V_g)^2} \quad (5-2)$$

Where n_0 represents residual carrier concentration at the Dirac point, which is non-zero owing to charged impurities in the dielectric or the graphene/dielectric interface. [156] $n(V_g)$ represents the carrier concentration induced by the electric gating, and can be obtained from a simple parallel capacitor model:

$$V_g - V_{CNP} = \frac{e}{c_{ox}} n + \frac{E_f}{e} \approx \frac{e}{c_{ox}} n \quad (5-3)$$

Where $c_{ox} = \epsilon_0 \epsilon_{ox} / t$, with ϵ_0 being the permittivity of free space and ϵ_{ox} the dielectric constant of SiO₂ (~3.9), is the geometrical capacitance. For oxide thickness of $t=450\text{nm}$, $c_{ox} = 8 \times 10^{-9} \text{ F / cm}^2$.

The square resistance across the source and drain is given by [161]

$$R_{tot} = R_{contact} + \frac{1}{en_{tot}u} \quad (5-4)$$

Where $R_{contact}$ represents the metal/graphene contact resistance.

By fitting the measured data with the above model, as shown in Fig. 5-8, we were able to extract the relevant parameters: $n_0 = 1.0 \times 10^{12} / \text{cm}^2$, $u = 924 \text{cm}^2 \text{V}^{-1} \text{s}^{-1}$, $V_{\text{CNP}} = 50.2 \text{V}$ and $R_{\text{contact}} = 440 \Omega$.

The graphene sheet conductivity in Fig. 5-2(d) is given by $1 / (R_{\text{tot}}(\text{measured}) - 440 \Omega)$, the Fermi level is calculated by $E_f = \hbar v_f \sqrt{\pi n_{\text{tot}}}$, where $v_f \approx 1 \times 10^6 \text{ms}^{-1}$ is the Fermi velocity [14].

Figure 5-8. Square resistance of the CVD graphene transistor as a function of the gate voltage. Symbols: measured data; Curve: modeling result.

5.7.4 Comparison of the graphene response at 78 K and 300 K

To investigate the graphene response at cryogenic and room temperature, we measured the electrical transport property and the optical modulation of the same graphene sheet at 78 K and 300 K temperature. As plotted in Fig. 5-9(a), at 78 K temperature, the graphene undergoes a larger change of conductivity across the gate voltage range. Treating graphene as a zero thickness conductive layer with free carriers and current, derived from Maxwell equations, the transmission intensity at the air/graphene/dielectric interface can be expressed as:

$$\frac{T(\omega, V_G)}{T(\omega, V_{\text{CNP}})} = \left(\frac{1 + n_{\text{diel}} + Z_0 \sigma_0(\omega)}{1 + n_{\text{diel}} + Z_0 \sigma(\omega, V_G)} \right)^2 \quad (5-5)$$

where n_{diel} is the refractive index of the dielectric, $Z_0 = 376.73 \Omega$ is the vacuum impedance, $\sigma_0(\omega)$ and $\sigma(\omega, V_G)$ are the graphene optical conductivity at the Dirac point and V_G gate voltage, respectively. The optical conductivity $\sigma(\omega, V_G)$ is related to the DC conductivity $\sigma(V_G)$ measured electrically as $\sigma(\omega, V_G) = \sigma(V_G) / (1 + \omega^2 \tau^2)$ with τ being the carrier momentum scattering time (~ 15 fs) [14,162]. The calculated $T(V_G)/T_{CNP} - V_G$ curves of the THz wave (~ 3.2 THz) at the air/graphene/SiO₂ interface is also plotted in Fig. 5-9(a), showing a slightly higher modulation depth ($\sim 1\%$) at 78 K than that at 300 K. However, the uncertainty in the THz modulation measurement smears out this small difference (Fig. 5-9(b)).

Figure 5-9. Comparison of the graphene response at 78 K and 300 K. (a) Calculated transmittance normalized to the value at the Dirac point and the electrical transport measurements of the graphene sheet at 78 K and 300 K. (b) Modulation of the THz radiation by the graphene at 78 K and 300 K with the effect of the Si substrate removed.

5.7.5 High-frequency circuit model of the graphene modulator

To investigate the high speed performance of the integrated modulator, we built an equivalent circuit model as will be discussed below. Fig. 5-10(a) presents the schematic of the modulation scheme, a function generator with output internal impedance $R_0 = 50 \Omega$ is applied between the two electrodes that were insulated by the SiO₂ layer. In this configuration, the main parts that affect the high frequency performance of the integrated modulator are shown in Fig. 5-10(b), whose cross-section view is schematically shown in Fig. 5-10(c). The slits are numbered as 1, 2, 3, ..., . The circuit

model for a single slit marked by a dashed box is also shown, where R_G^L and R_G^R are respectively the resistance of the graphene sheet from the left and right contact edge to the center of the sheet, C_p^L and C_p^R are respectively the capacitance of the left and right contact pads to the back gate, and C_G is the capacitance between the graphene sheet and the back gate. This circuit can be simplified as that shown in the lower right model in Fig. 5-10(c), where the parameters can be written as:

$$R_G = \frac{1}{1/R_G^L + 1/R_G^R} \approx \frac{\rho_c + d/2\sigma}{4\pi r} \quad (5-6)$$

$$C_G = C_p^L + C_p^R = \frac{\varepsilon_{ox}\varepsilon_0(2\pi rd)}{t} \quad (5-7)$$

where ρ_c is the contact resistance across the metal/graphene edge in unit of $\Omega \cdot \mu m$, σ is the graphene conductivity, r is the distance from the center of the slit to the center of the device, t is the thickness of the SiO₂ (450 nm), d is the width of the slits (14 μm), ε_0 is the vacuum permittivity and ε_{ox} is the relative permittivity of the SiO₂.

It is easily found that the slits are in parallel in the circuit, therefore, the equivalent circuit for the whole modulator is as that shown in Fig. 5-10(d). From the previous two equations, we have

$$\frac{R_G^1}{C_G^1} = \frac{R_G^2}{C_G^2} = \dots = \frac{R_G^6}{C_G^6}. \quad (5-8)$$

This means the voltages at the red points in Fig. 5-10 are equal, we can therefore simplified the circuit model as the one shown in Fig. 5-10(e), where

$$\begin{aligned} R_G^t &= \frac{1}{1/R_G^1 + 1/R_G^2 + \dots + 1/R_G^6} = \frac{\rho_c + d/2\sigma}{4\pi} \left(\frac{1}{r_1 + r_2 + \dots + r_6} \right) \\ &\approx \frac{\rho_c}{10^4 \mu m} + \frac{2 \times 10^{-4}}{\sigma} \\ &\approx 180 \Omega \end{aligned} \quad (5-9)$$

$$\begin{aligned} C_G^t &= C_G^1 + C_G^2 + \dots + C_G^6 = \frac{2\pi\varepsilon_{SiO_2}\varepsilon_0 d}{t} (r_1 + r_2 + \dots + r_6) \\ &\approx 6.5 pF \end{aligned} \quad (5-10)$$

$$C_p^t = C_p^1 + C_p^2 + \dots + C_p^6 = \frac{\varepsilon_{SiO_2} \varepsilon_0 A_p}{t} \approx 8.7 \text{ pF} \quad (5-11)$$

with A_p being the total area of the graphene contact for slit 1 to 6. In Eq. 5-9, we have used $\rho_c = (R_{contact} / 2) \times l_{contact} \approx (440 \Omega / 2) \times 8000 \mu\text{m}$, where $R_{contact}$ is the contact resistance obtained in Section 5.7.3 and $l_{contact}$ is the length of the graphene/metal edge of that separated device. Based on the circuit model, the applied gate voltage can be calculated as

$$V_p = V_s \times \frac{Z_{in}}{Z_{in} + R_0} \quad (5-12)$$

where Z_{in} is the total impedance of the modulator, enclosed by the dashed box in Fig. 5-10(e), and can be written as

$$Z_{in} = \frac{[R_G^t + (1 / i\omega C_G^t)](1 / i\omega C_p^t)}{R_G^t + (1 / i\omega C_G^t) + (1 / i\omega C_p^t)}. \quad (5-13)$$

where $\omega = 2\pi f$ with f being the frequency of the modulation signal V_s . The signal that actually driven the carriers in and out of the graphene is

$$\begin{aligned} V_G &= V_s \times \frac{Z_{in}}{Z_{in} + R_0} \times \frac{1}{1 + i\omega R_G^t C_G^t} \\ &= V_s \times \frac{1}{1 + i\omega R_0 C_p^t + R_0 / (R_G^t + 1 / i\omega C_G^t)} \times \frac{1}{1 + i\omega R_G^t C_G^t} \end{aligned} \quad (5-14)$$

To characterize the high-frequency response of the integrated modulator, we have performed an S-parameters measurement using a radio frequency (RF) network analyzer, the dynamic response (S_{21}) is shown in Fig. 5-11 (blue triangular symbol). The relation between S_{21} and Z_{in} for the circuit in Fig. 5-10(e) is [163]

$$S_{21} = \frac{2Z_{in}}{2Z_{in} + R_0} \quad (5-15)$$

Combine equation (5-13) – (5-15), we get

$$V_G = V_s \times \frac{1}{2 / S_{21} - 1} \times \frac{1}{1 + i\omega R_G^t C_G^t} \quad (5-16)$$

Using the estimated values $R_G^t \approx 180 \Omega$, and $C_G^t \approx 6.5 pF$, we are able to plot the normalized modulation depth (V_G/V_s) of the electrical signal applied to the graphene sheet as a function of the modulation frequency (black circular symbol in Fig. 5-11). The estimated 3-dB cutoff frequency is 110 MHz.

Figure 5-10. Equivalent circuit model of the integrated graphene modulator. (a) Schematic of the modulation scheme, the internal impedance of the function generator is R_0 (50Ω). (b) Main parts that affect the high speed performance of the integrated modulator. (c) cross-section view of modulator in (b) and the equivalent circuit for a single slit. (d) equivalent circuit for the whole modulator. (e) simplified circuit model for the modulator.

Figure 5-4. Frequency response of the integrated graphene modulator measured by a RF network analyzer. The blue triangular symbol represents the S_{21} response of the device and the black circular symbol corresponds to the electrical modulation applied to the graphene sheet, whose 3-dB cutoff frequency was estimated to be 110 MHz.

5.8 CONCLUSION

A monolithically integrated graphene modulator with surface-emitting concentric-circular-grating (CCG) THz quantum cascade lasers (QCLs) was reported with a 100% modulation depth and a fast response speed. The former was a consequence of a strong interaction of the graphene with the laser field, which was further enhanced at the output apertures of the laser cavity, while the latter is owing to the reduced device area enabled by the integration. An even faster operation frequency of the integrated THz modulator could be achieved through proper design of the device's architecture. Our integration scheme can also apply to Mid-infrared or Near-infrared frequency regime as the optical absorption of graphene is naturally broadband.

6. CONCLUSIONS AND FUTURE WORKS

In this thesis, I have described the engineering of terahertz (THz) quantum cascade laser (QCL) in terms of band structure, resonator, beam collimation and intensity modulation. In the course of this thesis, I have developed a series of recipes and techniques used to fabricate the devices including wafer bonding (Au-Au thermocompression method and Au-In reactive method), wet and dry etchings, material depositions, etc. In addition, various experimental setups have been built to automatize the device characterization.

At the beginning of this thesis, an overview of THz technologies was given, followed by an introduction of the development of the QCLs and the roles they have played in the THz areas. Chapter 2 reviewed the theoretical basics of the THz QCL and a comparison between the major kinds of the active region design. An indirectly pumped bound-to-continuum design was then proposed. Preliminary experimental results showed a very low threshold current density, which was attributed to the suppression of the parasitic leakage current prior to the threshold. However, the small dynamic range limited its temperature performance. Future works are to increase the rollover current density by, e.g., adjusting the coupling between the injection states and increasing the doping level of the structure.

Chapter 3 demonstrated a novel concentric-circular grating based resonator for achieving single-mode surface-emitting THz QCL with low beam divergence. The designed, fabrication and experimental characterization of the device has been discussed. Thanks to the circular symmetry of the structure, the design of the structure is much simpler than the photonic crystal resonators while inherits their benefits. Robust single-mode operation was achieved with a side-mode suppression ratio of ~ 30 dB. The device emits ~ 5 times the power of a ridge laser of similar dimensions with little degradation in the maximum operation temperature. Two lobes are observed in the far-field emission pattern, each of which has a divergence angle as narrow as $\sim 13^\circ \times 7^\circ$. Future efforts will be devoted to the demonstration of a single-lobe far-field pattern with narrower beam divergence, or a ring-shaped far-field pattern with radial polarization which is desirable for coupling the radiation to metal wire THz waveguides.

In chapter 4, I have described the integration of tapered THz QCLs with metasurfaces, which are surface plasmonic waveguides developed to provide a

versatile platform for THz beam engineering and, hopefully, active THz photonic circuit. As an illustrative example, the metasurface has been made into an efficient beam collimator by introducing periodically arranged scatterers. The resultant surface-emitting beams are highly directional with a small divergence of $\sim 4^\circ \times 10^\circ$. Future works are to apply the well-established surface plasmon techniques to the platform to realize more complex THz beam control, e.g., Airy beam, optical vortex, coherent beam combining, etc. More importantly, other optoelectronic components could be incorporated to construct a more functional device, a conceptual example is depicted in Fig. 6-1.

Finally, chapter 5 reported, for the first time, the realization of graphene modulators integrated with THz QCLs. A 100% modulation depth has been achieved as a result of a greatly enhanced interaction of the graphene with the laser field, enabled by the intimate integration scheme. Moreover, the small device area of the device allows fast modulation that could be further improved through improved design of the laser cavity and the modulator architecture, which is one direction of the future researches. Other future researches may be to realize mode locking of THz QCLs with graphene, or to implement the integrated graphene as a fast THz photodetector which is used to monitor and stabilize the THz power.

Figure 6-1. Schematic of a lab-on-a-chip device for molecule sensing application.

PUBLICATIONS

1. **G. Liang**, H. Liang, Y. Zhang, S. P. Khanna, L. Li, A. Giles Davies, E. Linfield, D. Fatt Lim, C. Seng Tan, S. Fung Yu, H. Chun Liu, and Q. J. Wang, "Single-mode surface-emitting concentric-circular-grating terahertz quantum cascade lasers," **Appl. Phys. Lett.** 102, 031119 (2013). (***Featured in Nature Photonics, April 2013***)
2. **G. Liang**, H. Liang, Y. Zhang, L. Li, a. G. Davies, E. Linfield, S. F. Yu, H. C. Liu, and Q. J. Wang, "Low divergence single-mode surface-emitting concentric-circular-grating terahertz quantum cascade lasers," **Opt. Express** 21, 31872 (2013). (***Featured in Laser Focus World, Feb. 2014***)
3. **G. Liang**, E. Dupont, S. Fatholouloumi, Z. R. Wasilewski, D. Ban, H. K. Liang, Y. Zhang, S. F. Yu, L. H. Li, A. G. Davies, E. H. Linfield, H. C. Liu, and Q. J. Wang, "Planar integrated metasurfaces for highly-collimated terahertz quantum cascade lasers.," **Sci. Rep.** 4, 7083 (2014).
4. **G. Liang**, N. Hu, X. Yu, L. H. Li, A. G. Davies, E. H. Linfield, H. K. Liang, Y. Zhang, Q. J. Wang, "Integrated terahertz graphene modulator with 100% modulation depth," Submitted, (2015).
5. H. K. Liang, B. Meng, **G. Liang**, J. Tao, Y. Chong, Q. J. Wang, and Y. Zhang, "Electrically pumped mid-infrared random lasers," **Adv. Mater.** 25, 6859–63 (2013).
6. X. Yu, J. Tao, Y. Shen, **G. Liang**, T. Liu, Y. Zhang, and Q. J. Wang, "A metal-dielectric-graphene sandwich for surface enhanced Raman spectroscopy," **Nanoscale** 6, 9925–9 (2014).

BIBLIOGRAPHY

1. W. L. Chan, J. Deibel, and D. M. Mittleman, "Imaging with terahertz radiation," *Reports Prog. Phys.* **70**, 1325–1379 (2007).
2. S. M. Kim, F. Hatami, J. S. Harris, A. W. Kurian, J. Ford, D. King, G. Scalari, M. Giovannini, N. Hoyler, J. Faist, and G. Harris, "Biomedical terahertz imaging with a quantum cascade laser," *Appl. Phys. Lett.* **88**, 153903 (2006).
3. K. L. Nguyen, M. L. Johns, L. Gladden, C. H. Worrall, P. Alexander, H. E. Beere, M. Pepper, D. A. Ritchie, J. Alton, S. Barbieri, and E. H. Linfield, "Three-dimensional imaging with a terahertz quantum cascade laser," *Opt. Express* **14**, 2123–9 (2006).
4. A. W. M. Lee, Q. Qin, S. Kumar, B. S. Williams, Q. Hu, and J. L. Reno, "Real-time terahertz imaging over a standoff distance (>25 meters)," *Appl. Phys. Lett.* **89**, 141125 (2006).
5. Q. Hu, "Terahertz quantum cascade lasers and real-time T-rays imaging at video rate," *Terahertz Sci. Technol.* **2**, 120–130 (2009).
6. D. M. Mittleman, R. H. Jacobsen, R. Neelamani, R. G. Baraniuk, and M. C. Nuss, "Gas sensing using terahertz time-domain spectroscopy," *Appl. Phys. B Lasers Opt.* **67**, 379–390 (1998).
7. J. F. Federici, B. Schulkin, F. Huang, D. Gary, R. Barat, F. Oliveira, and D. Zimdars, "THz imaging and sensing for security applications—explosives, weapons and drugs," *Semicond. Sci. Technol.* **20**, S266–S280 (2005).
8. Y. Ren, J. N. Hovenier, R. Higgins, J. R. Gao, T. M. Klapwijk, S. C. Shi, B. Klein, T.-Y. Kao, Q. Hu, and J. L. Reno, "High-resolution heterodyne spectroscopy using a tunable quantum cascade laser around 3.5 THz," *Appl. Phys. Lett.* **98**, 231109 (2011).
9. S. Koenig, D. Lopez-Diaz, J. Antes, F. Boes, R. Henneberger, a. Leuther, a. Tessmann, R. Schmogrow, D. Hillerkuss, R. Palmer, T. Zwick, C. Koos, W. Freude, O. Ambacher, J. Leuthold, and I. Kallfass, "Wireless sub-THz communication system with high data rate," *Nat. Photonics* **7**, 977–981 (2013).
10. T. Kleine-Ostmann and T. Nagatsuma, "A Review on Terahertz Communications Research," *J. Infrared, Millimeter, Terahertz Waves* **32**, 143–171 (2011).
11. Z. Chen, Z. Y. Tan, Y. J. Han, R. Zhang, X. G. Guo, H. Li, J. C. Cao, and H. C. Liu, "Wireless communication demonstration at 4.1 THz using quantum cascade laser and quantum well photodetector," *Electron. Lett.* **47**, 1002 (2011).

12. B. Sensale-Rodriguez, R. Yan, M. M. Kelly, T. Fang, K. Tahy, W. S. Hwang, D. Jena, L. Liu, and H. G. Xing, "Broadband graphene terahertz modulators enabled by intraband transitions.," *Nat. Commun.* **3**, 780 (2012).
13. L. Vicarelli, M. S. Vitiello, D. Coquillat, a Lombardo, a C. Ferrari, W. Knap, M. Polini, V. Pellegrini, and a Tredicucci, "Graphene field-effect transistors as room-temperature terahertz detectors.," *Nat. Mater.* **11**, 865–71 (2012).
14. S. H. Lee, M. Choi, T.-T. Kim, S. Lee, M. Liu, X. Yin, H. K. Choi, S. S. Lee, C.-G. Choi, S.-Y. Choi, X. Zhang, and B. Min, "Switching terahertz waves with gate-controlled active graphene metamaterials.," *Nat. Mater.* **11**, 936–41 (2012).
15. H.-T. Chen, W. J. Padilla, J. M. O. Zide, A. C. Gossard, A. J. Taylor, and R. D. Averitt, "Active terahertz metamaterial devices.," *Nature* **444**, 597–600 (2006).
16. M. Choi, S. Lee, Y. Kim, S. Kang, and J. Shin, "A terahertz metamaterial with unnaturally high refractive index," *Nature* **470**, 369–373 (2011).
17. C. M. Watts, D. Shrekenhamer, J. Montoya, G. Lipworth, J. Hunt, T. Sleasman, S. Krishna, D. R. Smith, and W. J. Padilla, "Terahertz compressive imaging with metamaterial spatial light modulators," *Nat. Photonics* **8**, 605–609 (2014).
18. R. Köhler, A. Tredicucci, F. Beltram, H. E. Beere, E. H. Linfield, A. G. Davies, D. A. Ritchie, R. C. Iotti, and F. Rossi, "Terahertz semiconductor-heterostructure laser," *Nature* **417**, 156–159 (2002).
19. S. Jung, A. Jiang, Y. Jiang, K. Vijayraghavan, X. Wang, M. Troccoli, and M. a Belkin, "Broadly tunable monolithic room-temperature terahertz quantum cascade laser sources.," *Nat. Commun.* **5**, 4267 (2014).
20. L. Li, L. Chen, J. Zhu, J. Freeman, P. Dean, A. Valavanis, A. G. Davies, and E. H. Linfield, "Terahertz quantum cascade lasers with >1 W output powers," *Electron. Lett.* **50**, 309–311 (2014).
21. H. C. Liu, C. Y. Song, A. J. SpringThorpe, and J. C. Cao, "Terahertz quantum-well photodetector," *Appl. Phys. Lett.* **84**, 4068 (2004).
22. J. Chamberlain, J. Gibbs, and H. Gebbie, "Refractometry in the far infra-red using a two-beam interferometer," *Nature* **198**, 874 (1963).
23. M. Tonouchi, "Cutting-edge terahertz technology," *Nat. Photonics* **1**, 97–105 (2007).
24. N. Nagai, R. Kumazawa, and R. Fukasawa, "Direct evidence of inter-molecular vibrations by THz spectroscopy," *Chem. Phys. Lett.* **413**, 495–500 (2005).
25. S. Nashima, O. Morikawa, K. Takata, and M. Hangyo, "Temperature dependence of optical and electronic properties of moderately doped silicon at terahertz frequencies," *J. Appl. Phys.* **90**, 837 (2001).

26. D. M. Mittleman, J. Cunningham, M. C. Nuss, and M. Geva, "Noncontact semiconductor wafer characterization with the terahertz Hall effect," *Appl. Phys. Lett.* **71**, 16 (1997).
27. P. A. George, J. Strait, J. Dawlaty, S. Shivaraman, M. Chandrashekhara, F. Rana, and M. G. Spencer, "Ultrafast optical-pump terahertz-probe spectroscopy of the carrier relaxation and recombination dynamics in epitaxial graphene," *Nano Lett.* **8**, 4248–4251 (2008).
28. M. Hoffmann, J. Hebling, H. Hwang, K.-L. Yeh, and K. Nelson, "Impact ionization in InSb probed by terahertz pump—terahertz probe spectroscopy," *Phys. Rev. B* **79**, 161201 (2009).
29. M. Nakajima, N. Takubo, Z. Hiroi, Y. Ueda, and T. Suemoto, "Photoinduced metallic state in VO₂ probed by the terahertz pump-probe spectroscopy," *Appl. Phys. Lett.* **92**, 011907 (2008).
30. T. Phillips and J. Keene, "Submillimeter astronomy," *Proc. IEEE* **80**, 1662–1678 (1992).
31. T. S. Hartwick, D. T. Hodges, D. H. Barker, and F. B. Foote, "Far infrared imagery," *Appl. Opt.* **15**, 1919–22 (1976).
32. P. K. Cheo, "Far-infrared laser system for detection of defects in polyethylene-insulated power cables," *Opt. Lett.* **2**, 42 (1978).
33. A. W. M. Lee, S. Member, B. S. Williams, S. Kumar, Q. Hu, S. Member, and J. L. Reno, "Real-Time Imaging Using a 4 . 3-THz Quantum Cascade Laser and a 320 240 Microbolometer Focal-Plane Array," **18**, 1415–1417 (2006).
34. M. Amanti, G. Scalari, M. Beck, and J. Faist, "Stand-alone system for high-resolution, real-time terahertz imaging," *Opt. Express* **20**, 2772–2778 (2012).
35. U. S. De Cumis, J. Xu, L. Masini, D. Innocenti, P. Pingue, F. Beltram, M. S. Vitiello, P. A. Benedetti, E. Harvey, and D. A. Ritchie, "Terahertz confocal microscopy with a quantum cascade laser source Abstract ;," **20**, 21924–21931 (2012).
36. P. Dean, A. Valavanis, J. Keeley, K. Bertling, Y. Leng Lim, R. Alhathloul, S. Chowdhury, T. Taimre, L. H. Li, D. Indjin, S. J. Wilson, A. D. Rakić, E. H. Linfield, and a. Giles Davies, "Coherent three-dimensional terahertz imaging through self-mixing in a quantum cascade laser," *Appl. Phys. Lett.* **103**, 181112 (2013).
37. A. a Danylov, T. M. Goyette, J. Waldman, M. J. Coulombe, A. J. Gatesman, R. H. Giles, X. Qian, N. Chandrayan, S. Vangala, K. Termkoa, W. D. Goodhue, and W. E. Nixon, "Terahertz inverse synthetic aperture radar (ISAR) imaging with a quantum cascade laser transmitter.," *Opt. Express* **18**, 16264–72 (2010).

38. P. Dean, N. K. Saat, S. P. Khanna, M. Salih, A. Burnett, J. Cunningham, E. H. Linfield, and a G. Davies, "Dual-frequency imaging using an electrically tunable terahertz quantum cascade laser.," *Opt. Express* **17**, 20631–41 (2009).
39. Y. Leng Lim, P. Dean, M. Nikolić, R. Kliese, S. P. Khanna, M. Lachab, A. Valavanis, D. Indjin, Z. Ikonić, P. Harrison, E. H. Linfield, a. Giles Davies, S. J. Wilson, and A. D. Rakić, "Demonstration of a self-mixing displacement sensor based on terahertz quantum cascade lasers," *Appl. Phys. Lett.* **99**, 081108 (2011).
40. F. P. Mezzapesa, L. L. Columbo, M. Brambilla, M. Dabbicco, M. S. Vitiello, and G. Scamarcio, "Imaging of free carriers in semiconductors via optical feedback in terahertz quantum cascade lasers," *Appl. Phys. Lett.* **104**, 041112 (2014).
41. A. J. Huber, F. Keilmann, J. Wittborn, J. Aizpurua, and R. Hillenbrand, "Terahertz near-field nanoscopy of mobile carriers in single semiconductor nanodevices.," *Nano Lett.* **8**, 3766–70 (2008).
42. T. Kürner, "Towards future THz communications systems," *Terahertz Sci. Technol.* **5**, 11–17 (2012).
43. S. Title, W. Communication, T. I. G. Thz, D. Submitted, T. U. Braunschweig, N. Address, G. Voice, and T. Tutorial, <https://mentor.ieee.org/802.15/dcn/12/15-12-0320-01-0thz-What-S-next-Wireless-Communication-beyond-60-Ghz-Tutorial-Ig-Thz.pdf> (n.d.).
44. C. Jastrow, K. Münter, R. Piesiewicz, and T. Kürner, "300 GHz transmission system," *Electron. Lett.* **44**, (2008).
45. L. Möller, J. Federici, A. Sinyukov, and C. Xie, "Data encoding on terahertz signals for communication and sensing," *Opt. Lett.* **33**, 393–395 (2008).
46. T. Liu, G. Lin, Y. Chang, and C. Pan, "Wireless audio and burst communication link with directly modulated THz photoconductive antenna," *Opt. Express* **13**, 10416–10423 (2005).
47. T. Kleine-Ostmann, K. Pierz, G. Hein, P. Dawson, and M. Koch, "Audio signal transmission over THz communication channel using semiconductor modulator," *Electron. Lett.* **40**, 2–3 (2004).
48. P. D. Grant, S. R. Laframboise, R. Dudek, M. Graf, a. Bezinger, and H. C. Liu, "Terahertz free space communications demonstration with quantum cascade laser and quantum well photodetector," *Electron. Lett.* **45**, 952 (2009).
49. F. Palma, F. Teppe, A. El Fatimy, R. Green, J. Xu, Y. Vachontin, A. Tredicucci, G. Goltsman, and W. Knap, "THz communication system based on a THz Quantum Cascade Laser and a Hot Electron Bolometer," 35th Int. Conf. Infrared, Millimeter, Terahertz Waves 1–2 (2010).

50. A. Maestrini, I. Mehdi, J. V. Siles, J. S. Ward, R. Lin, B. Thomas, C. Lee, J. Gill, G. Chattopadhyay, E. Schlecht, J. Pearson, and P. Siegel, "Design and characterization of a room temperature all-solid-state electronic source tunable from 2.48 to 2.75 THz," *IEEE Trans. Terahertz Sci. Technol.* **2**, 177–185 (2012).
51. F. Barath and M. Chavez, "The upper atmosphere research satellite microwave limb sounder instrument," *J. Geophys. Res.* **98**, 10751–10762 (1993).
52. T. Taniuchi, "Widely tunable terahertz-wave generation in an organic crystal and its spectroscopic application," *J. Appl. Phys.* **95**, 5984 (2004).
53. C. Staus, T. Kuech, and L. McCaughan, "Continuously phase-matched terahertz difference frequency generation in an embedded-waveguide structure supporting only fundamental modes," *Opt. Express* **16**, 13296 (2008).
54. K. Saito, T. Tanabe, and Y. Oyama, "THz-wave generation via difference frequency mixing in strained silicon based waveguide utilizing its second order susceptibility $\chi^{(2)}$," *Opt. Express* **22**, 16660–8 (2014).
55. C. W. Berry, M. R. Hashemi, S. Preu, H. Lu, A. C. Gossard, and M. Jarrahi, "Plasmonics enhanced photomixing for generating frequency-tunable terahertz radiation," **39**, 4522–4524 (2014).
56. W.-D. Zhang, J. R. Middendorf, and E. R. Brown, "Demonstration of a GaAs-based 1550-nm continuous wave photomixer," *Appl. Phys. Lett.* **106**, 021119 (2015).
57. H. Tanoto, J. H. Teng, Q. Y. Wu, M. Sun, Z. N. Chen, S. a. Maier, B. Wang, C. C. Chum, G. Y. Si, a. J. Danner, and S. J. Chua, "Nano-antenna in a photoconductive photomixer for highly efficient continuous wave terahertz emission," *Sci. Rep.* **3**, 1–6 (2013).
58. M. Inguscio, G. Moruzzi, K. M. Evenson, and D. A. Jennings, "A review of frequency measurements of optically pumped lasers from 0.1 to 8 THz," *J. Appl. Phys.* **60**, R161 (1986).
59. E. R. Mueller and B. H. Ave, "OPTICALLY-PUMPED THz LASER TECHNOLOGY Eric R. Mueller," www.coherent.com/downloads/opticallypumpedlaser.pdf (n.d.).
60. A. Andronov, I. Zverev, and V. Kozlov, "Stimulated emission in the long-wavelength IR region from hot holes in Ge in crossed electric and magnetic fields," *JETP Lett* **40**, 69–71 (1984).
61. S. Komiyama, N. Iizuka, and Y. Akasaka, "Evidence for induced far-infrared emission from p-Ge in crossed electric and magnetic fields," *Appl. Phys. Lett.* **47**, 958 (1985).
62. Y. L. Ivanov, "Generation of cyclotron radiation by light holes in germanium," *Opt. Quantum Electron.* **23**, S253–S265 (1991).

63. I. Altukhov, M. Kagan, and K. Korolev, "Hot-hole far-IR emission from uniaxially compressed germanium," *Sov. Phys. JETP* **74**, 404–408 (1992).
64. E. Bründermann, D. R. Chamberlin, and E. E. Haller, "High duty cycle and continuous terahertz emission from germanium," *Appl. Phys. Lett.* **76**, 2991 (2000).
65. Y. P. Gousev, I. V. Altukhov, K. A. Korolev, V. P. Sinis, M. S. Kagan, E. E. Haller, M. A. Odnoblyudov, I. N. Yassievich, and K.-A. Chao, "Widely tunable continuous-wave THz laser," *Appl. Phys. Lett.* **75**, 757 (1999).
66. R. Köhler, A. Tredicucci, F. Beltram, H. E. Beere, E. H. Linfield, A. G. Davies, D. A. Ritchie, R. C. Iotti, and F. Rossi, "Terahertz semiconductor-heterostructure laser," *Nature* **417**, 156–159 (2002).
67. R. F. Kazarinov and R. A. Suris, "Possibility of the amplification of electromagnetic waves in a semiconductor with a superlattice," *Sov. Phys. Semicond.* **5**, 707–709 (1971).
68. F. Capasso, K. Mohammed, and A. Cho, "Resonant tunneling through double barriers, perpendicular quantum transport phenomena in superlattices, and their device applications," *Electron. Struct. Semicond. ... QE-22*, 1853–1869 (1988).
69. P. Yuh and K. L. Wang, "Novel infrared band-aligned superlattice laser," *Appl. Phys. Lett.* **51**, 1404 (1987).
70. J. Faist, F. Capasso, D. L. Sivco, C. Sirtori, A. L. Hutchinson, and A. Y. Cho, "Quantum cascade laser," *Science* (80-.). **264**, 553–556 (1994).
71. M. Helm, P. England, E. Colas, F. DeRosa, and S. A. Jr, "Intersubband emission from semiconductor superlattices excited by sequential resonant tunneling," *Phys. Rev. Lett.* **63**, 1–4 (1989).
72. S. Fatholouloumi, E. Dupont, C. W. I. Chan, Z. R. Wasilewski, C. Jirauschek, Q. Hu, and D. Ban, "Terahertz quantum cascade lasers operating up to ~ 200 K with optimized oscillator strength and improved injection tunneling," *Opt. Express* **20**, 3331–3339 (2012).
73. B. Williams, S. Kumar, Q. Hu, and J. Reno, "High-power terahertz quantum-cascade lasers," *Electron. Lett.* **42**, 18–19 (2006).
74. S. Kumar, B. S. Williams, Q. Qin, A. W. Lee, Q. Hu, and J. L. Reno, "Surface-emitting distributed feedback terahertz quantum-cascade lasers in metal-metal waveguides," *Opt. Express* **15**, 113–128 (2007).
75. Y. Chassagneux, R. Colombelli, W. Maineult, S. Barbieri, H. E. Beere, D. A. Ritchie, S. P. Khanna, E. H. Linfield, and A. G. Davies, "Electrically pumped photonic-crystal terahertz lasers controlled by boundary conditions," *Nature* **457**, 174–8 (2009).

76. L. Mahler, A. Tredicucci, F. Beltram, C. Walther, J. Faist, B. Witzigmann, H. E. Beere, and D. A. Ritchie, "Vertically emitting microdisk lasers," *Nat. Photonics* **3**, 46–49 (2008).
77. N. Yu, Q. J. Wang, and F. Capasso, "Beam engineering of quantum cascade lasers," *Laser Photon. Rev.* **6**, 24–46 (2012).
78. A. W. M. Lee, B. S. Williams, S. Kumar, Q. Hu, and J. L. Reno, "Tunable terahertz quantum cascade lasers with external gratings," *Opt. Lett.* **35**, 910–2 (2010).
79. L. Mahler, A. Tredicucci, F. Beltram, H. E. Beere, and D. A. Ritchie, "Tuning a distributed feedback laser with a coupled microcavity," *Opt. Express* **18**, 19185–91 (2010).
80. Q. Qin, B. Williams, S. Kumar, J. Reno, and Q. Hu, "Tuning a terahertz wire laser," *Nat. Photonics* **3**, 732–737 (2009).
81. Q. Qin, J. L. Reno, and Q. Hu, "MEMS-based tunable terahertz wire-laser over 330 GHz," *Opt. Lett.* **36**, 692–4 (2011).
82. P. Khosropanah, a Baryshev, W. Zhang, W. Jellema, J. N. Hovenier, J. R. Gao, T. M. Klapwijk, D. G. Pavelev, B. S. Williams, S. Kumar, Q. Hu, J. L. Reno, B. Klein, and J. L. Hesler, "Phase locking of a 2.7 THz quantum cascade laser to a microwave reference," *Opt. Lett.* **34**, 2958–60 (2009).
83. A. L. Betz, R. T. Boreiko, B. S. Williams, S. Kumar, Q. Hu, and J. L. Reno, "Frequency and phase-lock control of a 3 THz quantum cascade laser," *Opt. Lett.* **30**, 1837–9 (2005).
84. D. J. Hayton, a. Khudchenko, D. G. Pavelyev, J. N. Hovenier, a. Baryshev, J. R. Gao, T. Y. Kao, Q. Hu, J. L. Reno, and V. Vaks, "Phase locking of a 3.4 THz third-order distributed feedback quantum cascade laser using a room-temperature superlattice harmonic mixer," *Appl. Phys. Lett.* **103**, 051115 (2013).
85. S. Barbieri, M. Ravaro, and P. Gellie, "Coherent sampling of active mode-locked terahertz quantum cascade lasers and frequency synthesis," *Nat. Photonics* **5**, 306–313 (2011).
86. J. R. Freeman, J. Maysonnave, N. Jukam, P. Cavalié, K. Maussang, H. E. Beere, D. a. Ritchie, J. Mangeney, S. S. Dhillon, and J. Tignon, "Direct intensity sampling of a modelocked terahertz quantum cascade laser," *Appl. Phys. Lett.* **101**, 181115 (2012).
87. D. Burghoff, T.-Y. Kao, N. Han, C. W. I. Chan, X. Cai, Y. Yang, D. J. Hayton, J.-R. Gao, J. L. Reno, and Q. Hu, "Terahertz laser frequency combs," *Nat. Photonics* **8**, 462–467 (2014).

88. M. a. Belkin, F. Capasso, A. Belyanin, D. L. Sivco, A. Y. Cho, D. C. Oakley, C. J. Vineis, and G. W. Turner, "Terahertz quantum-cascade-laser source based on intracavity difference-frequency generation," *Nat. Photonics* **1**, 288–292 (2007).
89. S. Jung, Y. Jiang, K. Vijayraghavan, A. Jiang, F. Demmerle, G. Bohm, X. Wang, M. Troccoli, M. C. Amann, and M. Belkin, "Recent Progress in Widely Tunable Single Mode Room Temperature Terahertz Quantum Cascade Laser Sources," *IEEE J. Sel. Top. Quantum Electron.* **N.A.**, 1–10 (2015).
90. C. Sirtori, S. Barbieri, and R. Colombelli, "Wave engineering with THz quantum cascade lasers," *Nat. Photonics* **7**, 691–701 (2013).
91. P. Harrison, *Quantum Wells, Wires and Dots: Theoretical and Computational Physics of Semiconductor Nanostructures* (Wiley, 2009).
92. M. G. Burt, "On the validity and range of applicability of the particle in a box model," *Appl. Phys. Lett.* **65**, 717 (1994).
93. A. Yariv, *Quantum Electronics*, 3rd ed. (John Wiley & Sons, 1989).
94. B. S. Williams, "Terahertz quantum cascade lasers (Ph. D dissertation)," Massachusetts institute of technology (2003).
95. J. H. Smet, C. G. Fonstad, and Q. Hu, "Intrawell and interwell intersubband transitions in multiple quantum wells for far-infrared sources," *J. Appl. Phys.* **79**, 9305 (1996).
96. P. Hyldgaard and J. Wilkins, "Electron-electron scattering in far-infrared quantum cascade lasers," *Phys. Rev. B* **53**, 6889–6892 (1996).
97. C. Gmachl, "Recent progress in quantum cascade lasers and applications," *Reports Prog. Phys.* **64**, 1533–1601 (2001).
98. M. I. Amanti, G. Scalari, R. Terazzi, M. Fischer, M. Beck, J. Faist, A. Rudra, P. Gallo, and E. Kapon, "Bound-to-continuum terahertz quantum cascade laser with a single-quantum-well phonon extraction/injection stage," *New J. Phys.* **11**, 125022 (2009).
99. G. Scalari, L. Ajili, J. Faist, H. Beere, E. Linfield, D. Ritchie, and G. Davies, "Far-infrared ($\lambda \approx 87 \mu\text{m}$) bound-to-continuum quantum-cascade lasers operating up to 90 K," *Appl. Phys. Lett.* **82**, 3165 (2003).
100. S. Kumar, Q. Hu, and J. L. Reno, "186 K operation of terahertz quantum-cascade lasers based on a diagonal design," *Appl. Phys. Lett.* **94**, 131105 (2009).
101. M. Rösch, G. Scalari, M. Beck, and J. Faist, "Octave-spanning semiconductor laser," *Nat. Photonics* **9**, (2014).

102. S. Kumar, C. W. I. Chan, Q. Hu, and J. L. Reno, "A 1.8-THz quantum cascade laser operating significantly above the temperature of $[k_B]$," *Nat. Phys.* **7**, 166–171 (2010).
103. S. Kumar, "Development of Terahertz Quantum-Cascade Lasers (Ph. D dissertation)," Massachusetts institute of technology (2007).
104. A. Wei Min Lee, Q. Qin, S. Kumar, B. S. Williams, Q. Hu, and J. L. Reno, "High-power and high-temperature THz quantum-cascade lasers based on lens-coupled metal-metal waveguides.," *Opt. Lett.* **32**, 2840–2842 (2007).
105. M. Amanti, M. Fischer, C. Walther, G. Scalari, and J. Faist, "Horn antennas for terahertz quantum cascade lasers," *Electron. Lett.* **43**, 125–126 (2007).
106. W. Maineult, P. Gellie, A. Andronico, P. Filloux, G. Leo, C. Sirtori, S. Barbieri, E. Peytavit, T. Akalin, J.-F. Lampin, H. E. Beere, and D. A. Ritchie, "Metal-metal terahertz quantum cascade laser with micro-transverse-electromagnetic-horn antenna," *Appl. Phys. Lett.* **93**, 183508 (2008).
107. N. Yu, Q. J. Wang, M. A. Kats, J. A. Fan, S. P. Khanna, L. Li, A. G. Davies, E. H. Linfield, and F. Capasso, "Designer spoof surface plasmon structures collimate terahertz laser beams.," *Nat. Mater.* **9**, 730–735 (2010).
108. M. I. Amanti, M. Fischer, G. Scalari, M. Beck, and J. Faist, "Low-divergence single-mode terahertz quantum cascade laser," *Nat. Photonics* **3**, 586–590 (2009).
109. J. A. Fan, M. A. Belkin, F. Capasso, S. Khanna, M. Lachab, A. G. Davies, and E. H. Linfield, "Surface emitting terahertz quantum cascade laser with a double-metal waveguide.," *Opt. Express* **14**, 11672–80 (2006).
110. G. Xu, R. Colombelli, P. Khanna, A. Belarouci, X. Letartre, L. Li, E. H. Linfield, A. G. Davies, H. E. Beere, and D. A. Ritchie, "Efficient power extraction in surface-emitting semiconductor lasers using graded photonic heterostructures," *Nat. Commun.* **3**, 952–957 (2012).
111. L. Mahler, M. I. Amanti, C. Walther, A. Tredicucci, F. Beltram, J. Faist, H. E. Beere, and D. A. Ritchie, "Distributed feedback ring resonators for vertically emitting terahertz quantum cascade lasers.," *Opt. Express* **17**, 13031–13039 (2009).
112. E. Mujagić, C. Deutsch, H. Detz, P. Klang, M. Nobile, A. M. Andrews, W. Schrenk, K. Unterrainer, and G. Strasser, "Vertically emitting terahertz quantum cascade ring lasers," *Appl. Phys. Lett.* **95**, 011120 (2009).
113. Y. Chassagneux, R. Colombelli, W. Maineult, S. Barbieri, H. E. Beere, D. A. Ritchie, S. P. Khanna, E. H. Linfield, and A. G. Davies, "Electrically pumped photonic-crystal terahertz lasers controlled by boundary conditions.," *Nature* **457**, 174–8 (2009).

114. Y. Chassagneux, R. Colombelli, W. Maineults, S. Barbieri, S. P. Khanna, E. H. Linfield, and A. G. Davies, "Predictable surface emission patterns in terahertz photonic-crystal quantum cascade lasers.," *Opt. Express* **17**, 9491–9502 (2009).
115. Y. Chassagneux, R. Colombelli, W. Maineult, S. Barbieri, S. P. Khanna, E. H. Linfield, and A. G. Davies, "Graded photonic crystal terahertz quantum cascade lasers," *Appl. Phys. Lett.* **96**, 031104 (2010).
116. G. Liang, H. Liang, Y. Zhang, S. P. Khanna, L. Li, A. Giles Davies, E. Linfield, D. Fatt Lim, C. Seng Tan, S. Fung Yu, H. Chun Liu, and Q. J. Wang, "Single-mode surface-emitting concentric-circular-grating terahertz quantum cascade lasers," *Appl. Phys. Lett.* **102**, 031119 (2013).
117. G. Liang, H. Liang, Y. Zhang, L. Li, a. G. Davies, E. Linfield, S. F. Yu, H. C. Liu, and Q. J. Wang, "Low divergence single-mode surface-emitting concentric-circular-grating terahertz quantum cascade lasers," *Opt. Express* **21**, 31872 (2013).
118. T.-Y. Kao, Q. Hu, and J. L. Reno, "Phase-locked arrays of surface-emitting terahertz quantum-cascade lasers," *Appl. Phys. Lett.* **96**, 101106 (2010).
119. Y. Chassagneux, R. Colombelli, W. Maineult, S. Barbieri, H. E. Beere, D. A. Ritchie, S. P. Khanna, E. H. Linfield, and A. G. Davies, "Electrically pumped photonic-crystal terahertz lasers controlled by boundary conditions.," *Nature* **457**, 174–8 (2009).
120. J. Scheuer and A. Yariv, "Annular Bragg defect mode resonators," *J. Opt. Soc. Am. B* **20**, 2285–2291 (2003).
121. S.-H. Kim, S.-K. Kim, and Y.-H. Lee, "Vertical beaming of wavelength-scale photonic crystal resonators," *Phys. Rev. B* **73**, 235117 (2006).
122. N. Yu, L. Diehl, E. Cubukcu, and D. Bour, "Coherent coupling of multiple transverse modes in quantum cascade lasers," *Phys. Rev. Lett.* **013901**, 1–4 (2009).
123. A. K. Wójcik, N. Yu, F. Capasso, and A. Belyanin, "Nonlinear optical interactions of laser modes in quantum cascade lasers," *J. Mod. Opt.* **58**, 727–742 (2011).
124. W. Bewley and J. Lindle, "Beam steering in high-power CW quantum-cascade lasers," *IEEE J. Quantum Electron.* **41**, 833–841 (2005).
125. T. W. Ebbesen, C. Genet, and S. I. Bozhevolnyi, "Surface-plasmon circuitry," *Phys. Today* **61**, 44 (2008).
126. B. Ng, J. Wu, S. M. Hanham, A. I. Fernández-Domínguez, N. Klein, Y. F. Liew, M. B. H. Breese, M. Hong, and S. a. Maier, "Spoof Plasmon Surfaces: A Novel Platform for THz Sensing," *Adv. Opt. Mater.* **1**, 543–548 (2013).

127. X. Luo and T. Ishihara, "Surface plasmon resonant interference nanolithography technique," *Appl. Phys. Lett.* **84**, 4780 (2004).
128. L. Li, T. Li, S. Wang, C. Zhang, and S. Zhu, "Plasmonic Airy Beam Generated by In-Plane Diffraction," *Phys. Rev. Lett.* **107**, 1–4 (2011).
129. H. J. Lezec, a Degiron, E. Devaux, R. a Linke, L. Martin-Moreno, F. J. Garcia-Vidal, and T. W. Ebbesen, "Beaming light from a subwavelength aperture.," *Science* **297**, 820–2 (2002).
130. E. Cubukcu, E. A. Kort, K. B. Crozier, and F. Capasso, "Plasmonic laser antenna," *Appl. Phys. Lett.* **89**, 093120 (2006).
131. B. Guo, G. Song, and L. Chen, "Plasmonic very-small-aperture lasers," *Appl. Phys. Lett.* **91**, 021103 (2007).
132. N. Yu, J. Fan, Q. J. Wang, C. Pflügl, L. Diehl, T. Edamura, M. Yamanishi, H. Kan, and F. Capasso, "Small-divergence semiconductor lasers by plasmonic collimation," *Nat. Photonics* **2**, 564–570 (2008).
133. J.-P. Tetienne, R. Blanchard, N. Yu, P. Genevet, M. a Kats, J. a Fan, T. Edamura, S. Furuta, M. Yamanishi, and F. Capasso, "Dipolar modeling and experimental demonstration of multi-beam plasmonic collimators," *New J. Phys.* **13**, 053057 (2011).
134. N. Yu, M. Kats, C. Pflügl, M. Geiser, Q. J. Wang, M. a. Belkin, F. Capasso, M. Fischer, A. Wittmann, J. Faist, T. Edamura, S. Furuta, M. Yamanishi, and H. Kan, "Multi-beam multi-wavelength semiconductor lasers," *Appl. Phys. Lett.* **95**, 161108 (2009).
135. N. Yu, Q. J. Wang, C. Pflügl, L. Diehl, F. Capasso, T. Edamura, S. Furuta, M. Yamanishi, and H. Kan, "Semiconductor lasers with integrated plasmonic polarizers," *Appl. Phys. Lett.* **94**, 151101 (2009).
136. A. Babuty, A. Bousseksou, J.-P. Tetienne, I. M. Doyen, C. Sirtori, G. Beaudoin, I. Sagnes, Y. De Wilde, and R. Colombelli, "Semiconductor Surface Plasmon Sources," *Phys. Rev. Lett.* **104**, 226806 (2010).
137. B. Schwarz, P. Reininger, D. Ristanić, H. Detz, A. M. Andrews, W. Schrenk, and G. Strasser, "Monolithically integrated mid-infrared lab-on-a-chip using plasmonics and quantum cascade structures.," *Nat. Commun.* **5**, 4085 (2014).
138. J. Pendry, A. Holden, W. Stewart, and I. Youngs, "Extremely low frequency plasmons in metallic mesostructures.," *Phys. Rev. Lett.* **76**, 4773–4776 (1996).
139. C. R. Williams, S. R. Andrews, S. a. Maier, a. I. Fernández-Domínguez, L. Martín-Moreno, and F. J. García-Vidal, "Highly confined guiding of terahertz surface plasmon polaritons on structured metal surfaces," *Nat. Photonics* **2**, 175–179 (2008).

140. M. Navarro-Cía, M. Beruete, S. Agrafiotis, F. Falcone, M. Sorolla, and S. a Maier, "Broadband spoof plasmons and subwavelength electromagnetic energy confinement on ultrathin metafilms.," *Opt. Express* **17**, 18184–95 (2009).
141. F. J. Garcia-Vidal, L. Martín-Moreno, and J. B. Pendry, "Surfaces with holes in them: new plasmonic metamaterials," *J. Opt. A Pure Appl. Opt.* **7**, S97–S101 (2005).
142. M. Wanke, E. Young, and C. Nordquist, "Monolithically integrated solid-state terahertz transceivers," *Nat. Photonics* **4**, 565–569 (2010).
143. H.-W. Hübers, "Terahertz technology: Towards THz integrated photonics," *Nat. Photonics* **4**, 503–504 (2010).
144. B. Y. Zhang, T. Liu, B. Meng, X. Li, G. Liang, X. Hu, and Q. J. Wang, "Broadband high photoresponse from pure monolayer graphene photodetector.," *Nat. Commun.* **4**, 1811 (2013).
145. S. Fatholouloumi, E. Dupont, Z. R. Wasilewski, C. W. I. Chan, S. G. Razavipour, S. R. Laframboise, S. Huang, Q. Hu, D. Ban, and H. C. Liu, "Effect of oscillator strength and intermediate resonance on the performance of resonant phonon-based terahertz quantum cascade lasers," *J. Appl. Phys.* **113**, 113109 (2013).
146. Y. Li, J. Wang, N. Yang, J. Liu, and T. Wang, "The output power and beam divergence behaviors of tapered terahertz quantum cascade lasers," *Opt. Express* **21**, 15998–16006 (2013).
147. Y. Chassagneux, R. Colombelli, W. Maineult, S. Barbieri, H. E. Beere, D. A. Ritchie, S. P. Khanna, E. H. Linfield, and A. G. Davies, "Electrically pumped photonic-crystal terahertz lasers controlled by boundary conditions.," *Nature* **457**, 174–178 (2009).
148. X. Cai, A. B. Sushkov, R. J. Suess, M. M. Jadidi, G. S. Jenkins, L. O. Nyakiti, R. L. Myers-Ward, S. Li, J. Yan, D. K. Gaskill, T. E. Murphy, H. D. Drew, and M. S. Fuhrer, "Sensitive room-temperature terahertz detection via the photothermoelectric effect in graphene," *Nat. Nanotechnol.* 1–6 (2014).
149. S.-L. Chen, Y.-C. Chang, C. Zhang, J. G. Ok, T. Ling, M. T. Mihnev, T. B. Norris, and L. J. Guo, "Efficient real-time detection of terahertz pulse radiation based on photoacoustic conversion by carbon nanotube nanocomposite," *Nat. Photonics* **8**, 537–542 (2014).
150. T. Kleine-Ostmann, P. Dawson, K. Pierz, G. Hein, and M. Koch, "Room-temperature operation of an electrically driven terahertz modulator," *Appl. Phys. Lett.* **84**, 3555 (2004).
151. D. Shrekenhamer, S. Rout, A. C. Strikwerda, C. Bingham, R. D. Averitt, S. Sonkusale, and W. J. Padilla, "High speed terahertz modulation from metamaterials with embedded high electron mobility transistors.," *Opt. Express* **19**, 9968–9975 (2011).

152. B. Sensale-Rodriguez, R. Yan, S. Rafique, M. Zhu, W. Li, X. Liang, D. Gundlach, V. Protasenko, M. M. Kelly, D. Jena, L. Liu, and H. G. Xing, "Extraordinary control of terahertz beam reflectance in graphene electro-absorption modulators.," *Nano Lett.* **12**, 4518–22 (2012).
153. W. Gao, J. Shu, K. Reichel, D. V Nickel, X. He, G. Shi, R. Vajtai, P. M. Ajayan, J. Kono, D. M. Mittleman, and Q. Xu, "High-Contrast Terahertz Wave Modulation by Gated Graphene Enhanced by Extraordinary Transmission through Ring Apertures.," *Nano Lett.* (2014).
154. L. Liao, J. Bai, Y. Qu, Y. Lin, Y. Li, Y. Huang, and X. Duan, "High-kappa oxide nanoribbons as gate dielectrics for high mobility top-gated graphene transistors.," *Proc. Natl. Acad. Sci. U. S. A.* **107**, 6711–5 (2010).
155. L. Ren, Q. Zhang, J. Yao, Z. Sun, R. Kaneko, Z. Yan, S. Nanot, Z. Jin, I. Kawayama, M. Tonouchi, J. M. Tour, and J. Kono, "Terahertz and infrared spectroscopy of gated large-area graphene.," *Nano Lett.* **12**, 3711–5 (2012).
156. S. Adam, E. H. Hwang, V. M. Galitski, and S. Das Sarma, "A self-consistent theory for graphene transport.," *Proc. Natl. Acad. Sci. U. S. A.* **104**, 18392–7 (2007).
157. D. K. Efetov and P. Kim, "Controlling Electron-Phonon Interactions in Graphene at Ultrahigh Carrier Densities," *Phys. Rev. Lett.* **105**, 256805 (2010).
158. A. Hsu, H. Wang, K. K. Kim, J. Kong, and T. Palacios, "Impact of Graphene Interface Quality on Contact Resistance and RF Device Performance," *IEEE Electron Device Lett.* **32**, 1008–1010 (2011).
159. W. Leong, H. Gong, and J. Thong, "Low-Contact-Resistance Graphene Devices with Nickel-Etched-Graphene Contacts," *ACS Nano* **8**, 994–1001 (2013).
160. J. Smith, A. Franklin, D. Farmer, and C. Dimitrakopoulos, "Reducing contact resistance in graphene devices through contact area patterning," *ACS Nano* **7**, 3661–3667 (2013).
161. S. Kim, J. Nah, I. Jo, D. Shahrjerdi, L. Colombo, Z. Yao, E. Tutuc, and S. K. Banerjee, "Realization of a high mobility dual-gated graphene field-effect transistor with Al₂O₃ dielectric," *Appl. Phys. Lett.* **94**, 062107 (2009).
162. F. Valmorra, G. Scalari, C. Maissen, W. Fu, C. Scho, J. W. Choi, H. G. Park, and M. Beck, "Low-Bias Active Control of Terahertz Waves by Coupling Large-Area," (2013).
163. D. Frickey, "Conversions between S, Z, Y, H, ABCD, and T parameters which are valid for complex source and load impedances," *IEEE Trans. Microw. Theory Tech.* **42**, 205–211 (1994).