Terahertz quantum cascade lasing in disordered photonic systems

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**NANYANG
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**Terahertz quantum cascade lasing in disordered
photonic systems**

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SCHOOL OF ELECTRICAL & ELECTRONIC ENGINEERING

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**A thesis submitted to the Nanyang Technological University
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Abstract

Scattering, which is created by dust, clouds, white paints or imperfections of artificial material such as photonic crystal, is normally undesirable in the field of photonics. Unequivocally, a deep understanding of disorder can help to overcome the issues arising from scattering. On the other hand, it is also possible to utilize the disorder to create useful photonic devices. A most well-known example is the random laser, in which lasing action is sustained by multiple scattering of light within the disordered media. Disordered structures are also proposed for the applications including wavelength filters, optical switches, sensors and cavity quantum electrodynamics. Most of the research work on disordered structures has been focused on short wavelength range. Several years ago, this tide spreads to mid-infrared range with the experimental demonstration and theoretical optimization of mid-infrared random laser at $\sim 10\ \mu\text{m}$. However, in the technically important terahertz (THz) frequency range, the disordered photonics are least explored. This thesis investigates disordered photonic systems based on the double-metal (DM) waveguide of THz quantum cascade laser (QCL) and explores the emission properties with the assistance of gain.

As the first demonstration, I design and fabricate one-dimensional disordered grating structures on top of the DM waveguide of the THz QCLs. Disorder is introduced into the waveguide by dislocating the position of each aperture from a periodic configuration. Numerical calculation indicates that the introduced disorder creates multiple microcavities with small optical losses near the bandgap frequency window of the underlying long-range order. With electrical pumping, 8 emission peaks at around 3.2 THz are observed, which are located near the bandgap region of the periodic counterparts. Simulation work and experimental measurements provide strong evidence for spatial localization of light.

Instead of utilizing long-range order for creating microcavities, I also investigate two-dimensional disordered structures consisting of randomly-distributed pillar scatterers fabricated from a quantum cascade gain medium, in which long-range order does not

exist. Due to the strong scattering efficiency of a high-index dielectric pillar for transverse-magnetic (TM) polarized wave, it is shown that such structures can achieve multimode lasing, with strongly localized modes at THz frequencies. This is a typical feature of random lasers. The weak short-range order induced by the pillar distribution is sufficient to ensure high quality-factor modes that have a large overlap with the active material. It is also found that the emission spectrum can be easily tuned by tailoring the size and filling fraction of the scatterers.

While the dielectric pillar scatterer is beneficial for better performance of multimode lasing (lower threshold, more lasing modes etc.) due to high scattering efficiency, it also limits the lasing to the Mie resonance frequency range, making the broadband random lasing applications difficult. The non-resonant scatterers feature a weaker scattering strength nonetheless a relatively frequency-insensitive scattering efficiency. Inspired by this, I further develop metallic pillars, a class of non-resonant scatterers with high scattering efficiency in the THz frequency range, enabling possible a broadband random laser. It is demonstrated that such random lasers, with metallic pillars embedded in THz QCL gain medium, outperform their dielectric-counterparts in terms of lasing spectral range, the number of lasing mode and laser threshold.

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Glossary of Acronyms

1-D	one-dimensional
2-D	two-dimensional
3-D	three-dimensional
BCB	benzocyclobutene
CW	continuous-wave
DM	double-metal
FDTD	finite-difference time-domain
FEM	finite-element method
FF	filling fraction
ICP	inductively coupled plasma
LIV	light-current-voltage
mid-IR	mid-infrared
PBG	photonic bandgap
Q-factor	quality factor
QCL	quantum cascade laser
RT	room temperature
SEM	scanning electron micrograph
SPR	surface plasmon resonance
TE	transverse-electric
THz	terahertz
TM	transverse-magnetic

1 Background and Motivation

In this chapter, the background of the research will be presented with a brief introduction of lasing in disordered medium. The motivation will then be clarified. Finally, the organization of this thesis will be outlined.

1.1 Background

The scattering of light is a form of scattering process during which the light ray is forced to deviate from its original trajectory due to the inhomogeneity of medium [1]. The inhomogeneous medium can be particles, tiny dopants inside a medium, roughness on the medium surfaces, or the interfaces between two media. According to this definition, the specular reflection and refraction at optically smooth interfaces are essentially the special cases of light scattering. Light scattering is one of the most common phenomena in our daily life. For example, the blue color of the sky is formed via the scattering of light by small particles in air at preferred short wavelengths; a colorful rainbow appears through the sunlight scattering by rain drops; people can see the sunlight/laser ray passing through the air or colloidal suspensions because the scattered light by dust or particles enters their eyes; The iridescent colors can be seen from the bird feather or butterfly wings [2] and can also be found with light illuminating the surface of man-made products, like CDs or DVDs, at different angles, which is all because of the angular scattering of the light at a certain wavelength by the ordered or disordered micro-structures.

Figure 1-1. The light scattering phenomena in our daily life.

In the field of photonics, light scattering phenomena are widely utilized for various applications, especially in photonic crystals. For instance, periodically-arranged gratings are utilized to differentiate wavelengths for spectroscopy analysis; structures with periodically modulated index profiles are created for photonic crystal lasers/waveguides; multiple-layered structures are employed for wavelength filters. However, light scattering due to unexpected material heterogeneities, as well as the structural disorder induced by fabrication error is indeed detrimental to many photonic devices, such as glass lenses, metallic mirrors, gratings, waveguides, micro-cavities and lasers, and is therefore undesirable. Significant theoretical efforts have been made to analyze the influences of surface roughness or structural disorder on the optical performance [3–7] and potential strategies to eliminate or overcome the influences have been proposed [8,9]. However, overcoming the unwanted random fabrication variations essentially requires stringent fabrication techniques and man-made disorder to some extent is inevitable. In addition, in some scenarios, the influence of disorder cannot be avoided: an example talks about the great challenge imposed by disordered cells/composites in human tissues and bones while performing the human body imaging [10,11].

Figure 1-2. Various applications of disordered structures. (a), (b), (c) A disordered thin film utilized for broadband and enhanced light capturing. (d), (e), (f) Disordered photonic crystal membrane waveguide used for cavity quantum electrodynamics researches. (g), (h), (i) A chip-based spectrometer based on multiple scattering in a disordered photonic structure. (Figure (a)-(c) are reprinted from [12], figure (e)-(f) are reprinted from [13], and figure (g)-(i) are reprinted from [14])

Rather than always following the ‘order’ rule, one might as well start from the presence of disorder and try to make use of its advantages. Perceiving the disorder as strength rather a weakness has inspired many revolutionary technologies. For example, two-dimensional disordered photonic structures have been demonstrated for efficient light trapping due to coherent optical effects and broadband/wide-angle properties induced by disorder [12]. This can be used for high-efficiency thin-film photovoltaic devices. Disordered photonic structures can also be used as sensitive compact spectrometer because multiple scattering of light in the medium helps to generate speckle patterns that vary delicately with wavelengths [14]. As being proposed to support the strong confinement of electrons under certain circumstance by P. W. Anderson [15], disordered electronic system has attracted enormous research interests to find the metal-insulator transitions. Likewise, the disordered photonic medium has also been extensively investigated for the analogous localization of light due to its attractive physical properties and promising application potentials. The strong localization of light caused by structural disorder/randomness has been demonstrated with a disordered photonic crystal membrane, which proves to be a desirable platform for the research on cavity quantum electrodynamics [13]. In addition, a kind of novel laser sources utilizing disordered photonic medium for feedback and optical amplification has also been demonstrated and proposed for various important applications.

In 1994, Lawandy *et al.*, demonstrated that the laser action can be achieved in a strongly scattering medium consisting of randomly distributed particles and a dye solution which provides light amplification [16]. The feedback of such a laser is provided by the multiple scattering of light in the disordered medium, without well-aligned mirror cavity as in traditional laser systems. Lasing takes place when the gain experienced by one or more of the complex modes overcomes the losses. Because of this property, this kind of lasers was then termed as “random laser” or “mirror-less laser”.

Since its first demonstration, random laser has attracted tremendous research interests due to its specific advantages including small sizes, simple composition, easy fabrication, low cost, broad-spectral/angular emission, flexibility, stretchability, and roll to roll processing, which bring new opportunities for the lighting, imaging, sensing, security, and medical diagnostics [17]. Intense research interests into random lasers also include the random lasing physics. Various theories, e.g. random matrix theory, semi-classical quantum mechanics, steady-state ab initio laser theory, have been proposed to study random lasing properties, including feedback mechanism, modal property, and multimode interaction [18]. Efforts have also been made to tailor the random laser emission in the spatial, spectral and temporal domains. Additionally, it is always appealing to look into the Anderson localization regime of light with the existence of gain materials, where the photon confinement and the strong interaction between the light and gain medium occur.

1.2 Motivation

Lasing in disordered medium has been demonstrated in various systems. Due to the easy accessibility of gain medium, including various dyes, semiconductor materials with large bandgap, and rare-earth doped ceramics, the random emission frequency mainly covers a range from the near-ultraviolet to the near-infrared (IR) [19]. Recently, a mid-IR random laser has been realized by drilling randomly distributed air holes onto a 10 μm quantum cascade laser (QCL) wafer [20]. However, before the commencement of this research, random laser has not been demonstrated in the technically important terahertz (THz) frequency regime yet.

THz frequency range (0.3 THz – 10 THz) is an important frequency span with great potential for diverse applications ranging from non-destructive imaging, spectroscopic sensing to ultra-high speed wireless communication. Numerous technological breakthroughs in the THz field have been achieved in the past two decades. Notable achievements include the invention and advancement of high-performance THz emitters, sensitive THz photodetectors, and THz modulators. THz QCL has emerged as

a compact semiconductor source of coherent THz radiation. Since its first demonstration in 2002 [21], the field of THz QCL has achieved tremendous progress including various practical applications, e.g. spectroscopy, imaging and communications. But these are all relying on conventional lasers. Realizing random laser based on THz QCLs is innovative, and it offers the opportunities of achieving novel potential applications, such as speckle-free imaging, multi-wavelength spectroscopy and multiple channel communication.

1.3 Objective

The main objective of this thesis is to investigate disordered photonic systems based on THz QCL and explore the optical and emission properties with the assistance of gain. To achieve the objective, it is necessary to come up with the strategies to integrate disordered photonic structures onto the THz QCL wafers which support operation wavelength much longer than the critical dimension of active region. In addition, it is important to design scattering medium which could support good confinement of photons, as the lasing in disordered structure always has larger laser threshold than conventional lasers due to the special feedback mechanism. Performances, including threshold, spectral coverage, modal confinement, are to be optimized, which is of great significance for various novel applications.

1.4 Thesis outline

This thesis consists of seven chapters. **Chapter 2** reviews the working principles of random laser with a brief introduction of recent development and application of random lasers. **Chapter 3** scrutinizes the working principles of THz QCLs, followed by a brief introduction of the THz QCL wafer design used in this thesis. Double-metal (DM) waveguide scheme is analyzed and conventional ridge lasers based on DM waveguide have been fabricated and characterized to have a better understanding of the wafer. **Chapter 4** reports the first trial to realize the localization of light by patterning one-dimensional disordered gratings on top of the DM waveguide of THz QCLs. With the

introduction of moderate positional disorder of the gratings, multiple microcavities near the bandgap frequency window of the underlying long-range order are created. Multiple emission peaks are observed and simulations and experimental measurements provide solid evidence for spatial localization of THz wave. **Chapter 5** presents the multimode random lasing of THz QCLs utilizing two-dimensional (2-D) amorphous lattices with high-efficiency dielectric pillar scatterers embedded in low-index benzocyclobutene (BCB). Due to the high scattering efficiency of dielectric pillars and the introduced weak short-range order during the structure generation, multiple high-quality (Q) modes with strongly localized field profiles can be supported. The detailed design, fabrication and experimental characterization will be presented. In **chapter 6**, a novel scatterer — metallic pillar, has been proposed for broadband random lasing application. For transverse-magnetic (TM) polarized light, the metallic pillar is a non-resonant scatterer, whereas it maintains a superior scattering efficiency than that of a resonant dielectric pillar over a broad frequency range. This makes a broadband random laser possible. 2-D random THz QCLs with metallic pillars have been fabricated and characterized. The lasing threshold is low and more than 25 emission peaks are observed across the gain spectral range of the laser. Random lasing in broader spectral range with more lasing peaks could be achieved if a THz QCL wafer with broader gain spectral range was used. Finally, **chapter 7** concludes the whole thesis and proposes future work.

2 The Working Principles and Development of Random Laser

In this chapter, the elastic optical scattering processes, scattering medium categorization and Anderson localization of light in the passive disordered medium will be reviewed, respectively. Two prevailing feedback mechanisms of random laser corresponding to different scattering regimes will then be introduced. Finally, the development and applications of random laser are reviewed.

2.1 Introduction

The gain medium and optical cavity are two basic elements of a conventional laser. The gain medium serves as a photon reservoir, sustaining the stimulated emission for optical amplification. And the optical cavity selectively provides coherent optical feedback for the stimulated emission at certain resonant wavelengths, determining the optical modes of the laser. Therefore, the scattering of photons, by inhomogeneous or roughness in the cavity is usually thought to be detrimental to laser action, as it randomly deflects the photons from their original paths, reducing the optical feedback and, thus, the gain. However, in 1994, Lawandy *et al.*, demonstrated that the laser radiation can occur in a strongly scattering medium consisting of randomly distributed particles and a dye solution which provides light amplification [16]. This kind of lasers was then termed as “random laser”. Since then, intense research interests have been devoted to lasing phenomena in disordered medium. Unlike a conventional laser, such a laser traps light via multiple scattering and interference in a disordered system, without any mirror. Therefore, optical scattering plays a positive role in this case. Before stepping into the random laser regime, a brief theoretical review of optical scattering in disordered medium without and with gain is necessary.

2.2 The theory of optical scattering in passive disordered medium

In this thesis, the scope of discussion is limited to the elastic scattering processes, where the energy of photon is conserved and only the direction can be altered. Typical examples of these are Rayleigh scattering and Mie scattering. Another category of optical scattering is inelastic scattering, where both the photon energy and propagation direction are changed. Raman scattering and Brillouin scattering are typical inelastic scatterings, which will not be considered here.

2.2.1 Single particle scattering

The simplest scattering is the interaction of light with a single particle embedded in a homogeneous medium. Strictly speaking, everything except a vacuum is inhomogeneous. Therefore, “homogeneous medium” here means that the heterogeneity is much smaller than the wavelength of incident light.

Scattering cross section. In optical scattering, the scattering cross section σ_{sca} is defined as the product of the total monochromatic power removed from the incident light as a result of optical scattering and the incident monochromatic light intensity. The concept is used to express effective cross section area that a particle interacts with light. It can also be regarded as the likelihood or strength of a particle to interact with the incident light. It is closely related to the wavelength and polarization state of incident light. By the same token, the absorption cross section σ_{abs} can be defined as the total monochromatic power removed from the incident light by particle absorption.

To determine the optical scattering cross section, the particle is assumed to be surrounded by an imaginary sphere of radius r large enough in the far-field zone. The background medium is assumed to be non-absorbing and the net rate of the electromagnetic energy crossing the surface S of the sphere is always non-negative and is equal to the power scattered by the particle [1]:

$$W^{\text{sca}} = r^2 \int_{4\pi} d\mathbf{r} \langle \mathbf{S}^{\text{sca}}(\mathbf{r}) \rangle \cdot \mathbf{r} \quad (2-1)$$

where $\mathbf{S}^{\text{sca}}(\mathbf{r})$ is the Poynting vector of the scattered optical field. Therefore, the scattering cross section is given by [1]

$$\sigma_{\text{sca}} = \frac{W^{\text{sca}}}{\frac{1}{2} \sqrt{\epsilon_1 / \mu_0} |E_0^{\text{inc}}|^2} = \frac{1}{|E_0^{\text{inc}}|^2} \int_{4\pi} d\mathbf{r} |\mathbf{E}_1^{\text{sca}}(\mathbf{r})|^2 \quad (2-2)$$

Rayleigh scattering. Rayleigh scattering is the elastic scattering of light by particles much smaller than the wavelength of incident light. Here it is normally to define the size parameter $x = ka$ with a being the radius of the particle and $k = \omega / c$ being the wave number of the incident light. Then, the scattering cross section can be given by

$$\sigma_{\text{sca}} = \frac{8\pi(n^2 - 1)x^4 a^2}{3(n^2 + 2)^2} \quad (2-3)$$

here, $n = n_1 / n_0$ is the refractive index contrast to the background.

The x^4 dependence of scattering cross section indicates that the scattering strength is stronger for shorter wavelength. Thus blue light tends to be more efficiently scattered in all directions by tiny particles in the atmosphere and thus forms a blue sky. In two-dimensional case where the particle is replaced by a uniaxial rod, the scattering cross section of incident wave propagating in the plane perpendicular to the rod axis has an x^3 dependence.

Mie scattering. When the particle size is on the order of the wavelength of the incident light, it comes to the regime of Mie scattering. For an isotopically and homogeneously dielectric sphere, the Mie scattering cross section is given by [1]

$$\sigma_{\text{sca}} = \frac{2\pi}{k^2} \sum_{m=1}^{\infty} (2m+1) (|a_m|^2 + |b_m|^2)$$

$$a_m = \frac{S_m(x)S'_m(nx) - nS_m(nx)S'_m(x)}{\varepsilon_m(x)S'_m(nx) - nS_m(nx)\varepsilon'_m(x)} \quad (2-4)$$

$$b_m = \frac{nS'_m(nx)S_m(x) - S_m(nx)S'_m(x)}{nS'_m(nx)\varepsilon_m(x) - S_m(nx)\varepsilon'_m(x)}$$

where S_m and ε_m are Riccati–Bessel functions. Obviously, by estimating coefficients a_m and b_m , the scattering cross section at a certain wavelength for a particle can be evaluated. In literatures, a dimensionless physical quantity – scattering coefficient is widely used, which is defined by $Q_{sca} = \sigma_{sca} / (\text{particle physical cross section})$.

Figure 2-1. The light scattering of a two dimensional pillar ($n=3$) in air. (a) The schematic diagram for the scattering process and the definition of scattering angle. The calculated scattering coefficient of the pillar versus size parameter x on (b) a linear plot and (c) a log-log plot. (d) The scattered far-field intensities versus scattering angle for $x = 0.15$ and $x = 0.65$.

In the Rayleigh scattering regime ($x \ll 1$), the particle can be treated as a dipole with a radiation pattern that is isotropic in the plane vertical to the orientation of the dipole. To the Mie scattering regime ($x \sim 1$), not only dipole mode, other higher order resonant modes with different angular preferences will also be excited by the incident wave. The scattered field is a superposition of the multiple resonant modes, each weighted by the appropriate coefficient a_m and b_m . Therefore, a strong intensity fluctuation with angle can be found in its scattered far field due to constructive and destructive interference. The scattered field and the scattering cross section of a particle can be approximated by numerical method. From the calculated scattering coefficient for a dielectric pillar with index of 3 embedded in air as shown in Fig. 2-1, a strong wavelength dependence of scattering efficiency at Mie scattering regime exists, contrary to that of the Rayleigh scattering which shows x^3 relationship (at $x < 0.3$ regime) in two-dimensional condition. From the scattered far-field, it can be found that the Rayleigh scattering (for $x = 0.15$) is isotropic whereas Mie scattering (for $x = 0.65$) preferentially scatters wave in the forward direction.

2.2.2 Multiple particle scattering

When a medium contains more than one particle within the characteristic length scale L , the light waves have the possibility of being scattered more than once. Each particle is excited by external incident field and the resultant field scattered by all the other particles. If the particle density ρ or scattering cross section σ_{sca} of the individual particle is small, there is no interference between the waves scattered by each particle and thus the scattering is independent. The overall scattering strength increases when the density ρ of particles increases. The overall scattered light intensity is the sum of scattered intensity by each particle. If the particle density ρ is large enough, the optical scattering is no longer independent. The scattered waves will interfere. Thus the overall scattered light intensity should be equal to the square of the overall scattered electric field.

Mean free path is a characteristic length scale to describe the scattering processes. Two important quantities are normally utilized to characterize the scattering strength of the complex disordered medium.

Scattering mean free path. Scattering mean free path is the average length of the photon that travels between two consecutive scattering events. For a disordered medium with monodispersed scatterers with a density of ρ , the scattering mean free path is given by [22]

$$l_s = \frac{1}{\rho \sigma_{sca}}. \quad (2-5)$$

If $l_s > L$, the single particle scattering is dominant; if $l_s < L$, the multiple scattering process dominates. In the multiple-scattering regime, individual scattering is not independent. The resonances between scatterers can affect the overall scattered light field. As the scatterers do not necessarily scatter the light isotropically, angular preference of the particle scattering needs to be taken into consideration.

Transport mean free path. The transport mean free path is defined as the average distance a photon has traveled before its propagation direction is randomized. It is related to scattering mean free path and defined by [17,22],

$$l_t = \frac{l_s}{1 - \langle \cos \theta \rangle} \quad (2-6)$$

where $\langle \cos \theta \rangle$ is the average cosine of the scattering angle. This quantity is normally utilized to characterize the scattering strength of the disorder medium in multiple-scattering regime. For lossless opaque or translucent medium, there should be $l_t < L$.

2.2.3 Structural modification

In the completely random medium, the directional preference of light scattering by individual scatterer is averaged statistically. However, when the scatterer is dense enough, the short-ranged or even long-ranged positional order can exist and evanescent

coupling of adjacent scatterers start to arise. Therefore, the local positional arrangement of scatterers will influence the multiple scattering of light and cannot be overlooked. In two-dimensional disordered structures, the transport mean free path modified by local geometrical arrangements can be given by [23,24],

$$l_t = \frac{k^6}{\pi} \left(\int_0^{2k} \rho(q) F(q) S(q) q^3 dq \right)^{-1} \quad (2-7)$$

where q is the spatial wavevector, $F(q)$ is the form factor of individual scatterer, $S(q)$ is the structure factor which can be given by,

$$S(\mathbf{q}) = \frac{1}{N} \sum_{i,j=1}^N e^{i\mathbf{q} \cdot (\mathbf{r}_i - \mathbf{r}_j)} \quad (2-8)$$

where $\mathbf{r}_i$ donates the center position of the i th scatterer. And $\rho(\mathbf{q})$ is the Fourier transform of the density function $\rho(\mathbf{r}) = \sum_i \delta(\mathbf{r} - \mathbf{r}_i)$.

There are several categories of structures in solids:

- 1) Periodic structure: Scatterers are strictly ordered in a periodic way. The structure has both translational and rotational symmetry. In reciprocal space, the Fourier transform of the structure features well-defined and periodic Bragg peaks. The typical example is a crystal at 0 °K.
- 2) Quasi-periodic structure: The scatterers are ordered but not periodic. The structure has rotational symmetry. In reciprocal space, the Fourier transform of the structure is featured by well-defined Bragg peaks but not periodic. This type of structure are discovered by Dan Shechtman [25] and normally found in aluminum alloys [26], e.g. Al-Li-Cu.

3) Long-range ordered structure: The particles are arranged in a certain range of translational periodicity. But in short distance, the positional disorder exists. One example is the crystal at room temperature (RT).

4) Short-range ordered structure: The scatterers are arranged periodic in short distance, but both translational and rotational symmetry are absent in long distance range. A typical example of this is amorphous silicon.

5) Random structure: The particles are randomly distributed. Neither rotational symmetry nor short-range/long-range translational symmetry exists. The typical example is gas.

2.2.4 Different scattering regimes

Normally, according to the scattering strength, the disordered medium can be classified into three regimes: ballistic regime, diffusive regime and localization regime [27].

Figure 2-2. The schematic diagram of light transportation in the disordered media at different scattering regimes: (a) ballistic regime, (b) diffusive regime and (c) localization regime.

Ballistic regime. In the ballistic regime, the transport mean free path is much larger than the system length ($l_t \gg L$), where the incident light travels through the disordered medium without scattering (as shown in Fig. 2-2(a)). If the light is scattered, but the trajectory is a nearly ballistic path, the medium is called quasi-ballistic medium.

Diffusive regime. In the diffusive regime, the transport mean free path is shorter than the system length, whereas much longer than the wavelength ($L \gg l_t \gg \lambda$). In this regime, the multiple scattering of light dominates however the light interference is neglected. The transport of light in the medium can be approximated by the diffusion equations.

Localization regime. When the transport mean free path is short enough ($l_t k \approx 1$, k is the wavevector in the disordered medium), interference between different multiple scattered paths dramatically modifies the average transport process and the wave starts to be trapped within the medium (as shown in Fig. 2-2(c)). A number of “hotspots”, the high intensity area induced by constructive interference, can be found in the different regions of the medium.

Apparently, from ballistic regime to localization regime, the scattering strength of disordered medium increases and the photon gets more and more easily trapped in the medium.

2.3 Anderson localization

In 1958, Philip Anderson proposed that when a certain amount of disorder is introduced into an otherwise periodic semiconducting crystal, the transport of electrons can come to a complete halt due to the interference of multiple scattering of electrons and the wavefunctions of electrons become strongly localized [15]. Such phenomenon is widely known as Anderson localization. Inspired by research on the Anderson localization of electrons, physicists began to ask whether scattering and interference of light can give rise to a similar localization phenomenon in the disordered photonic medium [28]. Practically, similarities can be found for an electron scattered by random potentials and a photon scattered by disordered scatterers. For an electron with effective mass of m^* moving in the semiconductor crystal with potential $V(\mathbf{x})$, which varies randomly in space, the Schrödinger equation can be given by Eq. (2-9). An electron with sufficiently negative energy E may get localized in a region with random

potential which is deep. In the case of light wave with frequency ω propagating in the disordered photonic medium, the classical Maxwell equation can be written in a form similar to the Schrödinger equation as by Eq. (2-10) [27]. Here, $\mathbf{E}(\mathbf{x})$ is the electric field amplitude and $\varepsilon(\mathbf{x}) = \varepsilon_{\text{fluct}}(\mathbf{x}) + \varepsilon_0$ is the position-dependent refractive index with ε_0 being the average value and $\varepsilon_{\text{fluct}}(\mathbf{x})$ being the fluctuating part, which scatters the wave and plays a role analogous to the random potential in the electron case.

For electron:
$$\left[\frac{-\hbar^2}{2m^*} \nabla^2 + V(\mathbf{x}) \right] \psi(\mathbf{x}) = E \psi(\mathbf{x}) \quad (2-9)$$

For photon:
$$-\nabla^2 \mathbf{E}(\mathbf{x}) + \nabla(\nabla \cdot \mathbf{E}(\mathbf{x})) - \frac{\omega^2}{c^2} \varepsilon_{\text{fluct}}(\mathbf{x}) \mathbf{E}(\mathbf{x}) = \varepsilon_0 \frac{\omega^2}{c^2} \mathbf{E}(\mathbf{x}) \quad (2-10)$$

However, based on the analogy between the Schrödinger equation (2-9) and Maxwell equation (2-10), several differences can be identified. Firstly, the quantity $\varepsilon_0 \omega^2 / c^2$ which is the energy analogue is always positive, and therefore the bound states of photon in deep negative potential wells are forestalled. Secondly, different from an electronic system where localization is more favored by lower electron energy, lowering the photon energy will not do good to the localization of light. This is because lowering the photon energy will weaken the scattering efficiency, as indicated by the comparison of Rayleigh and Mie scattering efficiency in Fig. 2-1. In the high-frequency regime, the Eq. (2-10) becomes less effective and the geometric optics dominates the light wave scattering. And finally, $\varepsilon(\mathbf{x}) = \varepsilon_{\text{fluct}}(\mathbf{x}) + \varepsilon_0 > 0$ everywhere in the disordered photonic medium indicates that the energy eigenvalue is always larger than the effective potential $|\varepsilon_{\text{fluct}}(\mathbf{x}) \omega^2 / c^2|$. Thus, it is more difficult for the light to get localized within the disordered photonic medium than electrons in the electronic system where $|E| < |V(x)|$ is often satisfied. The localization of light can only happen, if the localization indeed exists, in an intermediate frequency range with its energy

located at the positive energy continuum higher than the highest of the potential barriers [27].

In the disordered photonic medium, the photon is barely localized within a single “potential well”. However, in the Mie scattering regime where the scattering efficiency is high, the interference and multiple scattering of light can possibly form closed loops, in which photons are confined and randomly shaped standing-wave patterns are constructed. The likelihood of light localization depends heavily on the dimensionality of the disordered medium. According to the scaling theory [29], the localization can happen even with slight disorder in one and two dimensions only if the system size is sufficiently large. And in three-dimensional (3-D) conditions, the photon returning to the original point is more difficult than in lower dimensions. The possibility for photon localization depends on the scattering mean free path l_s . According to the well-known Ioffe-Regel criterion [30,31], the critical point of transition from extending electric field to localization in three dimension is $2\pi l_s / \lambda < 1$.

Anderson localization is an intrinsic wave-like phenomenon, which has been observed in many different systems of three dimension, including microwaves [32], acoustic waves [33,34], and even matter waves [35–37]. However, in the realm of light wave, achieving Anderson localization in 3-D medium is elusive and often controversial [38,39] as it is very challenging to find a material with simultaneous high scattering efficiency and low absorption. In 2007, Mordechai Segev and his coworkers reported the transverse localization of light in a two dimensional disordered photonic crystal, however the wave propagates in the third direction [40]. It has been proposed that introducing slight disorder into photonic crystal structures may contribute to the localization of light with the assistance of photonic bandgap [27,41]. However, no confirmative results have been reported [42].

2.4 Characteristic length scales for the random laser

In disordered photonic medium with gain, light scattering plays a positive role for random lasing. It increases the path length or dwell time of the photon within the active medium and thereby enhance the amplification. If the scattering strength is very strong, some photons may return to a coherence volume ($\sim \lambda^3$) they have visited before, and thus the optical feedback is provided. There are some parameters often used to characterize the photon behaviors with the existence of gain in the disordered medium.

Gain length l_g and amplification length l_{amp}

The gain length is defined as the path length over which the light intensity has been enhanced by e . And the amplification length is the average distance between the starting and ending point for paths of length l_g . In homogeneous medium, the two length scales are identical, while in random medium system, these two lengths are different. The relation between two lengths for 3-D diffusive medium is given by [43],

$$l_{amp} = \sqrt{Dl_g / v} = \sqrt{l_t l_g / 3} \quad (2-11)$$

where v is the speed of light, $D = vl_t / 3$ is the diffusion coefficient. The gain length is analogous to the inelastic length l_i , which is defined as the path length over which the light intensity has been reduced by e through absorption. Similarly, analogous to amplification length, the absorption length is given by $l_{abs} = \sqrt{l_t l_i / 3}$.

Critical length L_c

According to the diffusion model [44], the critical length is a length scale to judge whether the disordered medium can lase or not. It is related to the amplification length,

$$L_c = \pi \sqrt{l_t l_g / 3} . \quad (2-12)$$

If the random system size L is larger than the critical length, the threshold can be reached; otherwise the threshold cannot be reached no matter what the pumping intensity.

2.5 Feedback mechanism of random laser

Due to the system complexity, the physics, especially the feedback mechanism for lasing in disordered medium is still on debate. For more than 20 years, lasing in disordered medium has been intensively studied both theoretically and experimentally. Up to now, most popular theories of the feedback mechanisms for such lasers can be classified into two categories: incoherent (non-resonant) feedback and coherent (resonant) feedback.

2.5.1 Incoherent feedback

When spontaneously emitted photons are multi-scattered in the disordered medium, the path length and dwell time of the photon will be increased. Correspondingly, the amplification of light will be increased because additional photons are generated through stimulated emission. Longer path length or dwell time within the active medium also means that the chance of photon escaping from the gain area is smaller, leading to smaller cavity losses. When the scattering intensity of disordered medium is strong enough, the amplification of light will exceed the extinction or losses, thus there is a net gain for the light. This process is called incoherent feedback, or non-resonant feedback, because the feedback is phase insensitive and frequency independent. Due to the incoherent feedback mechanism, the emission spectrum is always featured by a linewidth of several nanometers and the mean frequency only depends on central gain frequency of the gain medium, where the gain coefficient is the highest. Over the broad emission spectrum envelope, small lasing spikes can be observed which are generated by some long optical paths. These lasing spikes change from pulse to pulse due to the randomness of scattering paths.

For incoherent feedback process, the propagation of light can be described as a diffusion process. Therefore, photon energy density in the disordered medium can be described by a diffusion equation [17,44]:

$$\frac{\partial W(\mathbf{r},t)}{\partial t} = D\nabla^2 W(\mathbf{r},t) + \frac{\nu}{l_g} W(\mathbf{r},t) \quad (2-13)$$

where D is the diffusion coefficient, $W(\mathbf{r},t)$ is the photon energy density. The general solution to Eq. (2-13) can be written as [44]

$$W(\mathbf{r},t) = \sum_n a_n \psi_n(\mathbf{r}) e^{-(DB_n^2 - \nu/l_g)t} \quad (2-14)$$

with $\psi_n(\mathbf{r})$ being the eigenfunctions and B_n being the eigenvalues.

The solution for $W(\mathbf{r},t)$ in Eq. (2-13) changes from exponential decay to exponential increase in time when crossing the threshold

$$DB_1^2 - \frac{\nu}{l_g} = 0 \quad (2-15)$$

where B_1 is the lowest eigenvalue [44]. Therefore, the threshold can be decided. Coupling the diffusion equation of light with rate equation of active material, the threshold, emission wavelength, and transient oscillation of the emission intensity can be predicted [43].

Figure 2-3. Schematic diagrams for the working principle of lasers with traditional cavity and disordered cavity. (a) Conventional laser cavity. (b) Disordered laser cavity with an illustration of coherent feedback (red dashed arrows) and incoherent feedback (yellow dashed arrows). (c) Typical emission spectra of a conventional laser and a disordered laser with coherent feedback and incoherent feedback respectively [19].

2.5.2 Coherent feedback

For coherent feedback, the photons are so strongly scattered that the propagation path forms a closed loop. This is like the scenario of conventional lasers. Therefore, the interference effect cannot be ignored. Fundamentally different from incoherent feedback, coherent feedback supports multiple lasing peaks with very narrow spectral

features. The frequencies of peaks are determined by the length of closed loops. For incoherent feedback, the photon travels a very long path within the gain medium and even out of the gain medium. Therefore, the strong mode overlap and, thus mode coupling, exist. Quality factors of modes are comparable except for some anomalies corresponding to localized modes. However, for coherent feedback, the closed loop isolates the different modes. The quality factors have large fluctuations. The threshold of the laser is determined by the mode with the highest quality factor. Unlike the incoherent feedback laser whose lasing spikes change from pulse to pulse, the lasing peaks of coherent feedback remain the same wavelength. This demonstrates the closed cavity feedback mechanism. The coherent feedback emission in disordered medium has been demonstrated to have a typical photon statistics close to that of the conventional emission [45].

By using the diffusion approach, it is not able to explain the consistent lasing peaks for strongly scattering medium as the interference effect is neglected. Time-independent wave equations [46,47] had been developed and found useful to predict the threshold of the complex system. However, it is hard to deal with the laser phenomena above the threshold. Then, a time-dependent theory based on the coupling of Maxwell equations and rate equations of the electronic population density has been developed [48]. The time-dependent Maxwell equations are given by Eq. (2-16), where $P(\mathbf{r},t)$ is the electric polarization density which gives amplification or gain. In the presence of an electric field, $P(\mathbf{r},t)$ obeys Eq. (2g-17), where $\Delta N(\mathbf{r},t)$, ω_α and $\Delta\omega_\alpha$ represent population inversion, the center frequency and linewidth of the atomic transition from upper laser level to lower laser level respectively. Consider a four-level gain medium, electrons pumped from level 0 to level 3 relax quickly to level 2 and then to level 1 through radiative and non-radiative transitions. Finally, the electrons in level 1 get quickly de-excited back to level 0. Therefore, $\Delta N(\mathbf{r},t)=N_2(\mathbf{r},t)-N_1(\mathbf{r},t)$. The polarization density $P(\mathbf{r},t)$ is also coupled in the rate equations when given an external pumping rate $P_r(t)$. Here, (N_3, N_2, N_1, N_0) are electron populations in four

levels and $(\tau_{32}, \tau_{21}, \tau_{10})$ represents the time constant for electron transition from one level to another level.

$$\nabla \times \mathbf{E}(\mathbf{r}, t) = -\frac{\partial \mathbf{B}(\mathbf{r}, t)}{\partial t}, \quad \nabla \times \mathbf{H}(\mathbf{r}, t) = \varepsilon(\mathbf{r}) \frac{\partial \mathbf{E}(\mathbf{r}, t)}{\partial t} + \frac{\partial \mathbf{P}(\mathbf{r}, t)}{\partial t} \quad (2-16)$$

$$\frac{d^2 \mathbf{P}(\mathbf{r}, t)}{dt^2} + \Delta \omega_\alpha \frac{d\mathbf{P}(\mathbf{r}, t)}{dt} + \omega_\alpha^2 \mathbf{P}(\mathbf{r}, t) = -\frac{6\pi\varepsilon_0 c^3}{\tau_{21} \omega_\alpha^2} \Delta N(\mathbf{r}, t) \mathbf{E}(\mathbf{r}, t) \quad (2-17)$$

$$\frac{dN_3(\mathbf{r}, t)}{dt} = P_r(t) N_0(\mathbf{r}, t) - \frac{N_3(\mathbf{r}, t)}{\tau_{32}} \quad (2-18)$$

$$\frac{dN_2(\mathbf{r}, t)}{dt} = \frac{N_3(\mathbf{r}, t)}{\tau_{32}} + \frac{\mathbf{E}(\mathbf{r}, t)}{\hbar \omega_\alpha} \cdot \frac{d\mathbf{P}(\mathbf{r}, t)}{dt} - \frac{N_2(\mathbf{r}, t)}{\tau_{21}} \quad (2-19)$$

$$\frac{dN_1(\mathbf{r}, t)}{dt} = \frac{N_2(\mathbf{r}, t)}{\tau_{21}} - \frac{\mathbf{E}(\mathbf{r}, t)}{\hbar \omega_\alpha} \cdot \frac{d\mathbf{P}(\mathbf{r}, t)}{dt} - \frac{N_1(\mathbf{r}, t)}{\tau_{10}} \quad (2-20)$$

$$\frac{dN_0(\mathbf{r}, t)}{dt} = \frac{N_1(\mathbf{r}, t)}{\tau_{10}} - P_r(t) N_0(\mathbf{r}, t) \quad (2-21)$$

The time-dependent theory [48] is powerful in determining the laser threshold, field patterns and emission spectra and particularly suitable for the simulation of laser dynamics above the threshold, such as gain competition, gain saturation. The drawback of this method is storage- and time-consuming.

Coherent feedback normally occurs in the strongly scattering medium with its l_t close to the wavelength. If the scattering strength of disordered medium is brought to the localization regime ($l_t k \simeq 1$, k is the wavevector in the disordered medium), connections between lasing and Anderson localization may exist, especially for low-dimensional medium where the probability of the scattered photons going back to its original point is much higher than that in 3-D case [49,50]. However, for the weak scattering medium where $l_t k$ is tens or hundreds of thousand, the coherent feedback has still been demonstrated experimentally and numerically [51,52]. This indicates the

complex physics behind the multiple scattering of light in disordered medium, especially when the gain is introduced.

2.6 The development and application of random laser

2.6.1 Lasing in different disordered media

Overall, the scatterers of the random laser can be categorized into two types: passive scatterers with no gain, and active scatterers, which work as scatterer and gain simultaneously. For the former one, the modulation of lasing property can be easily realized by changing the scatterer size, shape, density and polarization [53,54] with slight influence on the gain. This is advantageous for the research on laser physics of disordered medium. Dye solutions containing various dielectric particles (e.g. ZnO, TiO₂, GaAs) [16], metallic scatterers [55] and liquid crystals [53,54], has been a widely-utilized platform of this kind of random lasers. For the latter type, higher performance of lasing can be realized as the photon confinement can be better due to higher index contrast with the elimination of background material (e.g. no liquid solutions to dissolve the dye for light amplification). High-index semiconductor nanostructures with direct bandgap (e.g. ZnO [56,57], GaAs [58], ZnSe [59]) have been extensively exploited as both scatterers and gain medium for multimode random lasing with coherent feedback. Additionally, semiconductor membranes [23,60], and rare-earth ion doped powders/ceramics [61,62], random fibers [63], organic films [64] and nanofibers [65,66], hybrid organic-inorganic composites [67,68], etc., are also developed for random lasing applications.

Surface plasmonic resonances of metallic particles have been widely employed for various applications. And plasmonic scatterers have also been incorporated into disordered medium with gain material to improve the performance of the lasers with large scattering cross sections to enhance photon trapping and surface plasmon resonances induced near-field enhancement for larger gain [69,70]. Besides making use of the elastic scattering, inelastic scattering process is also proposed for photon

trapping and therefore, lasing. BaSO₄ powder has been used as both the scattering medium and Raman gain to achieve Raman lasing in the visible range [71]. And multiphoton absorption and energy transfer mechanism in ZnO [72,73] and Nd³⁺ doped fluoroindate glass powder [74] have been utilized to achieve upconversion emission.

With regard to the emission frequency which depends on the selected gain material, random lasing in different frequency range has been demonstrated. Due to its large bandgap, ZnO [56,57], GaAs [58], ZnSe [59] particles or films have been exploited as scattering and gain medium for the generation of UV and visible random emissions. Rare-earth ion doped ceramics [61,75] and various dyes [76] have been widely utilized as gain material for random emission in visible and near-infrared frequency range. In the near-infrared regime, semiconductor membranes containing quantum dot [23,77] or quantum well [60,78] as the gain medium are also usually for random lasing. In 2013, a quantum cascade laser (QCL) has been utilized to generate mid-IR random emission around 10 μm [20], which is the longest wavelength for lasing in disordered medium demonstrated before the commencement of this research. The scattering medium of the laser consists of air holes randomly distributed on the slab of QCL wafer as shown in Fig. 2-4 (h) and (i). Due to the weak scattering efficiency of air holes for transverse-magnetic polarized light of QCL, the threshold of random laser is high and the number of emission peaks is limited.

Figure 2-4. Various disordered media utilized for laser applications. (a) ZnO particle cluster [57]. Inset: spatially resolved photoluminescence (PL). (b) Random gold nano-island structures [70]. (c) Random photonic glass material made of polystyrene (PS) spheres [79]. (d) Random $\text{Nd}^{3+}:\text{LuVO}_4$ particles [62]. (e) Disordered crystalline nanofibers made of para-sexiphenyl (p-6P) [66]. (f) A fiber with disordered hollow core structure [63]. (g) A suspended semiconductor membrane with random air holes [77]. Quantum dots are embedded within the membrane and map of the measured PL intensity is illustrated. Inset: cross section view of the structure. (h) 3D schematic diagram of mid-IR random laser with randomly distributed air holes [80]. (i) The cross section view of the fabricated mid-IR random laser at $\sim 10 \mu\text{m}$ [20]. Inset: typical emission spectrum of the mid-IR random laser. (The figures are reprinted from the corresponding references)

2.6.2 Potential applications of random laser

Although both mirrorless and random, light in a random laser does not lose wave character and optical information [42]. This leads to a series of interesting physical phenomena that could reveal great potential for novel disorder-based optical applications.

As the feedback is provided by optical multiple scattering of the disordered medium rather than the reflection of the mirror, the fabrication cost of the disordered laser is quite low as compared to that of the conventional lasers. Lasing can be realized with very small sizes [81], flexible shapes [53,82] and on many substrates [83,84], which opens up possibilities for many new applications, including display, optical batteries [85], tumor detection [86,87] and photonic barcode for document encoding, material labeling or military identification of friend and foe [17,19]. More often than not, the laser emission spectrum of disordered medium contains many emission peaks and each emission peak corresponds to a different feedback loop. Therefore, the wavefronts corresponding to different emission peaks are not correlated, resulting in low spatial coherence of the emission as demonstrated in Reference [88]. This makes such kind of laser promising for full-field imaging and display lighting.

Low spatial coherence source for full-field speckle-free imaging

Conventional lasers as the source in the full-field imaging system should have high photo degeneracy (that is, the number of photons per coherence volume). However, they also have high spatial coherence due to the highly collimated beam path. When photons are scattered off a rough surface, random path lengths are generated. The mutually coherent photons would interfere at the detector and result in an artificial modulation in the measured intensity (speckles). Low coherent sources such as thermal white light and light-emitting diode (LED) have been sought to mitigate the speckle effect. However, they have been limited by their low emission intensity and conversion efficiency. On the other hand, random lasers have high photo degeneracy due to the coherent emission characteristic as well as large number of spatial modes with

uncorrelated phases. Their distinct and rich structures combine to produce spatially incoherent emission.

Figure 2-5. (a) Schematic diagram of the experimental set-up for full-field imaging. Images taken with LED (b), random laser (c) and conventional laser sources (d) respectively, showing that random laser produces a speckle-free image as that of LED, while conventional laser produces speckles in the bright area of the image. (reprinted from [89])

A random laser that is speckle-free has been demonstrated recently [89]. A scattering film was placed on the illumination side of the test chart in order to impart random phase delays to the incident light, resulting in speckled illumination of the object if the source has a high degree of spatial coherence. The test chart was imaged with three light sources: LED, random laser and conventional laser. From Fig. 2-5, it is noted that unlike the highly spatial-coherent source (conventional laser), random laser eliminates the interference effects and produce a clean image of the object as that of LED.

2.7 Summary

This chapter details the theory of single particle and multiple particle scattering processes. Influenced by the positional correlation of scatterers, the scattering strength

of disordered medium is characterized by a physical quantity — transport mean free path, with which the disordered media are categorized into different regimes. Anderson localization of light in the strongly scattering disordered medium has then been reviewed. When the gain is included, the critical length of the complex medium is introduced to judge whether the disordered system can lase or not. Then, incoherent and coherent random laser feedback mechanisms corresponding to different scattering regimes have been detailed with respective theoretical models. With more than twenty years' development, random lasers have been greatly developed in various systems. Many unique applications have been proposed and even demonstrated, of which the most significant example is speckle-free full-field imaging.

3 The Terahertz Technology and Terahertz Quantum Cascade Lasers

At the beginning of this chapter, the terahertz (THz) technology and THz quantum cascade laser (QCL) will be briefly reviewed. The fundamental theories of the THz QCL will then be presented, followed by a detailed analysis of THz QCL active region design which will be used in the whole thesis. Following this, double-metal (DM) waveguide structure will be introduced. The fabrication and characterization of typical THz QCLs based on DM waveguide will then be briefed, and finally some preliminary experimental results are to be analyzed.

3.1 The terahertz technology

Lying between the microwave and infrared frequencies, the electromagnetic spectral band ranging from ~ 0.3 THz to 10 THz is known as the “terahertz (THz) gap”, where the technology for wave generation, manipulation, and detection is least developed. THz wave shares some properties with the microwave and mid-infrared (mid-IR) optical wave. Similar to the mid-IR wave, THz wave propagates in a line of sight and has limited penetration through fog and clouds and cannot pass through liquid water due to severe scattering and absorption. As a result of the molecular rotations, vibrations and atomic fine-structure transitions, many molecules have their absorption “fingerprint” in the THz wave frequency range [90]. Therefore, like mid-IR wave, THz wave is also entitled a great utility for spectroscopy and has been applied to numerous materials in different applications, including gas detection [90], material analysis [91], pharmaceutical sciences, biomolecule dynamics [92], etc. THz wave is not ionizing yet can penetrate into biological tissues for some distance. Therefore, it is of significant interest for imaging application and being regarded as a promising replacement of X-rays because it is much safer. Since the photon energy is lower than the band-gap of nonmetallic materials, THz wave can penetrate various non-conducting materials,

including clothes, polymer, wood, plastic, paper, ceramics, and lightly doped semiconductors. It can therefore be used to inspect these materials for hidden contents, such as weapon and explosives. The THz wave is similar to microwave, but normally has higher resolution. THz wave also has potential applications in high bandwidth communication as its frequency is much higher than that of radio wave and microwave. As the atmosphere has strong absorption for THz wave at specific water vapor absorption bands, the application of THz wave for long-distance communications is critically limited. However, THz wave could still be useful in imaging and wireless networking at the indoor distance range [93]. Potential uses exist in empty space telecommunications, such as aircraft to aircraft, aircraft to satellite, and satellite to satellite communications.

Figure 3-1. The “Terahertz (THz) gap” lying between the infrared and microwave frequencies.

Driven by so many important potential applications and the recent technological development in photonics and nanotechnology, numerous technological breakthroughs in the THz field have also been achieved in the past two decades. THz quantum well photodetector is developed for sensitive and ultrafast THz wave detection [94–96]. The introduction and application of new materials such as metamaterials [97,98] and graphene [99–102] in novel THz photonic devices pave the way for high-speed modulation. Various THz radiation sources have also been developed to generate the THz wave. For example, 3 μ W THz wave at 1.9 THz was generated by upconverting the microwave source using solid-state frequency multipliers [103]. The uni-travelling-

carrier photodiode based on photomixing mechanism has been developed and a maximum continuous-wave (CW) output powers of 20 mW and 10 μ W have been achieved at 100 GHz and 1 THz [104], respectively. Layered superconductors, such as $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$, have been utilized to generate coherent CW THz radiation of several hundreds of microwatts [105,106]. The operation mechanism is based on the Josephson Effect, which converts a direct-current voltage into a high-frequency electric current, and thus the emission frequency is bias-voltage tunable. And the most significant THz source, THz QCL, which was first realized by Kohler [21] *et al.*, has demonstrated watt-level power emission ability in pulse mode [107] and 0.23 W in CW mode [108]. RT THz emission based on QCL has also been demonstrated utilizing difference-frequency generation mechanism with a mid-IR QCL [109]. In the following sections, THz QCL will be reviewed in detail.

3.2 Terahertz quantum cascade laser

As discussed in the previous section, various techniques have been demonstrated to generate THz wave. THz QCLs are probably the most promising coherent radiation source in the development of THz technologies due to its overall better performances, including compact size, high efficiency, high power, high temperature operation and broad frequency coverage. However, THz QCL is not easy to achieve.

3.2.1 Quantum cascade laser and its properties

In 1970s, two Russian physicists Kazarinov and Suris proposed to use quantized electronic intersubband transition in a semiconductor superlattice under high electric field to achieve optical amplification [110]. With more than two decades' endeavor, the first mid-IR intersubband laser based on $\text{AlInAs}/\text{GaInAs}$ heterostructures, named quantum cascade laser, has been demonstrated by Faist *et al.* at Bell laboratory in 1994 [111].

Figure 3-2. The conduction band energy diagram of the first quantum cascade laser (QCL) operating at 4.2 μm . The moduli squared of the wavefunctions and an overview of electron movement within the structures are illustrated, with electron tunneling indicated by direct arrows and laser transition indicated by wavy arrows. Each period of the structure consists of active region layers and digitally graded alloy layers working as an electron injector. (Reprinted from [111])

Fig. 3-2 is the conduction band energy diagram of first QCL. Grown by molecular beam epitaxy, QCL semiconductor heterostructures ($\text{Al}_{0.48}\text{In}_{0.52}\text{As}/\text{Ga}_{0.47}\text{In}_{0.53}\text{As}$) consists of 25 active regions alternated with injector regions (digitally graded alloy in this work). An electric field is applied as visualized by linear slope of electronic potential. Electrons are injected from injector to upper laser state (level 3) of the active region through resonant tunneling, and then undergo radiative as well as non-radiative transitions to lower laser state (level 2). In lower laser state, electrons are quickly depopulated by an underneath level (level 1) through LO-phonon scattering and then tunnel to the next injection region to be reused in the next period. The developed QCL operated around 4.2 μm with a high threshold current density of 15 kA/cm^2 and maximum operation temperature of 88 K, mainly limited by the low optical gain.

As compared to the conventional semiconductor lasers, QCL mainly has three unique properties. Firstly, QCLs are unipolar devices, of which the stimulated emission

happens between two subbands of the conduction band. For conventional diode lasers, stimulated emission occurs between conduction band and the valence band through electron-hole recombination. Secondly, due to the cascaded structure, the injected electrons can be recycled N_p times (N_p is the number of module of the QCL), leading to a quantum efficiency proportional to N_p . The output power can thus be greatly enhanced in QCLs [112]. Lastly, the emission properties of QCLs can be "band-structure engineered" [113]. The quantum well thickness, barrier thickness and band-offset can be designed to tailor the energy levels, carrier scattering rates, optical dipole matrix elements and other parameters to optimize the performance.

3.2.2 The first terahertz quantum cascade laser

Ever since its first demonstration, QCL has been a popular research topic around the world and rapid development has been made over the laser's performance, including expanding the emitting wavelength coverage from 2.6 μm [114] to 24 μm [115], improving the operational temperature to above RT [116], realizing CW operation [117] and improving the CW emission power to watt level [118] and pulsed peak power to the level of tens of watt [119]. With the rapid maturation of QCL wafer design and fabrication technology, mid-IR QCLs have been successfully commercialized in recent years [120].

Figure 3-3. (a) The conduction band energy diagram of the first THz QCL operating at 4.4 THz. The moduli squared of the wavefunctions are illustrated with miniband regions represented by shaded areas. The optical transition takes place between state 2 and state 1. Each period of the structure consists of a superlattice (SL) active region and an injector. (b) The optical modal profile along the growth direction of the device. The waveguide core consisting of 104 repetitions of the active region is sandwiched by top metal contact and bottom contact layer. (Reprinted from [21])

Despite the great breakthrough in mid-IR frequency region, it is much more difficult to extend the emission wavelength of QCLs to THz range due to multiple problems. Firstly, as the photon energy is quite small, it is difficult to selectively inject electrons into the upper laser state and selectively as well as efficiently extract them from the lower laser state. And the conventional waveguide scheme is not suitable for THz wave generation, as it would require extremely thick core and cladding layers due to the long wavelength. In addition, as the free carrier absorption has a wavelength dependence of λ^2 , large free carrier absorption losses impedes the THz lasing. Until 2002, the first THz QCL was realized with novel bandstructure and waveguide scheme [21]. As shown in Fig. 3-3(a), laser transition happens between laser state 2 and state 1. The chirped superlattice makes the lower miniband flat at the operation voltage, allowing efficient electron depletion from laser state 1. Meanwhile, the carriers are tunneling from the injector ground state g into the laser state 2 of the next active region. The adoption of a novel waveguide structure, the so-called “semi-insulating surface-plasmon waveguide”, also contributes to the laser emission greatly. As shown in Fig. 3-3(b), a n^+ GaAs layer with reasonable doping concentration below the 104 period of active superlattice was created, resulting in a large modal overlap ($\sim 47\%$) with the active region which has low absorption losses and provides gain when pumped. This laser emits at 4.4 THz with an output power of more than 2 mW in pulse mode at 8 K.

3.2.3 The development and application of terahertz quantum cascade laser

Since the first demonstration of THz QCL, tremendous development has been made in this field. The operation frequencies have been realized in a range of 1 – 5 THz. Watt-level power emission in pulse mode [107] and over 0.23 W in CW mode [108] at 10 K have been reported. The maximum operation temperature of THz QCLs has been pushed to 200 K [121], however much more efforts are necessary in terms of increasing the working temperature achievable by thermoelectric cooling (240 K). Even though RT emission of coherent THz wave based on difference-frequency generation

mechanism has been demonstrated using a mid-IR QCL [109], the output power is limited to milliwatt level due to small conversion efficiency [122]. Despite the limited temperature performance, wave engineering of THz QCL emission is readily developed to achieve specific functionalities with unprecedented performances. Various resonators (e.g. DFB gratings [123], photonic crystals [124], disks [125], external cavities [126], quasi-periodic structures [127,128], etc.) and beam engineering techniques [129,130] have been incorporated in the THz QCLs for spectrum or beam control. Wide frequency tunability of 330 GHz has been achieved with a moveable MEMS object to manipulate the transverse optical mode [131]. Phase locking [132] and active mode locking [133] of THz QCL lasing have been reported. More recently, THz frequency combs have been generated from a THz QCL with a chirped distributed-feedback reflector for dispersion compensation [134].

With such a great performance progress, THz QCLs have been applied in various practical applications. Due to high average output powers, THz QCLs have been used for THz real-time imaging (~30 frames per second) [135,136] and confocal microscopy [137] with relatively high spatial resolution as shown in Fig. 3-4. THz heterodyne receiver based on THz QCLs has been developed [138] owing to high power and good beam qualities. By virtue of the narrow linewidths (< 1 MHz) and electrical/thermal tuning ranges of several GHz, THz QCLs are suitable for high-resolution gas spectroscopy [139,140]. THz QCLs have been commercialized with interchangeable QCL modules for requirement of different applications. A built-in sterling cycle cooler contributes to a portable system with a size of tens of centimeters and a weight of only several kilogram weights [141].

Figure 3-4. The imaging applications of THz QCLs. First row: THz real-time imaging utilizing a microbolometer focal-plane array with (a) a photograph of a dried seed pod, (b) real-time THz image (Reprinted from [136]). Second row: confocal THz imaging using a three-dimensional (3-D) moving stage to target the focus point and scan the object surface with (c) a photograph of a fresh leaf, (d) confocal THz image of the leaf and (e) higher-resolution confocal THz image (200×200 pixels) of the white rectangular area in panel (d) (Reprinted from [137]).

3.3 Fundamental theories of terahertz quantum cascade laser

3.3.1 Subbands of terahertz quantum cascade laser

QCL is a superlattice structure which consists of multiple heterostructures. Quantum wells are sandwiched by barrier layers with much higher bandgaps. For THz QCL,

AlGaAs is for barrier layers and GaAs is for quantum well layers. The semiconductor layers have nanoscale thicknesses, on the order of the electron's DeBroglie wavelength. Thus, the electron motion is restricted in the growth (z-) direction with quantized energies. The electron wavefunctions and energy states in the QCL superlattice can be obtained by solving Schrodinger equation in the framework of effective mass theorem and envelope function approximation. The electron wavefunction can be given by [142],

$$\psi(\mathbf{r}) = \psi_n(\mathbf{r}) u_{n,\mathbf{k}}(\mathbf{r}) \quad (3-1)$$

where $u_{n,\mathbf{k}}(\mathbf{r})$ is the Bloch state wavefunction and $\psi_n(\mathbf{r})$ is the slowing varying envelop function which satisfies the effective mass approximation. The solution for envelop function is given by

$$\psi_n(\mathbf{r}) = \frac{1}{\sqrt{S_{\parallel}}} e^{i\mathbf{k}_{\parallel} \cdot \mathbf{r}_{\parallel}} \psi_n(z) \quad (3-2)$$

where $\mathbf{k}_{\parallel}$ is the in-plane wavevector, $S_{\parallel}$ is the cross-section area of the quantum well layer for normalization. $\psi_n(z)$ satisfies one-dimensional Schrödinger equation

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

where $m^*(z)$ is the effective mass, E_n is the n -th subband energy, and $E_c(z)$ the conduction bandedge potential profile including externally applied field and local variations due to space charge. Following this, the total energy of n -th subband including in-plane free electron kinetic energy can be given by

$$E_n(\mathbf{k}_{\parallel}) = E_n + \frac{\hbar^2 k_{\parallel}^2}{2m^*} \quad (3-4)$$

To solve Eq. (3-3), the conduction bandedge potential profile $E_c(z)$ has to be provided. Population of the accumulated electrons in different electronic states

introduces space charge effect which influences $E_c(z)$. Thus, it is necessary to solve Poisson equation [143]

$$\frac{d}{dz} \left[\varepsilon(z) \frac{d}{dz} \varphi(z) \right] = -\rho(z) \quad (3-5)$$

with $\varphi(z)$ being the electrostatic potential, $\varepsilon(z)$ being the position-dependent permittivity, and $\rho(z)$ the charge density. The conduction band profile is given by [143]

$$E_c(z) = E_{c,0}(z) - e\varphi(z) \quad (3-6)$$

where $E_{c,0}(z)$ is the inherent conduction band profile. The coupling of Poisson and Schrödinger equations here requires iterative calculation for a self-consistent solution. The validity of the model described above has been verified by M. G. Burt [144], who theoretically compared the effective mass envelope solution with the exact atomic wavefunctions and found that the model provides an excellent approximation even for structures with atomic abrupt boundaries, on the condition that the envelope function is slowly varying.

3.3.2 Intersubband radiative emissions and optical gain

The gain of a QCL is provided by stimulated emission of photons through the electron transition between two subbands of the quantum well superlattice. To calculate the optical gain, it is necessary to inspect the radiative emissions in a QCL, including spontaneous emission and stimulated emission.

3.3.2.1 Spontaneous emission

The electron transition rate between two subbands in QCL can be given by Fermi's golden rule [142],

$$W_{i \rightarrow f} = \frac{2\pi}{\hbar} \left| \langle \psi_i | \mathbf{H} | \psi_f \rangle \right|^2 \delta(E_i - E_f - \hbar\omega_0) \quad (3-7)$$

where ψ_i and ψ_f are the initial electron state and final electron state, E_i and E_f are the corresponding energy. ω_0 is the angular frequency of the emitted photon. By summing over all the photonic modes in the cavity, the spontaneous emission rate can be simplified to be [143]

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

with c the speed of light in vacuum, n the refractive index, ϵ_0 the permittivity of vacuum, and $z_{i \rightarrow f}$ the dipole matrix element defined as $z_{i \rightarrow f} = \left| \langle \psi_i | z | \psi_f \rangle \right|^2$.

In THz QCL, the typical spontaneous emission lifetime $\tau_{i \rightarrow f}^{sp}$, which is the inversion of the spontaneous emission rate, is microsecond level. This is much longer than the emission at short wavelengths, as the emission rate scales with ω_0^3 . Comparing to the picosecond non-radiative transition lifetime, such a long spontaneous lifetime indicates that spontaneous emission is very inefficient in a THz QCL. Therefore, it is very difficult to realize THz light emission diode based on the intersubband transition.

3.3.2.2 Stimulated emission

The stimulated emission rate can be calculated by considering the interaction of the dipole with the monochromatic incident light with a light intensity of $I(\nu)$ at frequency ν . The stimulated emission rate is given by [143]

$$W_{i \rightarrow f}^{st} = W_{i \rightarrow f}^{sp} \frac{\eta \lambda^2 I(\nu)}{8\pi \hbar \nu n^2} L(\nu) \quad (3-9)$$

with $\eta = \frac{3|z_{i \rightarrow f}|^2}{|\mathbf{r}_{i \rightarrow f}|^2}$ describing the interaction of the incident light with the dipole and a

lineshape function for homogeneously broadened transition expressed as

$$L(\nu) = \frac{\Delta\nu / 2\pi}{(\nu - \nu_0)^2 + (\Delta\nu / 2)^2} \quad (3-10)$$

where

$$\Delta\nu = \frac{1}{\pi T_2} = \frac{1}{\pi} \left(\frac{1}{2\tau_i} + \frac{1}{2\tau_f} + \frac{1}{T_2^*} \right) \quad (3-11)$$

is the full-width half maximum linewidth of the transition centered at ν_0 . In Eq. (3-11), T_2 is the total phase breaking time, τ_i and τ_f are the lifetimes of the initial electronic states and final electronic states and T_2^* is the pure dephasing time.

3.3.2.3 Intersubband optical gain

The optical power density with radiative transition between subbands i and f can be given by:

$$P = h\nu (N_i W_{i \rightarrow f}^{st} - N_f W_{f \rightarrow i}^{st}) = h\nu W_{i \rightarrow f}^{st} (N_i - N_f) \quad (3-12)$$

assuming $W_{i \rightarrow j}^{st} = W_{j \rightarrow i}^{st}$. If the population inversion is satisfied, i.e., $\Delta N = N_i - N_f > 0$,

the intensity of light will be amplified when propagating within the active region

$\frac{dI(\nu)}{dz} = g(\nu)I(\nu)$. Then the bulk gain coefficient per unit length can be given

by [143]

$$g(\nu) = \frac{\Delta N e^2 \pi \nu_0 |z_{i \rightarrow f}|^2}{\epsilon_0 n \hbar c} L(\nu) \quad (3-13)$$

where ΔN is three dimensional population inversion density which is related to the two dimensional population inversion density ΔN^{2D} as $\Delta N = \Delta N^{2D} / L_{\text{mod}}$, with L_{mod} being the length of an active region module.

3.3.3 Intersubband non-radiative transitions

Derived from steady-state rate equations of QCL, the population inversion between upper laser state (u) and lower laser state (l) can be expressed as

$$\Delta N = \frac{J_{\text{inj} \rightarrow u}}{e} \tau_u \left(1 - \frac{\tau_l}{\tau_{u \rightarrow l}} \right) \quad (3-14)$$

with τ_u being the lifetime of upper laser state, τ_l being the lifetime of lower laser state, $\tau_{u \rightarrow l}$ being the scattering time of an electron from upper laser state to lower laser state, and $J_{\text{inj} \rightarrow u}$ being the injected current density from injection region to the upper laser state. From this equation, it can be noticed that to realize population inversion and thus for optical gain for amplification, longer lifetime of upper laser state τ_u and scattering time $\tau_{u \rightarrow l}$ and shorter lifetime of lower laser state τ_l would be desirable. Therefore, a better understanding of the non-radiative relaxation mechanisms of the electron in the QCL system can provide strategies in the design of energy levels and wavefunctions to be engineered for a population inversion. The main intersubband non-radiative electron relaxation paths include electron-LO-phonon scattering, electron-electron scattering, acoustic-phonon scattering, ionized-impurity scattering and interface roughness scattering, as can be seen from Fig. 3-5(a). Comprehensive knowledge of these can be inferred from [145,146]. A brief introduction only on electron-LO-phonon scattering and electron-electron scattering will be made to shade light on the non-radiative electron scattering processes in this thesis.

Figure 3-5. (a) Schematic view of the typical intersubband relaxation processes. LO: electron-LO-phonon scattering; e-e: electron-electron scattering; ii: ionized impurity scattering; ac: acoustic scattering; ir: interface roughness scattering. (b) The electron-LO-phonon scattering between two subbands with initial electron wavevector k_i and final electron wavevector k_f , respectively. (c) The relationship between k_i , k_f and in-plane phonon wavevector $q_{||}$.

3.3.3.1 Electron-LO-phonon scattering

For subband separations greater than LO-phonon energy E_{LO} (~ 36 meV in THz QCL with AlGaAs/GaAs materials), the electron-LO-phonon scattering is the principal intersubband scattering mechanism, which occurs within subpicosecond. This seriously suppresses the state lifetime and therefore the energy separation between upper and lower laser states is designed to deviate from LO-phonon energy E_{LO} . However, this scheme is widely utilized to deplete the lower laser state [147]. For intersubband electron transitions with energy separation smaller than E_{LO} , the electron-LO-phonon scattering also plays an important role which takes place from the high energy tail of the upper laser state to the lower laser state. This process is trivial in low temperature, but turns to be a limiting factor to the upper laser state lifetime in high temperature environment.

For subbands i and f , the electron-LO-phonon scattering rate is given by [143]

$$W_{i \rightarrow f}^{abs} = \frac{m^* e^2 \omega_{LO}}{8\pi \hbar^2} \left(\frac{1}{\varepsilon_\infty} - \frac{1}{\varepsilon_{dc}} \right) n_{\omega_{LO}} \int_0^{2\pi} B_{i \rightarrow f}(q_{\parallel}) d\theta,$$

$$W_{i \rightarrow f}^{em} = \frac{m^* e^2 \omega_{LO}}{8\pi \hbar^2} \left(\frac{1}{\varepsilon_\infty} - \frac{1}{\varepsilon_{dc}} \right) (n_{\omega_{LO}} + 1) \int_0^{2\pi} B_{i \rightarrow f}(q_{\parallel}) d\theta \quad (3-15)$$

with ε_∞ and ε_{dc} being the high frequency permittivity and static permittivity, $n_{\omega_{LO}}$ being the Bose-Einstein occupation, and $B_{i \rightarrow f}(q_{\parallel})$ defined as

$$B_{i \rightarrow f}(q_{\parallel}) = \int_{-\infty}^{\infty} dz \int_{-\infty}^{\infty} dz' \psi_f^*(z) \psi_i(z) \psi_i^*(z') \psi_f(z') \frac{1}{q_{\parallel}} e^{-q_{\parallel}|z-z'|} \quad (3-16)$$

where $q_{\parallel} = |\mathbf{k}_i - \mathbf{k}_f| = \sqrt{k_i^2 + k_f^2 - 2k_i k_f \cos \theta}$ is the in-plane phonon wavevector with θ being the angle between initial and final wavevectors as shown in Fig. 3-5(c).

The total electron-LO-phonon scattering time between the subbands can be given by averaging over all the possible initial states in the subbands [143]

$$\tau_{i \rightarrow f} = \frac{\int f[k_i] dk_i}{\int f[k_i] W_{i \rightarrow f}^{LO}(k_i) dk_i} \quad (3-17)$$

with $f[k_i]$ being the distribution function of the in-plane wavevector.

3.3.3.2 Electron-electron scattering

Electron-electron scattering is an elastic scattering process, which is dominant for intersubband transitions between closely spaced subbands at low temperature where LO-phonon scattering is suppressed. It plays an important role in miniband electron transition by enhancing the inter-miniband scattering, which has sub-picosecond scattering time and was utilized in the first THz QCL to help depopulate the lower laser state [21]. Electron-electron scattering is a many-body problem and accurate calculation of scattering rates is sophisticated. Smet *et al.* [146] developed a single-subband-approximation model based on Fermi's golden rule to analyze the scattering. Considering an electron in subband i scattering with another electron in subband j , and then falling in to the subbands f and g respectively, the interaction Hamiltonian is given by [146]

$$H'_{i,j \rightarrow f,g}(\mathbf{k}_i, \mathbf{k}_j, \mathbf{k}_f, \mathbf{k}_g) = \frac{2\pi e^2}{S_{\parallel} \epsilon} A_{i,j \rightarrow f,g}(q_{\parallel}) \delta(\mathbf{k}_f + \mathbf{k}_g - \mathbf{k}_i - \mathbf{k}_j) \quad (3-18)$$

with $\mathbf{k}_i$, $\mathbf{k}_j$, $\mathbf{k}_f$, $\mathbf{k}_g$ being the wavevectors corresponding to different states, $q_{\parallel} = |\mathbf{k}_i - \mathbf{k}_f|$, and form factor of

$$A_{i,j \rightarrow f,g}(q_{\parallel}) = \int_{-\infty}^{\infty} dz \int_{-\infty}^{\infty} dz' \psi_f^*(z) \psi_i(z) \psi_g^*(z') \psi_j(z') e^{-q_{\parallel}|z-z'|} \quad (3-19)$$

Substituting Eq. (3-18) into the Fermi golden rule, the scattering rate of the electron in subband i is given by [146]

$$W_{i,j \rightarrow f,g}(\mathbf{k}_i) = \frac{e^4}{2\pi\hbar\epsilon^2} \int d^2\mathbf{k}_j \int d^2\mathbf{k}_f \int d^2\mathbf{k}_g \frac{|A_{i,j \rightarrow f,g}(q_{\parallel})|^2}{\epsilon_{sc}^2(q_{\parallel}, T) q_{\parallel}^2} f_{j,\mathbf{k}_j} (1 - f_{g,\mathbf{k}_g}) (1 - f_{f,\mathbf{k}_f}) \quad (3-20)$$

$$\times \delta(E_f(\mathbf{k}_f) + E_g(\mathbf{k}_g) - E_i(\mathbf{k}_i) - E_j(\mathbf{k}_j)) \delta(\mathbf{k}_f + \mathbf{k}_g - \mathbf{k}_i - \mathbf{k}_j)$$

where $\epsilon_{sc}^2(q_{\parallel}, T)$ is the effective dielectric constant and $f_{j,\mathbf{k}_j}$ is the possibility that the state j is occupied by an electron. Among various electron-electron scattering process, it was found that $22 \rightarrow 11$ and $22 \rightarrow 21$ processes (as shown in Fig. 3-5(a)) is the most efficient, with typical scattering time of ~ 100 ps.

3.3.4 Resonant tunneling transport

Resonant tunneling is one of the most important electron transport processes in the QCLs, which helps electron to transport through potential barriers. It is straightforward to have a qualitative understanding of the electron tunneling in the QCL mainly based on two tunneling models, i.e. coherent tunneling and incoherent tunneling.

When a certain external voltage is applied and two different energy states are brought into aligned in energy, resonances arise between these two energy states. Such interaction will split the aligned states into doublets with minimum energy separation of $\Delta_0 = |E_A - E_S|$, which is known as the anti-crossing gap representing the interaction strength between the two states. The resulting wavefunctions of the doublets are spatially extended across the calculated structure (Fig. 3-6(b)) and electron transport is considered to be coherent when crossing through the barriers. Carrier transport can be simplified as electrons scattered into and out of the doublets from the preceding state and into the subsequent state (Fig. 3-6(c)). Thus, with neglecting the interwell tunneling time due to the phase coherence, the current density is given by [143]

$$J = \frac{eN_A}{\tau_A} + \frac{eN_S}{\tau_S} \approx \frac{eN}{2\tau} \quad (3-21)$$

where $\tau^{-1} = \tau_A^{-1} + \tau_S^{-1}$ and $N_A \approx N_S = N/2$ for strong coupling (e.g., $\Delta_0 = 4-6$ meV) with N being the total electron density of the two states.

However, in the weak subband interaction regime (small Δ_0), such coherent-tunneling picture fails. In the weak coupling regime, the tunneling process is easily to get dephased through the ionized impurity scattering and interface roughness scattering. It is more suitable to use tight-bonding model to describe the process. The wavefunctions of two resonant states are considered to be localized within their individual quantum well. No energy splitting occurs and the coupling strength is $\Delta_0/2$. The interwell transport time of electrons is considered to be half of the Rabi period $\hbar/\Delta_0$. The current density can be given by [143]

$$J \approx \frac{eN}{2\tau} \left(1 - \frac{1}{1 + \left(\frac{\Delta_0}{\hbar} \right)^2 \tau \tau_{\parallel}} \right) \quad (3-22)$$

with $\tau_{\parallel}$ being the dephasing time. It can be found that a large Δ_0 and thus $(\Delta_0/\hbar)^2 \tau \tau_{\parallel} \gg 1$ will revive the coherent tunneling.

Figure 3-6. (a) Two energy states and their wavefunctions in adjacent quantum wells. (b) When quantum wells are suitably biased, the two energy states are aligned in energy and split into doubles with an energy gap of Δ_0 . This energy gap Δ_0 is known as anti-crossing gap, representing the interaction strength between the two states. (c) Coherent transport model for describing electron transport through the barrier with two energy states in strong coupling regime. The two energy states in resonance form doublets with an energy gap of Δ_0 . The wavefunctions are extended across the quantum wells. (d) Tight-binding model for electron transport with two energy states in weak coupling regime. The wavefunctions are localized within each quantum well with a coupling strength of $\Delta_0 / 2$.

3.4 Three-well resonant-phonon active region design

The THz QCL wafer utilized in this work is based on three-well resonant-phonon design, which is the most successful design for THz QCL so far. The conduction band diagrams are shown in Fig. 3-7 and the corresponding wafer growth sheet is shown in

Table 3-1. In this design, one module consists of two narrow quantum wells where radiative transition happens and one wide quantum well which works as an injector. After being injected from the injector of previous module via resonant tunneling, the electrons in the upper laser states will undergo radiative and non-radiative transitions to the lower laser states. The lower laser states are in resonance with the excited states of the adjacent wide quantum well. The excited states and fundamental injection state *inj* in the wide quantum well have an energy separation ~ 36 meV. Therefore, the electrons in the lower laser states can tunnel to the wide well and be quickly evacuated by the fundamental injection state *inj* through ultrafast electron-LO-phonon scattering.

Multiple advantageous of this design can be found comparing to the first THz QCL which is based on the Chirped lattice design [21]. As the upper laser states and lower laser states are localized in different narrow quantum wells, the optical transition is diagonal, leading to a longer upper state lifetime comparing to the vertical optical transition scheme in the first THz QCL as shown in Fig. 3-3(a). This also helps to enhance the injection efficiency, because the lower laser states have less spatial overlap with the injection states. The localization of upper laser states reduces the overlap with injection state *inj*, and thus minimizes the parasitic non-radiative relaxation. And a large energy separation between the injection state *inj* and the lower laser states *l* can help to reduce the thermal backfilling of the electron from the state *inj* to the lower laser state *l*.

Figure 3-7. Simplified conduction band diagrams and the moduli squared of wavefunctions for a typical three-well resonant-phonon active region design of the THz QCL. (Reprinted from [147])

Layer	Material	[nm]	Ratio	Doping (cm ⁻³)	Remark
1	GaAs	250			
2	AlGaAs	300	Al(15)Ga(85)As		Etch stop layer
3	GaAs	75		5.00E+18	Contact layer
Start of 218 repeat periods					
4S1	AlGaAs	5.1	Al(15)Ga(85)As		
5S1	GaAs	9			QCL doping
6S1	AlGaAs	2.4	Al(15)Ga(85)As		
7S1	GaAs	8.1			QCL doping
8S1	AlGaAs	4.6	Al(15)Ga(85)As		
9S1	GaAs	5.3			QCL doping
		5.7		5.00E+16	
		5.3			QCL doping
End of repeat periods					
10	AlGaAs	5.1	Al(15)Ga(85)As		
11	GaAs	50		5.00E+18	Contact layer

Table 3-1. Growth sheet of the three-well resonant-phonon active region design of the THz QCL.

3.5 Double-metal waveguide scheme

Under appropriate applied bias, the population inversion between two laser states will be achieved and therefore, the light will get amplified by stimulated emission. However, only stimulated emission is not sufficient for coherent laser radiation as there are many different optical losses that dampen the light intensity. There are three important sources of loss. Firstly, mirror loss or outcoupling loss, resulted from the semi-transparent mirrors or un-coated semiconductor facets, can be estimated by $\alpha_m = (1/L) \cdot (\ln(R))$, where R is the reflectivity of the mirror and L is the length of the cavity. The second important loss is the free-carrier absorption in doped semiconductor and metallic contact layers, which is often called waveguide loss α_w . The third source of loss is the resonant inter-subband transitions. Due to the considerable optical dipole matrix element of inter-miniband transitions, the extrinsic electrons in the injector region which undergo optical transitions resonant with the laser wavelength will cause significant absorption. This kind of loss can be suppressed during the design of QCL active region and injectors. It can be inferred from Eq. (3-10), Eq. (3-13) and Eq. (3-14), that the peak gain coefficient for a THz QCL is given by,

$$G(\nu_0) = \frac{2e\nu_0}{\epsilon_0 n \hbar c} \frac{\tau_u}{L_{\text{mod}}} \left(1 - \frac{\tau_l}{\tau_{u \rightarrow l}} \right) \frac{|z_{i \rightarrow f}|^2}{\Delta \nu} \text{ cm / kA} \quad (3-23)$$

Then, the threshold current density J_{th} of THz QCLs can be given by the equilibrium of gain and loss,

$$J_{th} = \frac{\alpha_m + \alpha_w}{G(\nu_0) \Gamma} \quad (3-24)$$

where Γ is the modal confinement factor with the active region.

From Eq. (3-24), it can be found that larger modal confinement factor, smaller mirror loss and material absorption are desirable to reduce the laser threshold and thus, a better laser performance. One of the most important steps achieved by R. Kohler *et al.*

to realize the first THz QCL was utilizing a novel waveguide design, which is called “semi-insulating surface-plasmon waveguide” [21]. By inserting a thin n^+ GaAs layer (~ 300 nm) between the active region and GaAs substrate, the waveguide enhances modal overlap ($\sim 47\%$) with the waveguide core, leading to a significant reduction of the free carrier absorption in the substrate. Another widely-used waveguide is double-metal (DM) waveguide [148], of which the active region (~ 10 μm) of the THz QCL is sandwiched by two metal plates, leading to near 100% modal confinement as shown in Fig.3-8(b). The free carrier absorption by GaAs substrate is completely avoided. Therefore a lower threshold and better temperature performance (higher maximum operation temperature) can be achieved. But the laser suffers from relatively low output power and extremely large beam divergence ($> 180^\circ$). This stems from the large impedance mismatch between the modes inside the DM waveguide and in free space due to the subwavelength confinement.

Figure 3-8. Schematic diagrams of (a) the semi-insulating surface-plasmon waveguide and (b) the double-metal (DM) waveguide used in THz QCLs. The electric field intensity (maroon curve) is plotted.

With regard to lasing in disordered semiconductor membrane structures, the most significant difference from conventional lasers is the huge cavity losses (conceptually

similar to the mirror loss of a conventional ridge laser). Due to the disordered structures and surface roughness induced by fabrication, the light propagating within the membrane gets more easily scattered into the vertical direction [149], especially into the substrate owing to smaller index contrast between the active region and substrate [20]. The gain materials used in short wavelengths, e.g. organic dyes, can achieve a peak gain coefficient in 100s or 1000s of cm^{-1} . Therefore, random lasing can be achieved even in the weakly scattering medium. However, such huge cavity losses (normally magnitudes larger than mirror loss of a conventional ridge laser), can hardly be compensated by the limited gain of the THz QCL (normally in the range of 30 – 40 cm^{-1}). In terms of structure fabrication, it is extremely challenging to create efficient scatterers on a THz quantum cascade wafer for such a long wavelength ($\sim 100 \mu\text{m}$ in free space) using traditional cleanroom processes. For example, to fabricate air hole scatterers on the QCL wafer as in Ref. [20], the AlGaAs/GaAs dry etching depth should be no less than 30 μm , which is quite challenging. Nevertheless, it is more practical to realize disorder structuring based on DM waveguide scheme which provides subwavelength ($\sim 10 \mu\text{m}$) confinement in the vertical direction. And in the following chapters it will be shown that the DM waveguide is a good platform for disordered laser applications.

3.6 Fabrication of double-metal waveguide ridge lasers

The fabrication processes of a DM waveguide ridge laser are summarized in Fig. 3-9. After the growth of a AlGaAs/GaAs heterogeneous quantum well superlattice by molecular beam epitaxy (MBE) onto a GaAs substrate, the THz QCL wafer, together with an n+ GaAs receptor wafer, were then cleaved into square pieces with size $\sim 8 \text{ mm} \times 8 \text{ mm}$. After cleaning the wafer surface, a Ti/Au (20 nm/600 nm) layer was deposited onto both QCL wafers and receptor wafers by electron-beam evaporator. The QCL wafers were put upside down onto the receptor wafers with crystal orientations of the wafer pairs carefully aligned to facilitate cleaving. Then, heat (300°C) and pressure ($\sim 5.4 \text{ Mpa}$) were applied to the wafer pairs by a commercial bonding machine for one

hour under the vacuum environment. After cooling down to the room temperature (RT), the substrate of QCL wafers was mechanically polished utilizing fine alumina powder to reduce the substrate thicknesses to $\sim 120 \mu\text{m}$. Then the remnant substrate was etched away by a solution of $\text{NH}_4\text{OH}:\text{H}_2\text{O}_2$ (1:19), which stopped at a $\sim 300\text{nm}$ $\text{Al}_{0.5}\text{Ga}_{0.5}\text{As}$ stop layer. Once the substrate was completely removed, the surface is smooth as mirror. Then the stop layer was removed by 49% HF solution.

The patterns of ridge laser were defined by a photolithography process using AZ5214 photoresist, which was followed by top contact (Ti/Au 15nm/300nm) deposition and a lift-off process. Then another photolithography process using S1818 photoresist was conducted to define wider strips to cover the metal strips and work as a mask for dry etching. The strips at this step are $\sim 10 \mu\text{m}$ wider than the metal strips, which is mainly used to suppress higher order lateral modes. The dry etching of the QCL active region was conducted by inductively coupled plasma (ICP) system with a gas ratio (in SCCM) of $\text{Ar}/\text{Cl}_2/\text{BCl}_3 = 5/5/5$ (units in SCCM).

Following the defining of laser ridges, the receptor substrate was lapped down to $\sim 120 \mu\text{m}$ by mechanical polishing using fine alumina powder. The thinner substrate facilitates chip cleaving and helps heat dissipation of the laser. Then a bottom contact layer (Ti/Au 15nm/300nm) was deposited. Finally, the fabricated chip was cleaved, indium soldered onto copper heatsink, wire-bonded, and finally mounted on the cold finger of a cryostat for characterization.

1. Au deposition for bonding

2. Wafer bonding

3. Substrate removal

4. Au deposition for top contact

5. Lithography for dry etching

6. Dry etching

7. Substrate polishing

8. Au deposition for back contact

9. Cleaving

GaAs substrate

Au

AZ5214

Stop layer

Active region

S1818

Figure 3-9. Schematic illustration of the fabrication flow for DM waveguide ridge THz QCL.

3.7 Experimental results of double-metal waveguide ridge lasers

The fabricated ridge lasers were driven by an electrical pulse generator with a repetition rate of 10 kHz and a pulse width of 500 ns. The emission spectra were characterized using a Fourier transform infrared spectrometer (FTIR, Bruker Vertex 70 series) with a room-temperature deuterated triglycine sulfate (DTGS) detector. The spectral scanning resolution is 0.5 cm^{-1} . As shown in Fig. 3-10, the threshold of laser at 10 K is around 1.33 kA/cm^2 and the highest working temperature is 145 K. The peak power is around 2 mW corrected by an estimated power collection efficiency of 30%. The performance of laser can be further enhanced by replacing gold with copper for metallic contacts, which has smaller free carrier losses in THz frequency range [150]. The dynamic range of the laser is large ($1.33 - 2.5 \text{ kA/cm}^2$), advantageous for lasing with very lossy cavities, e.g. disordered cavity. The typical emission spectrum of laser features multiple equally-spaced peaks with a free spectral range of $\sim 0.0208 \text{ THz}$.

According to the equation $\Delta f_{FSR} = \frac{c}{2n_{\text{eff}}L}$, where c is the light velocity in vacuum, L is

the length of cavity, the effective index n_{eff} of the DM waveguide is estimated to be 3.60. Due to almost 100% confinement of the mode by the double metal plates, the index of the THz QCL active region is indeed very close to 3.60. Thus, this value will be used as the refractive index approximation of the THz QCL active region in this thesis.

Figure 3-10. Emission characteristics of a DM waveguide THz QCL ridge laser. (a) The light-current-voltage (LIV) curve of the ridge laser with a dimension of $106\ \mu\text{m} \times 2\ \text{mm}$. (b) Emission spectrum of the laser at the rollover current density, measured at 10 K. The spectral resolution is $0.5\ \text{cm}^{-1}$.

During the emission spectra characterization, it was found that with increasing pumping current, the envelope of emission peaks ($T = 10\ \text{K}$) had a continuous blue-

shift as shown in Fig. 3-11. This phenomenon can be attributed to the Stark effect where the energy spacing between upper laser states and lower laser states becomes smaller with larger biased voltage. This is a typical property of diagonal transition scheme [121].

Figure 3-11. Emission spectra the THz QCL ridge laser at different pumping currents at 10 K.

3.8 Discussion and summary

In this chapter, the THz technologies, THz QCLs, and theoretical basis of the THz QCL have been reviewed sequentially, followed by an introduction of the three-well resonant-phonon design. The THz QCL wafer based on this design will be used in the following work. DM waveguide scheme is analyzed and found to be more suitable for lasing application with disordered feedback mechanism. To have a better understanding of this wafer, conventional ridge lasers based on DM waveguide have been fabricated and characterized. The ridge lasers achieve a large dynamic range and high temperature performance, indicating the advantages for disordered laser application which are

featured by larger cavity losses. The gain of the THz QCL wafer has a blue shift with increasing pumping current due to the Stark effect.

As highlighted in the introduction to the THz QCLs, it is promising to realize THz random emission by designing one/two-dimensional disordered structures with DM waveguide scheme for optical confinement in the vertical direction. Considering the limited gain of THz QCLs, efficient optical feedback is significantly necessary. For disordered structures, the transport mean free path, i.e. the scattering strength, is determined by the positional correlations and the scattering efficiency of individual scatterer. Therefore, the basic structure design strategy for efficient feedback is to incorporate beneficial structural order or to devise high-efficiency scatterers.

A typical property of random lasers with coherent feedback is the complex emission spectra with randomly distributed multiple peaks. These peaks correspond to different feedback paths within the disordered medium. Therefore, one of the major objectives is to realize multimode emission.. Moreover, multimode random emission is beneficial to reduce the spatial coherence, which enables potential application of speckle-free full-field imaging in the THz frequency range. Efficient trapping of light or strong localization of light enable more lasing modes in a disordered structure, which is advantageous for multimode random lasing. At the same time, it may pave the way for a research on the Anderson localization of light with the assistance of gain at such a long wavelength. As a result, it is of great significance to realize the localization of light by designing disordered structures with strong scattering property.

4 Terahertz Emission in One-Dimensional Disordered Systems

In this chapter, terahertz (THz) laser emissions around 3.2 THz from localized modes in one-dimensional (1-D) disordered grating systems are reported. The disordered structures are patterned on top of the double-metal (DM) waveguide of THz quantum cascade lasers (QCLs). Multiple emission peaks are observed near the bandgap region of the periodic counterparts. Numerical calculation indicates that the introduced disorder creates multiple microcavities near the bandgap frequency window of the underlying long-range order. Simulation results and experimental measurements provide strong evidence for spatial localization of THz wave.

4.1 Introduction

The scattering and diffusion of light in disordered/random media, have attracted considerable research interests for years due to intriguing physical phenomena. Particularly, as an analogy of Anderson localization in disordered electronic system [15], the spatial localization of light caused by structural disorder/randomness was investigated theoretically [27,28] and experimentally [38,40]. A wide range of applications based on this strong localization of light have been proposed, such as wavelength filters, optical switches, sensors [42] and cavity quantum electrodynamics [13]. The localization of light may also lead to important application of low-threshold laser with small figure print, which is prospective for sensing applications or active photonic micro-chips.

THz technology has been a highly topical research field in recent years. Notable achievements including the invention and advancement of THz QCLs [21,151,152], sensitive THz photodetectors [94,95], THz modulators [99,153] etc., make THz-frequency domain more accessible. THz QCLs are electrically-pumped semiconductor-

based lasers based on electronic transitions between the subbands of a quantum well superlattice [21]. Various one/two-dimensional structures including periodic structure [154], graded structure [155,156] and quasi-periodic structure [127,128] have been exploited in THz QCLs for specific functionalities. However, due to the long-range Bloch scattering feedback mechanism, the typical electromagnetic field distributions in these structures are extended. Purely random structures [20,157] are advantageous to realize the localization of light on the condition that the scattering efficiency of structures is strong enough. In the disordered medium, it is normally difficult to meet the Ioffe-Regel criterion for the localization of light [30,31]. However, in a photonic crystal structure with slight disorder, sharp Bragg peaks and photonic band gaps exist. Near the bandedge, a modified criterion $2\pi l_t / \lambda_{\text{crystal}} < 1$ was suggested for the wavelength because the standing wave is modulated by an envelope function of the crystal and it seems to be easily satisfied in the photonic bandedge [27,41]. It was also suggested that in the lower dimensional medium, it is easier to realize localization due to the reduced number of dimensions for efficient optical feedback [29]. Therefore, 1-D disordered structures are exploited to achieve the localization of light.

In this work, I report THz multimode lasing emission around 3.2 THz in 1-D disordered grating systems comprising of subwavelength aperture scatterers with average spacing of quarter-wavelength scale. A statistical study of disorder degree and average gain threshold of eigenmodes illustrates the localization mechanism. Spectra measurement and numerical analysis provide solid evidence of spatial localization of light.

Figure 4-1. Some photonic structures used in terahertz (THz) quantum cascade lasers (QCLs) (left column) and the typical field distributions of the lasing mode (right column): (a),(b) Photonic crystal [154]; (c), (d) one dimensional (1-D) graded photonic crystal [155]; (e), (f) 1-D photonic quasi-crystal based on Fibonacci sequence [127]; (g), (h) two-dimensional (2-D) photonic quasi-crystal [128]; (i),(g) 2-D random photonic structure with randomly distributed air holes [157].

4.2 Structure design

Our scheme for implementing 1-D disordered structure is illustrated in Fig. 4-2. A double-metal (DM) ridge waveguide comprising of 10 μm THz quantum cascade active material and a thin (~ 75 nm) heavily-doped contact layer was fabricated by conventional THz QCL process [148], which will be detailed later. Gratings were defined on the top metal plate with apertures opened for laser output. The width of waveguide and top metal plate are 100 μm and 90 μm respectively, leaving absorbing boundaries to suppress high-order lateral modes [124]. The total length of gratings is 2 mm, ending up with 100 μm wire bonding areas and long absorbing sections (> 500 μm) to eliminate the facet reflectivity on the two sides of the ridge waveguide. Therefore, the whole structure is a single-lateral-mode waveguide and the feedback is completely provided by the multiple scattering of gratings. The width of low-index apertures (A) and high-index DM region (B) are: $A(i) = a$ μm , $B(i) = (14-a)*(1+\delta)$ μm . Here, i is the period number, a is a constant value, δ is a random number uniformly distributed between $-\Delta d$ and Δd , where Δd represents the disorder degree.

Figure 4-2. 3-D schematic diagram of the THz QCL with 1-D disordered gratings. Inset: top view of the sketched device (top-left corner); scanning electron

microscope (SEM) image of the fabricated QCL with disordered gratings with aperture width of 3.5 μm and disorder degree of 0.2 (bottom-right corner).

QCLs are electrically pumped, and emit in the transverse-magnetic (TM) polarization due to inter-subband selection rules [111]. For THz QCLs with double-metal ridge waveguide, the subwavelength vertical confinement of the TM mode, leads to an impedance mismatch in the areas with and without metal coverage [123]. The effective refractive index contrast of the gratings is roughly estimated to be 2.52 : 3.53, by calculating the electric-field distributions of the fundamental propagating mode in the waveguide at 3.2 THz.

For disordered structures, statistical studies are usually adopted to extract the ensemble-averaged wave propagation properties [158–161]. In this work, for a qualitative understanding of the disorder influence on the photonic bandgap (PBG), I firstly use finite-difference time-domain (FDTD) method to calculate the ensemble-averaged transmission spectra for waveguides with disorder degrees ranging from $\Delta d = 0$ to $\Delta d = 0.4$. In the simulation, the 3-D disordered waveguide structure was simplified to be 1-D grating using estimated refractive index contrast, without considering waveguide losses. The transmission spectra were averaged over 100 different configurations at each disorder degree. As shown in Fig. 4-3(a), for the periodic structure, a clear PBG ($\Delta\omega/\omega_0 \sim 0.12$) exists. When disorder is introduced, it can be observed that the transmission gap gets narrower and shallower, while the transmission at passband decreases. Multiple narrow transmission peaks (for the spectrum of $\Delta d = 0.2$) arise within the original PBG range, which are related to resonant modes induced by the disorder. At this disorder degree, a clear transmission gap still persists, indicating the underlying long-range order. When disorder degree gets larger (e.g $\Delta d = 0.3$ or 0.4), the long-range order is even more strongly destroyed by the introduced disorder with a shallow transmission dip filled up with resonant peaks. It can be inferred that the transmission spectra will get flat at the whole frequency range for sufficient disorder degree (say, $\Delta d = 0.5$ or 0.6) when the underlying long-range order is completely disrupted.

Figure 4-3. (a) Ensemble-averaged transmission spectrum over 100 different configurations of 1-D disordered gratings for each disorder degree. (b) Ensemble-averaged gain threshold. For $\Delta d = 0$, the peak at $\Delta\omega/\omega_0 = -0.15$ is induced by the resonance of wire bonding areas. This resonance is disrupted and therefore disappears when disorder is introduced. The free carrier loss of active material was not considered.

Theoretical work has been conducted for a better understanding of the interplay of localization and amplification in disordered or random media [48,162]. As in the conventional laser system, the lasing threshold of the disordered or random system is determined by the eigenmode which experiences the largest net gain. Although statistical study over light propagation and resonances in the disordered gratings has been carried out, the analytical results may not decide essential eigenmode properties for the investigated particular structures, which have subwavelength vertical confinement, vertical out-coupling channels and wire-bonding pads. To have a better understanding of the eigenmode properties, finite-element method (FEM) simulations have been performed for the 1-D grating systems using commercial software Comsol Multiphysics. As full-structure 3-D simulation is time- and memory-consuming, the simulation was simplified to 2-D calculation of the structural cross sections where the x-z plane was considered, while the grating was assumed to be infinite in the y direction. The losses of the active region material were not considered. From the calculated complex eigenfrequency $\omega_r + i\omega_i$, the gain threshold to compensate the

eigenmode loss can be extracted by $2n_{eff}\omega_i/c$, where n_{eff} is the modal effective refractive index, c is vacuum velocity of light. The ensemble-averaged gain threshold was calculated from 20 different configurations at each disorder degree, taking the mean gain threshold at each frequency bin.

As shown in Fig. 4-3(b), with disorder degree increasing from $\Delta d = 0.1$ to 0.4, the optical gap gets narrower and the ensemble-averaged gain threshold increases continuously. In the pass band, the ensemble-averaged gain threshold decreases firstly and then fluctuates around a certain level. When $\Delta d = 0.1$, the long-range Bloch scattering effect dominates the feedback since the optical gap obviously exists and the eigenmodes in the pass band obey the overall trend of the eigenmodes in the periodic structure. Inside the underlying PBG, weak disorder-induced low-loss modes appear. These low-loss modes are exponentially localized inside the structures. However, the electric profiles of eigenmodes in the passband are all extended. In the structure with larger disorder degree ($\Delta d = 0.2$), more localized modes appear due to higher density of optical states inside the underlying PBG; meanwhile the loss is slightly higher due to stronger disorder perturbation. When disorder gets strong (e.g. $\Delta d = 0.4$), the random scattering effect gradually dominates the feedback mechanism. The electric profiles of the eigenmodes outside original PBG are featured by large, random amplitude fluctuations [161] and the average gain threshold decreases to around 10 cm^{-1} . Inside the underlying PBG, even more eigenmodes appear and the average gain threshold increases such that the dip induced by long-range effect is trivial. The optical localization lengths, $L_e = \left(\int \text{abs}(E_z) dx \right)^2 / \int (E_z)^2 dx$, have been estimated for the localized eigenmodes inside the underlying PBG. For disorder level of $\Delta d = 0.1$ and 0.2, the optical localization lengths are normally in the range from 300 μm to 450 μm , much smaller than the system size. The optical localization lengths almost double when larger disorder degree, e.g. $\Delta d = 0.4$, is introduced. This indicates a smaller disorder degree will be more beneficial for optical localizations. However, the optical density (number of eigenmodes) inside the underlying PBG region can be enhanced by a larger disorder degree. Therefore, disordered grating structures with $\Delta d = 0.2$ are chosen for

experimental demonstrations, a compromise between the optical density and the gain threshold, localization lengths of eigenmodes.

4.3 Device fabrication and characterization

The fabrication processes started with conventional DM ridge laser processes, i.e. Au deposition on QCL wafers and receptor wafers for bonding, wafer bonding, and QCL wafer substrate removal. After these, a photolithography process was conducted with AZ5214 to pattern strips with embedded disordered gratings on QCL active region, which was followed by top contact (Ti/Au 20 nm/200nm) deposition and a lift-off process. A layer of SiO₂ (500 nm) was deposited by PECVD which worked as a hard mask in the following inductively coupled plasma (ICP) dry etching process. Then another photolithography process utilizing S1818 was conducted to define wider strips to cover the metal strips and gratings. The strips at this step are ~ 10 μm wider than the metal strips, which is employed to suppress higher order lateral modes. The hard mask of SiO₂ was defined by a reactive ion etching (RIE) process. Then, the ridges were formed through dry etching of the QCL active region, which was conducted by ICP system with a gas ratio (in SCCM) of Ar/Cl₂/BCl₃ = 5/5/5. Finally, the receptor substrate was lapped down to ~ 120 μm by mechanical polishing using fine alumina powder and a bottom contact layer (Ti/Au 15nm/300nm) was deposited. The fabricated chip was cleaved, indium soldered onto copper heatsink, wire-bonded, and finally mounted on the cold finger of a cryostat for characterization.

The emission spectra were characterized using a Fourier transform infrared spectrometer (FTIR, Bruker Vertex 70 series) with a room-temperature deuterated triglycine sulfate (DTGS) detector. The scanning resolution was set to be 0.2 cm⁻¹. The devices were driven by an electrical pulse generator with a repetition rate of 10 kHz and a pulse width of 500 ns.

4.4 Experimental results and discussion

4.4.1 Light-current-voltage and emission spectrum characterization

The THz QCL active region used in this work was based on three-well resonant-phonon design with GaAs/Al_{0.15}Ga_{0.85}As heterostructure active layers, whose gain spectrum center continuously shifts from 3.05 THz to 3.20 THz with increasing pumping intensity as shown in Fig. 3-11. As the gain spectral range is smaller than the bandgap size of periodic structure, the lower bandedge was located to be within this frequency regime. The fabricated QCLs have 1-D gratings with three typical disorder degrees, i.e. perfectly periodic distribution ($\Delta d = 0$), moderate disordered ($\Delta d = 0.2$) distribution and highly-disordered ($\Delta d = 0.6$) distribution.

Fig. 4-4 shows the light-current-voltage (LIV) curves and the corresponding emission spectra of the QCLs with 1-D periodic grating and disordered grating ($\Delta d = 0.2$), respectively. The threshold of periodic grating laser is around 1.58 kA/cm² at 10 K. A stable single mode was observed at 3.07 THz for the whole dynamic range, even though the gain spectrum center shifts with increasing pumping current. This indicates the position of lower band edge. For 1-D disordered QCL with $\Delta d = 0.2$, the threshold at 10 K is 1.79 kA/cm², slightly higher than that of periodic counterpart. More than 8 peaks arise in its emission spectrum with frequency located inside the PBG of the underlying periodic gratings (the peak frequencies and the corresponding thresholds are listed in Table.4-1). It can be found that multiple peaks appear almost simultaneously, indicating the corresponding eigenmode losses are of the same level. The emission peaks should correspond to low-loss localized modes of the disordered grating system. For the QCL with highly-disordered gratings (not shown in this thesis), the threshold is much higher and the number of peaks is limited (normally 2 peaks). This is attributed to larger localization lengths and much higher scattering losses of stochastic distributed gratings. From the simulation results, for QCLs with moderate disorder degree, the lasing preferentially occurs at wavelengths within the bandgap of periodic counterparts due to the smallest gain threshold and tightest modal confinement in the center of the

system; for QCLs with large disorder degree, the optical modes with largest gain coefficient and comparably smaller optical losses will be selected to lase.

Figure 4-4. (a) The light-current-voltage (LIV) curve and (b) emission spectrum at 4.26 A of the fabricated 1-D periodic QCL. (c) The LIV curve and (d) emission spectra of the fabricated 1-D disordered QCL with $\Delta d = 0.2$. The emission spectra were measured at heat-sink temperature of 10 K with a resolution of 0.2 cm⁻¹. The aperture width of devices is 3.5 μm . The green line is vertically shifted by 0.2 for clarity.

Frequency (THz)	3.123	3.131	3.145	3.160	3.181	3.204	3.279	3.315
Threshold (A)	3.624	3.716	3.812	3.624	4.305	4.421	3.656	4.069

Table 4-1. The peak frequencies and the corresponding lasing thresholds corresponding to the spectra in Fig. 4-4(d).

4.4.2 Calculated field distributions for emission modes

The emission spectra with multiple peaks can always be observed for different disordered grating structures with $\Delta d = 0.2$. The field distributions have been calculated for another disordered laser with its emission spectra shown in Fig. 4-5(a). The calculation was done for the 2-D cross-section plane (x-z plane, as shown in Fig.4-2) and top-view plane (x-y plane within the active region, as shown in Fig.4-2) of the structure, respectively. For 2-D calculations of the structural cross section (x-z plane), the field distribution along the z-direction is uniform in the DM waveguide (as shown in Fig. 3-7(b)). Therefore, the 2-D field distributions can be embodied by the intensity curves within the active region along the x direction. In both sets of calculations, four eigenmode field distributions with the lowest gain threshold are plotted in Fig. 4-5(b). A good match between the emission frequency and eigenmode frequency can be found. Even though the calculation models are the simplification of 3-D actual laser structure, it can be concluded that the emission modes correspond to the localized eigenmodes with the lowest gain thresholds.

Figure 4-5. (a) The emission spectrum of another 1-D disordered QCL with $\Delta d = 0.2$ at pumping current of 4.44 A. The peaks are labeled by the frequency values. (b) The calculated mode distributions (red curves) corresponding to the four peaks of the laser. The simulation was simplified to 2-D calculation of the structural cross sections where the x-z plane was considered, while the grating was assumed to be infinite in the y direction. Inset: field distribution snapshots of the top-view structure calculation (x-y plane within the active region, as shown in Fig. 4-2). In the calculation, the waveguide losses were not considered.

4.4.3 Indirect proof for the localization of light

Near-field scanning optical microscopy (NSOM) can be used for direct observation of the mode profile. Unfortunately, it normally cannot be used at very low temperatures unless a special cryocooling system is designed [163]. I don't have the access to low temperature NSOM system in our group, thus a simpler method is proposed to further verify the localization of light.

Figure 4-6. The emission spectra of a 1-D disordered QCL without (red curve) and with (green curve) partial covering by a copper sheet. The pumping currents are (a) 3.90 A, (b) 4.09 A. All the peaks are labeled by the frequency values.

Experimental evidence for optical localization is acquired by partially covering the surface of the disordered laser device (see Fig. 4-4(c) and (d)) with a copper sheet for emission spectrum measurement. The copper sheet was fixed at a distance of ~ 1 mm above the device surface, and half of the device was covered. Emission spectra were then compared without (red curve) and with (green curve) the copper sheet in place, with the position of the device and the driving current remaining fixed between the measurements. It can be found that the emission from 3.133 THz and 3.283 THz was completely blocked by the copper sheet, indicating these modes are located at the covered region. The intensity of emission peak at 3.146 THz was reduced, implying that the emission was partially blocked. However, for the peaks at 3.127 THz and 3.164 THz, the intensity got no obvious change with the copper covering. Therefore, these modes are located at the uncovered region. The measured spectra at different current have similar phenomena, indicating the stability of the field distributions. When the copper sheet is removed, all the peaks recovered to original intensity level, confirming the reliability of measurement. From the above observations, we may conclude that the emission modes of 1-D disordered QCL are localized.

Recently, the strong localization of THz surface plasmon polariton (SPP) wave on a copper foil with disordered gratings was observed [164]. However, this is a passive structure. As the optical modes in THz QCL cavity are essentially SPP modes and the index contrast of disordered gratings is much higher than that in Ref. [164], principally it is easier to realize the localization of light in our devices.

4.5 Summary

In conclusion, 1-D THz disordered gratings QCLs with localized modes have been developed. Statistical numerical results show that the PBG gradually vanishes and random multiple scattering arises when disorder degree increases. Multimode localization can be supported by disordered gratings with moderate disorder degree. Modal localization in disordered system with such disorder degree is evidenced by the numerical simulation and experimental analyses.

5 Multimode Localized Random Lasing in Amorphous Lattices

In this chapter, devices consisting of randomly-distributed pillars fabricated from a terahertz (THz) quantum cascade gain medium are studied, and shown that such structures can achieve transverse-magnetic (TM) polarized multimode random lasing, with strongly localized modes at THz frequencies. The weak short-range order induced by the pillar distribution is sufficient to ensure high quality-factor modes that have a large overlap with the active material. Furthermore, the emission spectrum can be easily tuned by tailoring the size and filling fraction (FF) of the scatterers. These ‘designer’ random lasers, realized using standard photolithography techniques, provide a promising platform for investigating disordered photonics with pre-designed randomness in the THz frequency range, and may have potential applications such as speckle-free imaging.

5.1 Introduction

In conventional laser cavities, randomness is usually undesirable because it distorts the optical modes and leads to additional cavity losses. However, it has been recognized that a special class of laser, random lasers [16], can exhibit interesting and potentially useful properties. In random lasers, light is confined through multiple scattering and interference in a randomly-arranged scattering medium. Lasing takes place when the gain experienced by one or more of the complex modes overcomes the losses. The resulting multimode lasing emission spectrum has many potential applications. For example, it has been suggested that randomly-arranged nanoparticle clusters can be used for “fingerprinting” applications in document encoding and material labeling [17]. Multimode random lasers can also be employed as light sources for speckle-free imaging applications, owing to the low spatial coherence of an ensemble of

uncorrelated wavefronts [88,89]. Other possible applications can be found in display lighting, tumor detection [86] and sensing [165].

To date, random lasing has been achieved in a wide range of media, including nanocrystalline ZnO powers [56,81], ceramics [75], organic composites [166] and nanoparticles (e.g., TiO_2 , ZnO and GaN) dispersed in fluorescent dyes [16,167–170]. The lasing frequencies have been demonstrated to range from the near-ultraviolet to the mid-infrared (mid-IR) [20]. At lower frequencies, THz quantum cascade lasers (QCLs) based on quasi-periodic structures have been developed [127,128] and a hyperuniform disordered laser has been demonstrated [171]. THz lasing in purely random structures with air holes has also been realized [157], however, the lasing modes are extended due to the weak scattering efficiency of scatterers. By fabricating disordered gratings with moderate disorder degree on the top contact of THz QCL ridge lasers in the previous work, it has been demonstrated that the localization of light can be supported by the created microcavities near the bandgap frequency window of the underlying long-range order. However, starting from completely random structures with inclusion of high-efficiency scatterers and short-range order [23,24,149] is also applicable to create multiple microcavities for multimode random lasing with excellent performances.

Here, I report on the development of surface-emitting, multimode THz random QCLs comprising dielectric pillars arranged in a two-dimensional (2-D) amorphous lattice [172]. These QCLs are electrically pumped, and emit in the TM polarization due to inter-subband selection rules [111]. Compared to air hole scatterers [20,157], isolated dielectric pillars have superior scattering efficiency for TM polarized light [173]. In our structure, this leads to a substantial quality (Q) factor enhancement of localized modes near the edge of the TM gap. The pillars are formed by dry etching the quantum cascade active material, and serve not only as in-plane scatterers but also as the gain medium. Strongly localized multimode random lasing was observed at around 3.3 THz, with more than 13 lasing modes. Furthermore, I demonstrate that the lasing frequency can be fine-tuned by modifying structural parameters such as the pillar radius and the lattice FF.

5.2 Structural design of the two-dimensional terahertz random laser

Previously, a mid-IR random lasing was demonstrated in QCLs at $\sim 10\ \mu\text{m}$ by Liang *et al.*, through drilling randomly-distributed circular holes in a quantum cascade laser active region [20]. Nevertheless, this design was hampered by the weak scattering efficiency of air holes for TM modes, which limited the number of lasing modes to three. Sebastian *et al.* demonstrated THz random lasing using similar air hole structures [157]. However, the role of air holes was concealed by the highly reflective circular boundary and the emission modes are extended. Such a scheme is therefore unsuitable for a QCL in the THz frequency range, where the gain coefficient is much lower than that of mid-IR QCLs. Instead, isolated dielectric pillars provide much stronger scattering efficiency than air holes for TM polarized light. Previous studies show that a strong bandgap can be supported by photonic lattices formed by isolated pillars [171,174,175]. Since the electrical pumping requires direct electrical contact to the pillars, I adopt a double-metal waveguide design, enclosing the pillars between parallel metal layers. This waveguide is commonly employed for THz QCLs, achieving near-unity vertical optical confinement, owing to the low losses of metal at THz frequencies [148,176].

The device design is shown in Fig. 5-1. Circular quantum cascade (QC) pillars, with radii of a few microns and $10\ \mu\text{m}$ height, are randomly distributed in a 2-D lattice. The pillars were formed through inductively coupled plasma (ICP) etching of the THz QCL active region, and embedded in benzocyclobutene (BCB) [177–180] for mechanical support. BCB is a non-conducting resin with good planarization property, low losses at THz frequencies, excellent thermo-stability, and low refractive index. The 2-D scattering medium (formed by the pillars and BCB) is sandwiched between two thin layers of Ti/Au, which are used for current injection as well as vertical confinement. To allow for surface (i.e. vertical) emission, micro-apertures of $1.5\ \mu\text{m}$ radius were etched into the top plate, at the center position of each pillar. Fig. 5-1(b)

and (c) show scanning electron microscope images of the QC pillars after ICP dry etching, and of the finished device, respectively. The refractive index contrast in the scattering medium is approximately 3.6 (GaAs): 1.58 (BCB), which is higher than in many other random laser systems [86,167,181,182], allowing for stronger in-plane mode confinement. As the size of the output aperture on top of each pillar is in deep subwavelength regime and the lasing modes are largely anti-symmetric with respect to the apertures, only a small fraction of the light is coupled out from the laser cavity (see **Section 5.5.2** for details). By 3-D finite-element method (FEM) simulations using Comsol Multiphysics, the vertical Q factor corresponding to the surface emission losses was estimated to be several thousand, which is relatively high as compared to in-plane Q factors including the consideration of material losses as discussed below. Thus, in the following theoretical analysis, 2-D simplified models are used to approximate 3-D full-structure simulations which are storage- and time-consuming.

Figure 5-1. Terahertz (THz) random laser with two-dimensional (2-D) amorphous pillar lattice. (a) 3-D schematic diagram of a surface-emitting terahertz random quantum cascade laser. (b) Scanning electron microscope (SEM) images of the quantum cascade (QC) pillars at an oblique angle after inductively coupled plasma (ICP) dry etching, and (c) the surface of the finished device.

Figure 5-2. Spectral and spatial analysis of the random structures. (a) Simulated power spectra (vertically offset for clarity) of 2-D structures. The black and red curves correspond to minimum edge separations of $\Delta = 0 \mu\text{m}$ and $\Delta = 2.5 \mu\text{m}$, respectively. The refractive index contrast is approximately 3.6 (QC pillar): 1.58 (BCB). (b) Ensemble averaged spatial autocorrelation function $C(\Delta r)$ for structures with $\Delta = 2.5 \mu\text{m}$; the black dashed curve is an exponential fit. The inset shows the spatial Fourier transform of the structure, where the scale bar is $2\pi/a$; a is the average center-to-center distance between the pillars. (c) Power spectrum (red curve) and modal in-plane quality (Q) factors (purple dots) in the vicinity of the gap. The frequency region 3.34–3.55 THz has been omitted to show the Q factors at the edges of the gap clearly. Material losses are neglected. The modes highlighted by the blue ellipses correspond to the intensity distribution plots in Fig. 5-3(a). (d) Modal Q factors (blue triangles) with the BCB loss of $\sim 10.9 \text{ cm}^{-1}$ taken into account. The power spectrum is overlaid as a guide to the eye.

The positions of the QC pillars are determined by a random placement algorithm, subject to the constraint of a ‘minimum center-to-center distance’: $|P_i - P_j| \geq 2r + \Delta$, where P_i and P_j are the center positions of any two pillars, r is the radius of the pillar, and Δ is the minimum edge-to-edge separation between pillars (e.g. $\Delta = 0$ means each pillar is allowed to touch another one but without overlap). The pillars are placed sequentially, with any subsequent placement rejected if it violates this constraint. The constraint introduces weak short-range order into the random structure. To qualitatively show the effects of the short-range order, the power spectra of structures with different Δ were compared. A substantial gap in the power spectrum can be found for the structure with larger Δ , as shown in Fig. 5-2(a). The short-range order can be further enhanced for a wider bandgap by using more sophisticated placement algorithms, such as molecular dynamics-based close-packing; however, even with the present simple algorithm, the intensity in the power spectrum spans several orders of magnitude near the gap. This can be attributed to the scattering efficiency of the dielectric pillars.

The power spectra in Fig. 5-2(a) were calculated using 2-D finite-difference time-domain (FDTD) simulations, with pillars of 6.4 μm radius distributed within a 1000 $\mu\text{m} \times 1000 \mu\text{m}$ area; the total FF is 36.5%. In the simulations, randomly distributed multiple TM polarized electric dipoles are used to inject Gaussian pulses into the structure. Time evolutions of the electric field at different positions are recorded and summed before being Fourier transformed. The frequency range spans from 2 THz – 5.5 THz. For $\Delta = 0 \mu\text{m}$ and $\Delta = 2.5 \mu\text{m}$, dense peaks clustering around 3.3 THz are observed, corresponding to multiple resonant modes. For the $\Delta = 2.5 \mu\text{m}$ case (vertically shifted down by 10^3 a.u. for clarity), there is, however, a prominent spectral bandgap. By contrast, for an air-hole structure (without pillars), the power spectrum (vertically shifted down by 10^6 a.u. for clarity) is uniform with small random peaks over a broad spectral range. As a further probe of the structure’s short-range order, I calculated the spatial autocorrelation function of the ensemble, $C(\Delta r) = \langle \Theta(r)\Theta(r + \Delta r) \rangle / \langle \Theta(r) \rangle^2 - 1$, averaged over 100 independent configurations (For this calculation, I take $\Theta = 1$ inside the rods, and $\Theta = 0$ outside.). The result shown

in Fig. 5-2(b) reveals that an estimated correlation length L_c ($\sim 0.3-0.4a$), obtained from an exponential fit $\exp(-\Delta r / L_c)$ of the autocorrelation curve, is smaller than a (the average center-to-center distance of the pillars). This indicates that the short-range order is fairly weak, compared to amorphous structures such as that in Ref. [23]. Likewise, the power Fourier spectrum of the structure geometry, shown in the inset of Fig. 5-2(b), displays a weak isotropic scattering ring around the spatial frequency $2\pi/a$.

To determine the in-plane modal Q factors, 2-D FEM simulations were performed. Note that the metallic loss of the double metal waveguide and the vertical surface emission loss from apertures are not considered for this Q factor calculation. Fig. 5-2(c) shows the in-plane Q factor of the mode in a typical structure, overlaid on the power spectrum near the gap. With the BCB loss neglected, it can be found that a large number of modes with Q factors as high as 10^6 (indicating strong in-plane optical confinement) on either side of the gap exist. If a realistic level of BCB loss is included ($\sim 11 \text{ cm}^{-1}$), the Q factors are substantially reduced; however, as shown in Fig. 5-2(d), the band edge modes at lower frequencies of the bandgap still retain relatively high Q factors. Note also that the Q factors just below the gap ($Q \sim 1300$) are much larger than those above the gap ($Q < 300$), which can be attributed to the different spatial overlap of the modes with the BCB region, as discussed below.

Figure 5-3. Optical mode distributions of the random structures. (a) Field intensity distributions of the modes highlighted by the blue ellipses in Fig. 5-2(c). The mode intensities have been normalized for clarity. All modes are localized. The field intensity distributions of the two modes at 3.33 THz and 3.56 THz are amplified and shown as insets. As indicated in the insets, the modes near the bottom of the gap (at 3.33 THz) are mainly concentrated in the pillars, while modes near the top of the gap (at 3.56 THz) are mainly concentrated in the BCB. (b) Overlap factor and in-plane Q factors, with BCB losses accounted for, averaged over 100 modes with field energy mainly confined in the QC pillars for different FFs. The pillar radius is 6.4 μm , and the total device area is 1000 $\mu\text{m} \times 1000 \mu\text{m}$. Squares indicate structures with $\Delta = 0 \mu\text{m}$, and circles indicate structures with $\Delta = 2.5 \mu\text{m}$.

Fig. 5-3(a) shows the field intensity distributions of a selection of random modes on each side of the gap (the corresponding modes are highlighted by the blue ellipses in Fig. 5-2(c)). It is noteworthy that the modes at the lower edge of the gap match well with the main emission peaks which will be shown in the results section. Due to the strong scattering efficiency, the short-range ordered amorphous structures demonstrated here support multiple localized random lasing modes, some with nearly diffraction-limited localization lengths (see **Section 5.5.1** for details). These strongly localized modes are beneficial for emission with low spatial coherence and the realization of low-threshold lasing. Furthermore, there is a clear distinction between the

modes below and above the gap: the modes below the gap are mainly concentrated in the pillars (e.g. at 3.33 THz), while the modes above the gap are mainly located in the BCB (e.g. at 3.56 THz). This explains why the latter have much lower in-plane Q factors, as seen in Fig. 5-2(d). The high-Q modes below the gap are thus more favorable for lasing, and we shall see below that this frequency region indeed matches the distribution of the experimentally-observed emission peaks.

Comparing to the structure with $\Delta = 0 \text{ }\mu\text{m}$, the presence of a non-zero separation $\Delta = 2.5 \text{ }\mu\text{m}$ between pillar edges makes the structure more favorable for lasing. It creates a gap in the power spectrum, with more pronounced Q-factor enhancement at the band edge, as well as a larger overlap of the modes with the active region (and thus higher net gain). This can be quantified using an “overlap factor”, defined as the fraction of the field energy confined in the QC pillars:

$$\Gamma = \frac{\int_{xy=AR} \varepsilon(x, y) I(x, y) dx dy}{\int \varepsilon(x, y) I(x, y) dx dy} \quad (5-1)$$

where “AR” denotes active region, $\varepsilon(x, y)$ is the dielectric constant and $I(x, y)$ is the intensity distribution. Fig. 5-3(b) shows the overlap factor versus FF for $\Delta = 0 \text{ }\mu\text{m}$ and $\Delta = 2.5 \text{ }\mu\text{m}$, where each data point is averaged over 100 modes at the band edge below the gap. As expected, the overlap factors are significantly higher for the $\Delta = 2.5 \text{ }\mu\text{m}$ structure. The parameter $\Delta = 2.5 \text{ }\mu\text{m}$ further restricts the movement of the pillars so as to introduce a perceptible short-range order into the structure, leading to the opening of the bandgap and the enhanced Q factors at the band edges. This makes the maximum of field tend to be better confined inside the pillars for the lower band edge modes (see Fig. 5-2(d) and **Section 5.5.1** for details), which is the reason for larger overlap factors and higher Q factors.

5.3 Device fabrication and characterization

The schematic figure for the fabrication processes of the developed THz random QCLs are shown in the Fig. 5-4. To fabricate the random THz QCLs, Ti/Au was firstly

deposited both on top of the QCL structure and onto an n^+ doped GaAs host substrate; this was followed by Au/Au thermo-compression wafer bonding. The QCL wafer was then polished and selectively wet-etched to remove the substrate down to an etch-stop layer. Then the etch-stop layer was removed by HF solution. Through conventional photolithography and ICP dry etching of the active region, with gas ratio (in SCCM) of $\text{Ar}/\text{Cl}_2/\text{BCl}_3 = 5/5/5$, the QCL active region was etched into pillars of the intended radius (ranging from 6–8 μm) and 10 μm height. The structure was next spin-coated with BCB (CYCLOTENE 3022-46) until the top surface was planar; the detailed BCB planarization process was the same as in Ref. [178]. The BCB was then etched by reactive ion etching (RIE) with a gas ratio of $\text{O}_2/\text{CF}_4 = 25/100$ (SCCM) to expose the top surface of the QC pillars. Photolithography using AZ5214 was conducted to define the pumping area and extraction holes on top of the pillars, followed by a 20/600 nm Ti/Au layer deposition and lift-off. After this, a $\text{BCl}_3/\text{CH}_4/\text{Cl}_2$ RIE plasma mixture with ratio of 100/20/5 (SCCM) was used for 1 min to remove the highly-doped contact layer in the holes. This prevents the otherwise strong absorption of THz radiation, allowing it to be coupled out. The host substrate was then lapped down to around 180 μm and a 15/200 nm Ti/Au layer was deposited as bottom contact. Finally, the sample were cleaved, indium-soldered onto copper submounts, wire-bonded and mounted on the cold finger of a cryostat for characterization.

Figure 5-4. The fabrication processes for a THz random quantum cascade laser (QCL).

As in the previous work, the fabricated 2-D THz random QCLs were characterized using a Fourier transform infrared spectrometer (FTIR, Bruker Vertex 70 series) with a room-temperature deuterated triglycine sulfate (DTGS) detector. The scanning resolution was set to be 0.2 cm^{-1} . The devices were driven by an electrical pulse generator with a repetition rate of 10 kHz and a pulse width of 500 ns.

5.4 Experimental results

5.4.1 Light-current-voltage and emission spectrum characterization

The THz QCL wafer used here was based on a three-well resonant-phonon GaAs/Al_{0.15}Ga_{0.85}As design as introduced in **Section 3.5**, with gain centered around 3.1 THz. Based on the random QCL design described above, I fabricated a variety of structures with different pillar sizes and FFs. The experimental results of conventional ridge lasers (width 106 μm and length 2 mm) fabricated and characterized in **Chapter 3** are used as a reference.

Figure 5-5. Emission characteristics of random QCLs. (a) Light-current-voltage (LIV) curve of a random QCL with pillar radius of 6.4 μm , FF = 36.5%, and $\Delta = 2.5$ μm . The light-current curve of a ridge laser at 10 K is plotted for comparison (violet dashed lines); its intensity is reduced by a factor of four for clarity. (b) Emission spectrum of the same device at the rollover current density, measured at 10 K. The spectral resolution is 0.2 cm^{-1} . The spectral peaks occur between 3.1–3.4 THz, in agreement with the simulation results shown in Fig. 5-2(d) and Fig. 5-3(a).

Fig. 5-5(a) shows the light-current-voltage (LIV) curves of a random QCL with a pillar radius 6.4 μm , a FF of 36.5% and $\Delta = 2.5$ μm . The lasing threshold at 10 K is around 1.60 kA/cm^2 . This value is slightly larger than that of the ridge laser (1.33 kA/cm^2), mainly attributable to the BCB losses. Just above threshold, the light intensity

increases sharply due to the rapid appearance of many modes with comparable Q factors. The emission spectra are recorded by a FTIR with a DTGS THz detector (response time $\sim 1\text{ms}$) with a total scanning time of over 1 minute, and thus represent time averages over multiple pulses (it would be interesting to study dynamical shot-to-shot fluctuations of the lasing modes, but this would require THz detectors with a much faster response time). Measured by our existing system, the spectrum and power of the device are stable during the characterization. At the rollover current density, 13 lasing peaks can be easily distinguished in the emission spectrum (Fig. 5-5(b)), measured using an FTIR resolution of 0.2 cm^{-1} ; additional peaks are likely to be resolved with a higher resolution spectral measurement. The lasing peaks occur within the range 3.1–3.4 THz, which is in good agreement with the simulation results showing that this is the frequency range where the modes are strongly localized and confined in the QC pillars (Fig. 5-2(d) and Fig. 5-3(a)). Experimental evidence were also found that the lasing modes are strongly localized, by partially covering the device surface with a copper sheet, whereupon some of the peaks were greatly suppressed (due to the light emitted from the covered area not reaching the FTIR system), while other peaks were largely unaffected. For further details, see **Section 5.5.3**.

In comparison to the emission spectra of a QCL ridge laser, no apparent spectral periodicity can be found in the random laser spectrum. Moreover, the dependence of the lasing spectra on applied current reveals a qualitative difference between the behavior of the random QCL and the reference ridge laser. As shown in Fig. 5-6(a), the emission of the ridge laser is continuously blue-shifted from 3.05 THz to 3.15 THz with increasing pumping current. This is due to a blue-shift in the gain center of the QC medium, a typical feature of the three-well resonant phonon active region design employed here. By contrast, the lasing peaks of the random QCL experience negligible shift with current; the principal peaks are clustered around 3.3 THz, away from the gain center of the QC medium. Just above the threshold current, multiple peaks appear almost simultaneously and their intensities increase monotonically with pumping current until the rollover current. This is consistent with the interpretation that the

random lasing modes are determined by the high-Q band edge modes of the 2-D scattering medium.

Figure 5-6. Comparison of the emission spectra between a random QCL and a ridge QCL. (a) Emission spectra of a double-metal waveguide ridge laser (width = 106 μm and length = 2 mm) at different pumping current densities (the spectra are selected to better present the gradual blue shift of emission peaks). (b) Emission spectra of a random QCL (pillar size 6.4 μm , FF = 36.5%, Δ = 2.5 μm) at different pumping current densities. The spectral resolution is 0.2 cm^{-1} for the random laser, and 0.5 cm^{-1} for the ridge laser, respectively.

5.4.2 Tunability of the emission spectral range

The emission characteristics of the random QCL can be tailored by changing the parameters of the underlying scattering medium, such as the scatterer size and/or the density of the scatterers. The position of short-range order induced gap is approximately inverse to the effective lattice constant $a \propto r/\sqrt{FF}$. Fig. 5-7(a) shows the simulated power spectra for structures with varying FF, as well as varying pillar radius; Fig. 5-7(b) shows the corresponding experimental emission spectra. As expected, the gap in the calculated power spectra, and the peaks in the emission spectra, both undergo blue-shifts when r is reduced with fixed FF, or when FF is increased with

fixed r . The experimentally-determined spectral frequency range and their shift with r and FF agree well with the calculated high-Q band edge frequencies.

Figure 5-7. Emission tuning of the random structures. (a) Calculated power spectra for different random structures, with device parameters indicated in the figure, without considering the BCB waveguide losses. The arrows indicate the trend of the band-edge shift with varying parameters. (b) Measured emission spectra of the fabricated random QCL devices. The asymmetric profile of emission spectra is caused by the fact that the higher frequency band edge modes have a larger overlap with the BCB waveguide (thus with higher losses) compared to lower frequency band edge modes. See Fig. 5-2(d) for the simulation results.

5.5 Supplementary information

5.5.1 Multiple strongly localized modes within one structure

For the short-range order amorphous structure used for simulation and experiment in the main text, the modal properties have been calculated over a wide frequency range (2.8 THz-3.8 THz) by the FEM using the Comsol Multiphysics software. Fig. 5-8 shows a selection of multiple localized random modes supported by this structure, apart from those shown in Fig. 5-3(a) of the main text. As can be seen, these modes are strongly localized at different lateral positions.

Figure 5-8. The calculated field intensity distributions of different frequencies for the structure with pillars radius of $6.4\ \mu\text{m}$, filling fraction 36.5%, total area $1000\ \mu\text{m} \times 1000\ \mu\text{m}$, and $\Delta = 2.5\ \mu\text{m}$. The display windows of the intensity distributions are $300\ \mu\text{m} \times 300\ \mu\text{m}$, taken from different sites across the $1000\ \mu\text{m} \times 1000\ \mu\text{m}$ structure. The refractive indices used in this simulation for the pillars and BCB, which fills the space between the pillars, are 3.6 and 1.58, respectively. The white scale bars in all the figures represent a length of $50\ \mu\text{m}$.

Figure 5-9. The calculated field intensity distribution corresponding to the mode (3.305 THz) in Fig. 5-8(e), when the surrounding scatterers are eliminated. The mode still persists with only a very small frequency shift (to 3.247 THz) and the mode distribution is very close to that in Fig. 5-8(e), even in a scattering system with a very limited area of $100\ \mu\text{m} \times 100\ \mu\text{m}$.

To evaluate the localization strength of the modes, the localization length was calculated using the following equation:

$$L_{\text{eff}} \equiv \sqrt{\left[\int I(x, y) dx dy \right]^2 / \left[\int I(x, y)^2 dx dy \right]} \quad (5-2)$$

where $I(x, y)$ is the intensity distribution. The value of L_{eff} ranges from $50\ \mu\text{m}$ to $70\ \mu\text{m}$, which represents a near diffraction-limited localization length, given the emission wavelength $\sim 100\ \mu\text{m}$. Indeed, it is notable through simulations that when pillars surrounding a typical localized mode are eliminated, the mode still persists even with very limited number of pillars remaining (Fig. 5-9). This indicates that the short-range order leads to strong mode localization.

5.5.2 Vertical quality (Q) factor approximation

As the apertures are randomly distributed, the optical modes lack the excellent anti-symmetry that allows good far-field cancellation as in the periodic structures. But this is not to say the modes are totally symmetric relative to the output apertures. In fact, the modes of our random lasers can be treated as a hybrid of the symmetric and anti-symmetric modes. As shown by the example in the figure below (Fig. 5-10), most of the nodes of in-plane Magnetic field are located in the center of pillars, just below the apertures. Therefore, the lasing optical modes are largely antisymmetric with regard to the apertures. This, combined with the fact that the diameter of the aperture is deep sub-wavelength ($3\ \mu\text{m}$ compared to $100\ \mu\text{m}$ wavelength), leads to the large Q factor (several thousand) in vertical direction, estimated by 3-D finite-element method (FEM) simulations.

Figure 5-10. (a) Field intensity distribution of the eigenmode with frequency 3.305 THz, corresponding to the mode in Fig. 5-8(e). The Magnetic components of the field distribution in x (b) and y (c) direction.

5.5.3 Modal localization demonstration

To demonstrate the modal localization, a copper sheet was fixed at a distance of $\sim 1\ \text{mm}$ above the device surface, such that around half of the device surface was covered. Emission spectra were then compared with, and without, the copper sheet in place, with the position of the device and the driving current remaining fixed between the measurements. As shown in Fig. 5-11, the peaks at 3.218 THz, 3.305 THz and 3.335

THz (black arrows) are greatly suppressed by the presence of the copper sheet, as the corresponding modes are physically covered by the sheet. The peak at 3.295 THz (red dashed arrow), however, remains the same, indicating this mode is totally located in an uncovered region. This further indicates that the optical modes of our random laser are spatially localized and separated.

Figure 5-11. The emission spectra of the fabricated random laser with (black) and without (red) partial covering by a copper sheet. The spectra were measured at the same pumping current (~ 5 A) and with a fixed device position. The spectral resolution is 0.2 cm^{-1} . The red dashed and black solid arrows are used for guiding eyes.

5.6 Discussion and summary

By fabricating the dielectric pillars from a THz quantum cascade gain medium with low-index BCB as planarization material for top contact defining, 2-D disordered structures with high scattering efficiency have been achieved in THz QCL platform. TM polarized light is found to be strongly confined within the disordered medium.

Multimode random lasing has been achieved around 3.3 THz. The weak short-range order introduced during the structure generation is beneficial to ensure high quality-factor modes that have a large overlap with the active material. The fact that there are multiple modes, subject to strong localization, is highly promising for future applications; for example, these devices may be able to serve as high-intensity, low-spatial-coherence sources for speckle-free full-field imaging at THz frequencies. Based on these results, even more strongly multi-modal behavior could be achieved in three ways: (i) by replacing the BCB with a lower-loss supporting material, such as cyclic olefin copolymer [183,184], which would significantly increase the modal Q factors; (ii) by connecting the pillars with dielectric veins, which would reduce or eliminate the need for the BCB support [175]; and (iii) by increasing the short-range order through more sophisticated placement algorithms, such as molecular dynamics-based jam packing, which would again increase the modal Q factors.

It has been found that the emission spectrum of the random QCLs can be easily controlled by the size and FF of the scatterers, but is relatively insensitive to the pumping current. Since the individual mode appears to be quite strongly localized, this raises the interesting prospect of designing a random QCL where the scatterer parameters vary across the surface. This would lead to random lasing over a wide spectral range. In terms of emission power, optimization work on for example the aperture size can be done to enhance the power level.

6 Multimode Terahertz Random Lasing with Metallic Pillars

In this chapter, a new class of random laser with optical feedback provided by multiple scattering of metallic pillars in terahertz (THz) frequency range are developed. Such metallic scatterers are especially suitable for transverse-magnetic (TM) polarized light. In absence of the plasmonic resonance for TM wave, the metallic pillar maintains a high scattering efficiency and small ohmic losses over a broad bandwidth, making a broadband random laser promising. It is demonstrated that such random lasers, with metallic pillars embedded in THz quantum cascade laser gain medium, outperform their dielectric-counterparts in terms of the lasing threshold, the spectral range and number of lasing modes. Complex emission spectra are captured and more than 25 emission peaks can be observed across the gain spectral range of the laser.

6.1 Introduction

The random lasing frequencies have been demonstrated in the frequency range from the near-ultraviolet to the mid-infrared. Even recently, the random laser has been demonstrated to work at terahertz (THz) frequencies by fabricating randomly-distributed dielectric pillars [171,185] or air holes [157] onto a THz quantum cascade wafer. Especially for random THz lasers with dielectric pillars embedded in a surrounding low-index polymer, typical multimode emission with strong modal localization has been demonstrated [185] mainly due to the efficient optical scattering of the scatterers around Mie resonance frequencies. Although such a high scattering efficiency is beneficial to form high-quality (Q) modes and thus for random lasing with better performances (lower threshold, more lasing modes, compact system size, etc.), it also limits the random lasing to the Mie resonance frequency range, impeding the broadband random lasing applications. One possible strategy for random lasing over a wide spectral range is to utilize random structures with a certain parameter

polydispersity (e.g. the polydispersity of scatterer size). However, there is an expense of laser performance [182,186]. Alternative way is to adopt ‘non-resonant’ scatterers, featuring a relatively frequency-insensitive scattering efficiency over a broad spectral range. Typical scatterers of such kind are air hole or dielectric scatterer with small index contrast with the ambient medium. However, the scattering efficiency is very poor. Therefore, to devise a scatterer with both ‘non-resonant’ property and high scattering efficiency is desired.

Metallic scatterer normally utilized in random lasers at visible frequency range owing to surface plasmon resonances (SPRs), can be regarded as a perfect reflector in THz or microwave range. The index contrast is very large due to the huge dielectric constants of metal at these ranges and thus, the metallic scatterers are often arrayed to be a photonic crystal (i.e. metallic photonic crystal) for exceptional photonic bandgap properties. For example, in two-dimensional (2-D) metallic photonic crystal (MPhC) with periodically arranged metal pillars, an exceptionally broad PBG spanning from zero frequency to a cutoff frequency can be realized for TM polarized waves, implicating important applications, such as waveguides, filters, polarizers and low-threshold lasers. The passive MPhC with defects or disorder have been explored in terms of stop band and waveguiding [187–189], whereas systematic research on scattering property of individual scatterer, laser cavities and experimental demonstration of lasing with metallic scatterers has not been reported yet to the best of our knowledge.

In this work, I propose 2-D THz lasers with a random matrix of Au pillars, embedded in quantum cascade active material. With very high refractive index contrast between the scatterer and the background host medium, the Au pillar contributes to a large scattering efficiency over a broad low-frequency range without resonances for TM polarized waves. Consequently, Au pillars are particularly suitable for usage in a gain medium with a preference of TM polarization, such as the quantum cascade lasers (QCLs) [21,111] in this work. Multiple random cavities can be created with strongly localized modes when positional randomness of Au pillars is introduced. Advantages of

the random metallic structure over reported high-efficiency random dielectric structure [185] are also demonstrated in terms of density of optical states and broadband application. By incorporating such metallic pillars in a THz quantum cascade wafer with optimized boundary conditions, multimode random lasing with more than 25 emission peaks can be observed over a broad spectral range.

6.2 Structural design of two-dimensional random structures with metallic scatterers

6.2.1 Scattering properties of Au pillar

Since the feedback is provided by the multiple scattering of light, the scattering strength of a scatterer is vital for the performance of random lasers. As a result, it is necessary to study the optical scattering properties of the scatterers. Here, the optical scattering of a circular dielectric pillar or Au pillar has been calculated using finite-difference time-domain (FDTD) simulation. The complex dielectric function of Au is estimated by

$$\varepsilon_c = \varepsilon_\infty - \frac{\omega_p^2}{\omega^2 + i\omega\omega_\tau} \quad (6-1)$$

where $\omega_p = 1.37 \times 10^{16}$ rad/s is the plasma frequency, $\tau = 0.15$ ps is the Drude scattering time, and $\varepsilon_\infty = 1$ is the high-frequency limit of the dielectric constant; these are standard values based on a parameter fit in the far-infrared frequency range [190]. The results are shown in Fig. 6-1(b), where the optical scattering efficiency is defined as the ratio of scattering cross section size to the physical size of the scatterer. The size of dielectric pillar is optimized for random lasing at the gain spectral range of the QCL wafer used in this work.

Figure 6-1. In-plane scattering properties of a pillar scatterer. (a) Schematic diagrams for optical scattering of single pillar at different conditions. (b) The calculated scattering efficiency for different cases, I, dielectric pillar ($R = 6.4 \mu\text{m}$) made of quantum cascade gain medium (mainly GaAs, $n = 3.6$) embedded in benzocyclobutene (BCB, $n = 1.58$); II, Au pillar ($R = 6.4 \mu\text{m}$) embedded in GaAs, the incident wave is transverse-electric (TE) polarized; III, Au pillar ($R = 2 \mu\text{m}$) embedded in GaAs, the incident wave is transverse-magnetic (TM) polarized. The light green shaded region is the gain spectral range of the quantum cascade laser used in this work.

For both TM- and TE-polarized terahertz waves, the scattering efficiency curves are relatively flat over a broad frequency range due to the fact that Au pillar does not support SPRs in the terahertz region. This is in contrast to the dielectric pillar case

where the scattering efficiency peaks at the Mie resonant frequencies (purple curve in Fig. 6-1(b)). For TM polarization, the scattering efficiency of Au pillar (red curve in Fig. 6-1(b)) is higher than that of dielectric pillar over a broad frequency range. And since the penetration depth is orders of magnitude shorter than the wavelength, the absorption of the electromagnetic wave is small over the whole terahertz frequency range, which is beneficial for reducing the lasing threshold. These simply imply potential advantages of Au pillars over dielectric pillars for TM polarized THz waves. In addition, a great distinction of the scattering property can be found for Au pillar with different wave polarization. Comparing to the optical scattering of TE polarized light, the scattering efficiency of metallic scatterer for TM polarization is clearly higher and the advantage is remarkable even for longer wavelengths. This illustrates the specialty of metallic scatterer for TM polarized light.

The scattering curve of the Au pillar for TM polarization can be explained by the Mie theory, which indicates that the scattered far field of a perfect metal cylinder is given by [191],

$$E_z^s = E_0 \sqrt{\pi/2kr} \left[(\ln kR)^{-2} + (kR)^2 \cos \theta - (kR)^4 \cos(2\theta)/8 + \dots \right] e^{ikr - i\pi/4} \quad (6-2)$$

where (r, θ) are the cylindrical coordinates, k is the wave-vector inside the background medium and R is the radius of the perfect metal cylinder. Then the scattering efficiency should be given by

$$Q_{sca} = \pi^2 / \left[2kR(\ln kR)^2 \right] + \pi^2 (kR)^3 / 2 + \dots \quad (6-3)$$

From this equation, it is prominent that the scattering efficiency tends to be infinite for wavelength much larger than the scatterer size ($kR \ll 1$).

Typically, in a random laser, both the structural overall scattering efficiency level which indicates the optical feedback efficiency, and the gain volume which is closely related to the strength of gain that the system can supply to overcome the losses, are

critical for high-performance random lasing. According to Eq. (2-5) and Eq. (2-6), the transport mean free path in 2-D random system can be given by

$$l_t = \frac{1}{\rho\sigma_{\text{sca}}(1-\langle\cos\theta\rangle)} = \frac{\pi R}{\text{FF} \times \text{ScatteringEfficiency}(1-\langle\cos\theta\rangle)} \quad (6-4)$$

Where FF is the filling fraction (FF) of metallic pillars, θ is the scattering far-field angle of single scatterer. Except for the critical role of the FF, the pillar size plays a dual role in determining the scattering strength of random structure. Firstly, a smaller pillar size directly reduces the transport mean free path. And secondly, a smaller pillar size induces a higher scattering efficiency as shown in Fig. 6-1(b), which further reduces the transport mean free path, and thus a stronger scattering strength of the random structure. For the Au pillar scatterer ($R = 2 \mu\text{m}$), the averaged cosine of the scattering angle $\langle\cos\theta\rangle$ is estimated to be around 0.028 for free space wavelength of $100 \mu\text{m}$. Therefore, the transport mean free path is around $33 \mu\text{m}$ for FF of 6%, slightly larger than the effective wavelength in the medium. This calculation indicates the high efficiency of the metallic pillar scatterers. As the small FF (e.g. 6%) of scatterers can maintain an overall strong scattering efficiency, the gain volume is especially large to provide gain to the random modes. As a result, the metallic pillar scatterer is also beneficial for high-performance random lasing in terms of gain volume.

6.2.2 Optical modal properties of two-dimensional random structures with Au pillars

To shed light on the optical modal property of 2-D random medium with Au pillars, 2-D finite-element method (FEM) simulations using Comsol Multiphysics have been conducted to study the eigenmode properties. In the simulations, only metallic ohmic loss and in-plane loss are considered. Au pillar size is chosen to be $R = 4 \mu\text{m}$ with the consideration of experimental fabrication capability. The pillars are randomly distributed within a region of $500 \mu\text{m} \times 500 \mu\text{m}$ without any overlap. Random structures with scatterer filling fraction (FF) ranging from 6% to 20% have been

calculated over the gain spectral range (2.95 THz – 3.45 THz) of the QCL wafer. It can be found that for low-FF structures (e.g. 6%, as shown in Fig. 6-2(a)), the typical optical mode extends over a broad area, with a significant amount of energy escaping through the boundaries. When the FF gets larger (e.g. 12%, as shown in Fig. 6-2(b)), the in-plane confinement gets better and the electric field becomes more localized due to the stronger back scattering by the pillars. When the FF further increases (e.g. 18%, as shown in Fig. 6-2(c)), the optical field becomes strongly localized within the structure with very small modal size; and energy can barely leak outward through the boundaries. To have more insights into the behavior of the random media with various FFs, I calculated the Q factors with consideration of the metal loss and the effective modal size L_{eff} of optical modes for each FF. The latter is defined as

$$L_{eff} \equiv \sqrt{\left[\int I(x, y) dx dy \right]^2 / \left[\int I(x, y)^2 dx dy \right]} \quad (6-5)$$

Both Q and L_{eff} were obtained by averaging over the 150 highest-Q eigenmodes. The results are shown in Fig. 6-2(d). It can be found that the modal size dramatically decreases with increasing FF of metallic pillars, indicating the improvement of in-plane modal confinement. When FF comes to 20%, the modal size decreases to the wavelength scale and sub-wavelength confinement can be achieved with even higher FF. In terms of Q factors, with the increase of FF, the averaged Q factor first increases, up to certain value for FF in the range of 12% – 16%, and then decreases for even higher FF. The last feature can be attributed to the increasingly severe metallic absorption effect in a smaller cavity when FF becomes large. As can be seen in Fig. 6-2(d), the metallic absorption losses, averaged over 150 eigenmodes for each structure, increase almost linearly with FF. Overall, the optimal FF range for random laser application is 12% – 16%.

Figure 6-2. Optical properties of two-dimensional (2-D) random structures with Au pillars. Typical field intensity distributions of random medium with metallic pillar filling fraction (FF) of (a) 6%, (b) 12%, (c) 18%. The white dots correspond to Au pillars and the intensity is normalized. (d) Averaged quality (Q) factors, the averaged effective modal size, and averaged metallic ohmic loss of eigenmodes with the corresponding Q factor ranking top 150 for different FF structures.

In addition, the calculations have also been applied to the TE polarization cases with the same structures. Comparing to the calculation results with TM polarization, the optical losses induced by metallic absorption for TE polarization is in the same level, but the in-plane confinement is worse as shown in Fig. 6-3. It is also feasible to enhance the in-plane localization by increasing FF, whereas this is accompanied by a higher metallic absorption and a reduced gain medium area. These results illustrate that such Au pillars is especially suitable for TM-polarized random lasers.

Figure 6-3. Typical field intensity distributions of a 2-D random structure with Au pillars for TE polarized light. The Q factors of selected eigenmodes are among the highest in the frequency range of 3 – 3.2 THz. The structure is the same as the one in Fig. 6-2(b). A direct comparison between the TM and TE modes indicates that TM eigenmodes are better localized within the structure.

In our previous work, the 2-D THz quantum cascade random lasers (QC-RLs) with dielectric scatterers embedded in benzocyclobutene (BCB) [185] have been demonstrated. A scatterer size of $R = 6.4 \mu\text{m}$ (structural FF~35%) was chosen to locate the bandgap range to be 3.45 – 3.63 THz to fully make use of the gain spectral range. Eigenmodes have been calculated over a broad frequency range for multiple structures with the same generation parameters. All the in-plane Q factors larger than 200 are gathered. At every frequency interval of 0.1 THz, the Q factors are averaged and the number of the modes got averaged over structures.

As shown in the Fig. 6-4(a) and (b), an obvious bandedge effect can be observed around 3.2 – 3.45 THz, where the highest averaged Q factor with large standard deviation exists. When it deviates from this bandedge range at lower frequency direction, the averaged Qs decrease and the number of modes increase firstly and then decrease to very small values. In the frequency range larger than 3.45 THz, the modes with Q larger than 200 rarely exist, indicating the existence of a stop band. In the even higher frequency range, the Q factor of eigenmodes is severely limited, however,

localized modes can always be found. This is mainly caused by the BCB absorption due to the large modal overlap with BCB at higher bandedge as discussed in **Section 5.2**.

Figure 6-4. Optical property comparison of the random structures with dielectric pillars and Au pillars respectively. (a) The averaged Q factors with $Q > 200$ and (b) the corresponding number of eigenmodes at every frequency interval of 0.1 THz for random dielectric pillar structures (purple curve), random Au pillar structures (red curve). The light gray shaded area indicates the bandgap region of dielectric pillar structures. The structures have an overall size of $500 \mu\text{m} \times 500 \mu\text{m}$ with dielectric pillar size of $R = 6.4 \mu\text{m}$ and Au pillar size of $R = 4 \mu\text{m}$, respectively.

With replacing the $6.4 \mu\text{m}$ dielectric pillars by $4 \mu\text{m}$ Au pillars and background BCB by quantum cascade active material, the same simulation work has been done to the random metallic structures (FF~13%). The bandgap effect induced by short-range positional order in metallic structures is trivial, due to the absence of scatterer resonances. As shown in Fig. 6-4, the averaged Q factors and the number of modes remain a fairly high level over the whole frequency range, superior to the dielectric counterparts. With increasing frequency, both the Q factors and the number of modes slightly increase. This can be attributed to relatively larger cavity size and thus, higher density of optical states. As a matter of fact, the larger density of high-Q modes of random metallic structures is also due to the higher gain volume than the dielectric

structures (87% vs 35%). In conclusion, the random structures with Au pillars are more advantageous for broadband random lasing application with higher Qs, large density of high-Q states over a broad frequency range and larger gain volume.

6.3 Device fabrication and characterization

To fabricate the THz QC-RLs with Au pillars, Ti/Au was firstly deposited both on top of the QCL structure and onto an n+ doped GaAs host substrate; this was followed by Au/Au thermo-compression wafer bonding. The QCL wafer was then polished and selectively wet-etched to remove the substrate down to an etch-stop layer. Then the etch-stop layer was removed by HF solution. Through conventional photolithography, mask defining and inductively coupled plasma (ICP) dry etching of the active region with gas ratio (in SCCM) of $\text{Ar}/\text{Cl}_2/\text{BCl}_3 = 10/5/5$, the random air holes were etched through the QCL wafer with intended radius ($\sim 4 \mu\text{m}$). Then, an insulation layer of 200 nm SiO_2 was deposited by a process of tetraethyl orthosilicate (TEOS) to cover the bottom and side wall of the air holes. The SiO_2 on top of active region was then removed and a thick top contact layer ($\sim 25 \text{ nm}/1400 \text{ nm}$ Ti/Au) was sputtered onto the structure, guaranteeing that random air holes were completely covered by the metal layer with minimum thickness of 100 nm. Irregular boundaries of top contact layer were formed by photolithography and wet etching of the metallic layer. Focused ion beam (FIB) etching process was utilized to fabricate random apertures in the top contact metallic layer. The etching time was carefully controlled to make sure that the semiconductor under the metal was not etched significantly. The 3-D schematic diagram of a random structure with metallic pillars is shown in Fig. 6-5. The host substrate was then thinned down to around $180 \mu\text{m}$ and a $15 \text{ nm}/200 \text{ nm}$ Ti/Au layer was deposited as bottom contact. The samples were cleaved, indium-soldered onto copper submounts, wire-bonded and finally attached to the heatsink in a cryostat for measurement. The fabricated 2-D THz random QCLs with Au pillars were characterized using the same methods as in **Section 4.3** and **Section 5.3**.

Figure 6-5. 3-D schematic diagram of a random structure with metallic pillars. (a) Zoom-in cross section view of single Au pillar. (b) 3-D full structure with a cross section view. The passive and electrical-pumped quantum cascade active material regions are colored by light pink and purple, respectively.

6.4 Experimental results

6.4.1 Preliminary experimental result

For random lasers with ‘resonant’ scatterers (such as dielectric pillars with Mie resonance, or metallic particles with SPRs), to optimize the laser performance, the scatterer size should be carefully chosen to align its resonance peak with the gain peak of the laser medium [181,185,192]. In this work, there is no such requirement on the Au pillar due to the frequency-insensitive scattering efficiency. As the gain is provided by the background medium, which is the THz QCL active region, it is desirable to maximize the area ratio between the gain medium and the pillars while keeping the overall scattering strength of pillars. In this regard, smaller scatterers are preferred, especially considering that the scattering efficiency of the smaller pillar is even higher than that of the larger ones (red curve in Fig. 6-1(b)). In this work, the radius of the Au pillar was chosen to be 4 μm; even smaller size will impose great fabrication challenge.

The same THz QCL wafer based on a three-well resonant-phonon GaAs/ $\text{Al}_{0.15}\text{Ga}_{0.85}\text{As}$ design used in previous work was also employed in this work for performance comparison. For the fabrication of randomly distributed metallic pillars, it was realized by etching air holes through the QCL active region and then covering it completely, the side wall and bottom, with $\text{SiO}_2/\text{Ti}/\text{Au}$ (200 nm /30 nm /1400 nm) (see **Section 5.3**). As the electric field can't penetrate through the metallic layer, the scatterer resembles an Au pillar. The insulating gap between Au scatterer bottom and bottom contact layer is very small. Thus, its influence to the scattering efficiency of the scatterer and the field distributions of the eigenmodes is negligible. To minimize the specular reflection at the boundary, irregular jagged boundaries were created. The reflection of boundaries is estimated to be less than 1.5% and all the light escaped through the boundaries can be absorbed by the passive quantum cascade material outside. The light inside the random cavity is extracted from the holes randomly opened on the top contact metal layer with the restriction that the openings cannot overlap with the pillars. As the scattering efficiency of these openings is very weak, the light extraction holes impose little influence on the in-plane modal localization which is verified by 3-D FEM simulations using Comsol Multiphysics.

However, it was found that the fabricated devices have limited power performance due to the low collection efficiency induced by poor far fields (see weak emission peaks in Fig. 6-6), which is one of the drawbacks of random laser. Optimizing the structures for directional emission will contribute to higher collection efficiency, but it is out of the scope of this work. Another two factors also deteriorate the performance of random laser. The absorbing boundaries, which were created to verify the Fabry-Perot cavity-less random lasing, practically significantly suppress the Q factors of eigenmodes. And the extraction holes will also induce considerable vertical emission losses if the position is not optimized. As there are hundreds of tightly confined high-Q modes promising for lasing and the typical field distributions are random without symmetry preference with regard to the position of scatterers/extraction holes [179,185], no systematic strategy has been proposed to optimize the position of

light extraction holes. Therefore, it is difficult to control the influence of extraction holes over Q factors and the laser performance.

Figure 6-6. Emission spectrum of a THz quantum cascade random laser (QC-RL) with absorbing boundaries. The Au pillar radius is 4 μm , FF = 13% and total area is 515 $\mu\text{m} \times 515 \mu\text{m}$. The extraction apertures have a radius of 2.5 μm and FF of 10%. The device was characterized at 9 K with a repetition rate of 10 kHz and a pulse width of 500 ns.

6.4.2 Performance optimization

For the issues described above, fabricating mesa boundaries by etching the passive quantum cascade material away (Fig. 6-5(a)) is feasible to make up for the deficiencies. The etching was conducted by ICP with top contact metallic layer as a self-aligned hard mask, which was followed by FIB aperture opening process. In this way, the laser performance can be enhanced in two aspects. Firstly, the in-plane Q factors and number of high-Q modes can be improved by a mesa boundary with higher index contrast to the environment. Many modes, originally suffering from large boundary absorption, experience a higher reflection by the mesa and thus get more energy emitted through the extraction apertures. Secondly, the wall-plug efficiency of random lasers will be enhanced by the mesa boundaries. Because the escaped light through the boundaries

can be captured in direction normal to the laser surface induced by the large emission divergence of the edge-emitting THz QCL (note that a gain medium of only 10 μm thickness is sandwiched by metal layers) [129].

Therefore, I have also fabricated a series of mesa structures with random Au pillar FF spanning from 10% to 17%. Fig. 6-7 shows the light-current-voltage (LIV) curves and emission spectra of a random QCL with a FF of 12%. The lasing threshold at 9 K is $\sim 1.53 \text{ kA/cm}^2$, slightly higher than that of a ridge laser (1.33 kA/cm^2) with a comparable effective size of $2 \text{ mm} \times 106 \mu\text{m}$. However, this threshold value is smaller than that of the random dielectric pillar structures with larger size ($1 \text{ mm} \times 1 \text{ mm}$), as discussed in **Section 5.4**. The highest working temperature is around 95 K and the peak power reaches 0.46 mW without a correction of light collection efficiency. The power performance is mainly limited by the quantum cascade wafer as the highest peak power achieved by the ridge laser is only 1 mW. As the gain spectral center of QCL will have blue-shift with increasing pumping intensity [185], a typical feature of the three-well resonant-phonon active region design employed here, the principle peaks of emission spectra will also be transferred to higher frequencies. Three complex emission spectra at different pumping currents are shown in Fig. 6-7(d). These emission spectra are selected to fully present all the emission peaks in the whole dynamic range. The emission peaks cover a frequency range of 3.05 THz – 3.45 THz, mainly limited by the gain spectral range of the material. More than 25 emission peaks can be observed, which is almost twice of the emission peak number for the reported dielectric random structure with a larger structure size [185]. The performance of the laser can be further improved by employing even lower-loss material, e.g., Cu [150], to replace the Au used in our current devices.

Figure 6-7. Structural optimization and the characterization results. (a) 3-D schematic graph of THz QC-RL with Au pillars utilizing mesa boundaries. (b) Scanning electron microscope (SEM) image of the fabricated device with mesa boundaries. (c) Light-current-voltage (LIV) curves at different temperatures and (d) emission spectra at 9 K for different pumping currents of the fabricated THz QC-RL with mesa boundary. The total structure size is around $515 \mu\text{m} \times 515 \mu\text{m}$ and the FF of Au pillars is 12%.

Peak No.	1	2	3	4	5	6	7	8	9
Freq. (THz)	3.0535	3.0930	3.1091	3.1457	3.1674	3.1778	3.1912	3.2052	3.2133
Peak No.	10	11	12	13	14	15	16	17	18
Freq. (THz)	3.2301	3.2325	3.2389	3.2461	3.2538	3.2682	3.2721	3.2816	3.2922
Peak No.	19	20	21	22	23	24	25		
Freq. (THz)	3.3051	3.3225	3.3598	3.3700	3.3910	3.4005	3.4215		

Table 6-1. The peak frequencies corresponding to the spectra in Fig. 6-7(d).

6.5 Discussion and summary

2-D multimode THz QC-RLs with metallic (Au) pillars have been demonstrated for the first time. Metallic scatterers have been utilized in random lasers at visible frequency range [55,69,70,192,193], owing to the surface-plasmon-enhanced optical scattering efficiency and localized electromagnetic field near the particle surface for larger optical amplification. However, the implementation of such scatterers also introduces serious light absorption which is detrimental for optical oscillation in the disordered medium. In the THz frequency range, the metallic ohmic loss is moderate, implicating the advantage of the metallic pillars for THz random laser application. The most significant specialty of metallic pillar is the non-resonant, however, efficient scattering property, which is advantageous over conventional dielectric scatterer with Mie resonances and the metallic scatterer with surface electromagnetic wave coupling, leading to a broadband and efficient modal confinement with large gain medium volume. With the structure boundary optimization, good threshold and power performance have been realized. Most importantly, more than 25 emission modes have been achieved with a limited structure size. Random lasing in broader spectral range with more lasing peaks could be achieved if a THz QCL wafer with broader gain spectral range was used. This work may broaden the intriguing view of metallic scatterers in random laser applications.

7 Conclusions and future work

7.1 Conclusions

Arising from the material growth and fabrication processes, disorder is a double sided sword for photonic applications. Eliminating the disorder is always difficult and must come at a high cost. However, it is also possible to tame the disorder by exploring its advantages. The optical properties of disordered medium have been extensively studied and the disordered structures have been exploited for various novel applications, of which the most significant example is the random laser. Most of the research on disordered photonics is in the short-wavelength range. Nevertheless, in the technically important terahertz (THz) frequency range, the disordered photonics are least explored.

In this thesis, I pioneer the investigation of modal and emission properties of various disordered structures based on THz quantum cascade laser (QCL) platform. **In chapter 2**, the working principle and the state-of-the-art of lasing in disordered medium have been reviewed. As the feedback of random laser is sustained by the multiple scattering of light, the scattering strength of disordered structures is critical for the operation mechanism and also the performance of the laser. The scattering strength of disordered structures is determined by the positional correlations and the scattering efficiency of individual scatterer. Therefore, the basic design strategy of high-performance disordered structure is to incorporate beneficial structural order or to devise high-efficiency scatterers.

In chapter 3, the THz QCL wafer used in this thesis has been scrutinized in terms of the theoretical basis, working principles, wafer design, waveguide scheme, fabrication processes and the preliminary experimental results of the fabricated conventional laser. It has been pointed out that the double-metal (DM) waveguide is advantageous for the realization of disordered laser in terms of restricting photon escaping in the vertical direction and fabrication accessibility. The gain property of the

wafer has also been analyzed according to the preliminary results, as guidance for the lasers to be developed.

In chapter 4, the first trial has been made by utilizing the long-range order to enhance the confinement of the disordered eigenmodes near the bandgap region. The idea is to introduce the positional disorder into periodic configurations. Numerical calculation indicates that the disordered eigenmodes always feature tight confinement and small optical losses near the bandgap frequency window of the underlying long-range order. Through fabricating one-dimensional disordered grating structures on top of the DM waveguide of the THz QCL, 8 emission peaks at 3.2 THz have been observed, which are located near the bandgap region of the periodic counterparts. The spatial localization of light is proved by the simulation work and experimental measurements.

Instead of utilizing long-range order for microcavity formation, two-dimensional (2-D) disordered structures with high-efficiency scatterers embedded in benzocyclobutene (BCB) and short-range positional order are studied **in chapter 5**. Due to the strong scattering efficiency of a high-index dielectric pillar for transverse-magnetic polarized (TM) wave, multiple strongly localized modes can be sustained by such structures. The weak short-range order induced by the pillar distribution is sufficient to ensure high-Q modes that have a large overlap with the active material. By fabricating the 2-D structures on a THz QCL wafer, more than 13 random emission peaks with localization modal property have been demonstrated. And the emission threshold is obviously reduced. In addition, the emission spectra are found to be easily tuned by tailoring the scatterer size and filling fraction.

The resonant scatterer is beneficial for better performance of multimode lasing (lower threshold, more lasing modes etc.) due to its high scattering efficiency. However, the emission spectral range is at the same time limited to the resonance frequency range of the scatterer, impeding the broadband random laser application. **In chapter 6**, a novel scatterer, metallic pillar, is proposed for the multimode broadband random lasing.

The non-resonant property of metallic pillar features a relatively frequency-insensitive scattering efficiency, and the high scattering efficiency is maintained over a broad frequency range for TM-polarized light. The random structures consisting of metallic pillars embedded in THz QCL gain medium have been numerically demonstrated to outperform their dielectric-pillar counterparts in terms of the spectral range and the density of high-Q modes. By fabricating the random metallic scatterers via covering the etched air holes on a THz QCL wafer with a thick metal layer, random lasing over a broad spectral range with more than 25 emission peaks have been observed. Even lower lasing threshold has also been achieved. The lasing spectral range is mainly limited by the gain spectral range of the wafer used.

7.2 Future work

Some foreseeable future works include but are not limited:

Firstly, the modal localization has been verified by partially covering the device surface using a copper sheet to prevent some of the emissions from being captured by the characterization system. However, using a near-field scanning optical microscopy (NSOM) with a special cryocooling system to characterize the mode profile is always desirable, even though there is no access to this equipment at present. Direct observation of mode profiles will not only provide a better understanding of the modal properties of the laser and further improve the performance, but also enables us to investigate more interesting problems, such as the mode competition/couplings, the statistical laser physics.

Secondly, it has been demonstrated that the random structure with metallic pillar scatterer supports broadband random lasing. It is promising to adopt a THz QCL wafer with broader gain spectral range to further expand the random emission spectral range for low-spatial coherence applications. And using metals with even lower ohmic losses, such as Cu, may further enhance the laser performances.

Thirdly, for the typical random lasers, the collection efficiency of coherent radiation is quite low due to the poor emission directionality, which is a characteristic of random feedback. This is extremely severe for the lasers developed in this thesis where deep-subwavelength DM waveguide and emission apertures are used, causing almost 180° emission divergence. A broadband surface emission grating can be fabricated at the device periphery to extract the light in vertical direction. Special cavities, such as chaotic cavities [194] which can sustain numerous chaotic modes similar to random modes and emit in certain preferential directions, can also be developed for speckle-free full-field imaging application.

Finally, in this thesis, I mainly design disordered structures with beneficial structural order or efficient scatterers to realize the localization of light and multimode lasing. It is also possible to apply similar design strategy to the mid-IR frequencies. However, all of these designs feature work well at THz frequencies where metallic losses are extremely low, but may lead to unacceptably high losses if one seeks to scale these devices towards mid-IR frequencies. To realize a multimode mid-IR QCL, one promising strategy would be to utilize a dielectric pillar design without an enclosing metallic top plate; the pillars can be joined with dielectric veins to provide the connectivity required for electrical pumping. This has been shown theoretically to provide sufficiently high-Q modes for multimode lasing [175].

Publications

Journal Papers:

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2. B. Meng, **Y. Zeng**, G. Liang, J. Tao, X. Hu, E. Rodriguez, and Q. J. Wang, “Broadly continuously tunable slot waveguide quantum cascade lasers based on a continuum-to-continuum active region design”, *Applied Physics Letters* 107, 111110, 2015.
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6. **Y. Zeng**, G. Liang, H. K. Liang, B. Qiang, S. Mansha, L. Li, A. G. Davies, E. H. Linfield, Y. Zhang, Y. Chong, and Q. J. Wang, “Terahertz Emission in One-Dimensional Disordered Systems”, (manuscript in preparation).
7. **Y. Zeng**, G. Liang, B. Qiang, K. Wu, S. Mansha, L. Li, A. G. Davies, E. H. Linfield, Y. Zhang, Y. Chong, and Q. J. Wang, “Two-Dimensional Multimode Terahertz Random Lasing with Metallic Pillars”, (manuscript in preparation).

Conference Papers:

1. **Y. Zeng**, G. Liang, H. K. Liang, S. Mansha, S., L. Li, A.G. Davies, E. Linfield, Y. Zhang, Y. Chong, and Q. J. Wang, "Amorphous Random Lasing at Terahertz Frequency." In *CLEO: Science and Innovations*, pp. SM2L-5. Optical Society of America, 2016.
2. **Y. Zeng**, G. Liang, H. K. Liang, S. Mansha, L. Li, A.G. Davies, E. Linfield, Y. Zhang, Y. Chong, and Q. J. Wang, "Random Lasing at Far-infrared Frequency Range." In *International Quantum Cascade Lasers School and Workshop* (Cambridge, UK, 2016).
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