Designing Subwavelength-structured Light Sources

by

Song-Liang Chua

B.S.E., University of Michigan, Ann Arbor (2006)
S.M., Massachusetts Institute of Technology (2007)

Submitted to the Department of Electrical Engineering and Computer Science
in partial fulfillment of the requirements for the degree of

Doctor of Philosophy in Electrical Engineering and Computer Science

at the

MASSACHUSETTS INSTITUTE OF TECHNOLOGY

February 2013

© Massachusetts Institute of Technology 2013. All rights reserved.

Author

Department of Electrical Engineering and Computer Science

December 17, 2012

Certified by

Marin Soljačić

Professor of Physics and MacArthur Fellow

Thesis Supervisor

Accepted by

Leslie A. Kolodziejski

Chairman, Department Committee on Graduate Students
Designing Subwavelength-structured Light Sources

by

Song-Liang Chua

Submitted to the Department of Electrical Engineering and Computer Science
on December 17, 2012, in partial fulfillment of the
requirements for the degree of
Doctor of Philosophy in Electrical Engineering and Computer Science

Abstract

The laser has long been established as the best possible optical source for fundamental
studies and applications requiring high field intensity, single mode operation, a high degree
of coherence, a narrow linewidth and short pulses. There are many applications that require
lasers of varying frequencies, powers, and far-field properties. In science, the laser is a
useful source in spectroscopy and microscopy, for investigating nonlinear optics phenomena
and nuclear fusion. More commonly, we find them in barcode readers, laser pointers, and
printers. They are also widely used for military, medical, and industrial applications. This
thesis is focused on achieving new understanding of the principles and phenomena involved
in the interaction of light with a variety of material systems, which will in turn guide the
designs of compact lasers with feedback structures having features at the subwavelength-
scale.

The thesis begins by describing the interaction of light with an arbitrary complex ma-
terial system, and implementing them into the electromagnetic model using two different
theoretical techniques suitable for analyzing microstructured lasers: exact finite-difference
time-domain calculations and a semi-analytic coupled-mode formalism. These methods are
first applied to analyze lasing action in the photonic crystal (PhC) slabs. This class of lasers,
commonly referred to as the photonic crystal surface-emitting lasers (PCSELs), can be in-
tegrated on-chip and is essentially the two-dimensional (2D) versions of the second-order
distributed feedback lasers, where the higher quality factor lasing mode (dark Fano reso-
nance) is selected through the symmetry mismatch to the free-space modes. The PCSELs
have not only achieved the highest surface-emitting single-mode power but also the ability
to control the shapes, polarizations and directions of their far-fields. However, as in all laser
cavities, the lasing areas of PCSELs are limited by two fundamental constraints; a large area
tends to promote both multi-mode and multi-area lasing. We propose to overcome both
constraints to achieve single-mode PCSELs of larger areas, and thus higher output powers,
by tuning the regular lasing bandedges of quadratic dispersions in typical PCSELs to form
a single accidental Dirac cone of linear dispersion at the Brillouin zone center. Moreover,
an additional frequency-locking phenomenon at the accidental point, with potentially high
density of states, is analyzed. We demonstrate and distinguish experimentally the exis-
tence of the dark Fano resonances in a macroscopic 2D silicon nitride PhC slab consisting
of a square array of holes. We characterize the passive PhC slab in terms of its resonant
frequencies and radiation behaviors using temporal coupled-mode theory and symmetry
considerations. We also realize lasing at a dark Fano resonance with diluted solutions of
R6G molecules as the gain medium.

Next, we turn our attention to the organic dye lasers whose high tunability in the
visible wavelengths has attracted interests for many years due to their low-cost processing, flexible choice of substrates, and large emission cross sections that can cover the spectral region from ultraviolet to the near infrared. We investigate the laser dynamics in systems of sub-wavelength photonic structures consisting of organic dye molecules, including their photobleaching effects. Our analysis considers both the chemical properties of the dyes and optical properties of the cavities. We also systematically studied the feasibility of lasing under continuous-wave excitations in optically pumped monolithic organic dye lasers. This study suggests routes to realize an organic laser that can potentially lase with a threshold of only a few W/cm².

Lastly, we investigate far-infrared (FIR) (∼ 0.2 – 2 THz) laser emission from optically-pumped rotationally excited molecular gases confined in a metallic cavity. Terahertz radiation has already been used in packaging inspection for quality control, chemical composition analysis, and security screening. Submillimeter spatial resolution imaging and incredibly specific molecular recognition are other compelling uses for terahertz radiation. To apply terahertz radiation beyond laboratory or close range (< 10 m) applications, more powerful (> 100 mW) and efficient sources are required to see through highly attenuating obscurants (including the atmosphere). The fundamental limitations in the performances of FIR molecular gas lasers reside in the molecular gas physics due to the so-called vibrational bottleneck. We seek to overcome the resulting challenges through novel optical designs of the feedback structures. To undertake this task, we generalize previous works to allow for a realistic description of the spatio-temporal dynamics characterizing the molecular collisional and diffusion processes. This work expands the current understanding of lasing action in FIR gas lasers and, thus, could contribute to the development of a new class of terahertz sources able to operate efficiently at room temperature. The advent of quantum cascade lasers to replace CO₂ pump lasers may combine to produce truly compact submillimeter-wave laser sources in the near future.

Thesis Supervisor: Marin Soljačić
Title: Professor of Physics and MacArthur Fellow
Acknowledgments

I would like to express my most sincere gratitude to my thesis advisor, Prof. Marin Soljačić for his tutelage, consistent support and encouragement in numerous aspects, and being an inspiration throughout the course of my PhD. I am also thankful to Marin for his advice and guidance on the personal and career level. I enjoyed the numerous discussions we shared.

I would like to thank my Masters thesis advisor, Prof. Jin Au Kong for his guidance and wisdom. I am grateful to Prof. John Joannopoulos for his precious advice and insightful discussions, and Prof. Steven Johnson for his wonderful instructions in photonic crystals and help in several projects. In addition, I would like to thank Prof. Erich Ippen and Prof. Franz Kärtner for serving on my thesis committee, and Prof. James Fujimoto for advising my academic progress.

I am deeply grateful to Prof. Jorge Bravo-Abad at Universidad Autonoma de Madrid, Dr. Ofer Shapira at QD Vision Inc., and Dr. Ling Lu for their patience, guidance and friendship. I have been working closely with each of them on many exciting projects. They are excellent researchers who offered me tremendous help in so many aspects, from project initiations to hands-on simulations and writing papers. I am particularly grateful to Dr. Henry Everitt at Duke University for giving me the opportunity to embark on the FIR gas laser project and sharing his extensive knowledge in this area. I also wish to thank the DSO National Laboratories for providing me the opportunity and funding to pursue a PhD in the United States, especially to Mr. Chee Seng Tan for making this possible in the first place.

I wish to express my warmest thanks to members in Marin’s group for all the exciting conversations we had. Special thanks to Bo Zhen and Jeongwon Lee for the fruitful collaborations and their hard work in experiments, and to Wenjun Qiu for being the best officemate. I would also like to acknowledge friends in Cambridge who have generously shared their experiences and advices to make a pleasant difference in my MIT experience: Yi Xiang, Trina, Shyue Ping, Henry, Shireen, Wui Siew, Swee Ching, Vincent and Huili.

Finally, I dedicate this thesis to my family, Tong Guan Chua, Sharon Lee, Kellie Chua, and Xingfang Su. This thesis would not have been possible without their understanding and support.
4.3.1 1D periodic slabs (unit-cell calculations) .......................... 57
4.3.2 Finite-sized 1D periodic slabs ........................................ 60
4.3.3 2D periodic slabs (unit-cell calculations) .......................... 62
4.4 Conclusion ..................................................................... 63

5 Experimental realization of photonic crystal surface emitting lasers 65
5.1 Unique high-Q optical resonances in macroscopic photonic crystal slabs 65
5.1.1 Introduction .............................................................. 66
5.1.2 Measured band diagram ............................................... 67
5.1.3 Measured quality factors .............................................. 70
5.1.4 Conclusion .............................................................. 73
5.2 Low-threshold organic laser realized with photonic crystal slabs .... 73
5.2.1 Introduction .............................................................. 73
5.2.2 Lasing measurements and comparisons with model .......... 74
5.2.3 Conclusion .............................................................. 77
5.3 Appendix: Fabrication .................................................... 78
5.4 Appendix: Optical setup (passive system) ............................ 78
5.5 Appendix: Computation .................................................. 79
5.6 Appendix: Coupled-mode theory ...................................... 79
5.7 Appendix: Optical setup (lasing system) ............................. 80
5.8 Appendix: Fraction of pump power absorbed, η .................. 80

6 Threshold and dynamics behavior of organic nanostructured lasers 83
6.1 Introduction .............................................................. 83
6.2 Theoretical framework .................................................. 85
6.2.1 Sub-wavelength structured cavity effects ......................... 88
6.2.2 Continuous-wave lasing system (steady-state analysis) .... 89
6.2.3 Pulsed lasing system ............................................... 92
6.3 Results and discussion .................................................. 93
6.3.1 Intersystem crossing lifetime ratio (τi/τisc) ....................... 96
6.3.2 Dye concentration .................................................. 96
6.3.3 Confinement factor (Γs) ............................................ 99
6.3.4 Spontaneous emission enhancement factor (Fp) ............. 99
7  Spatio-temporal theory of lasing action in optically-pumped rotationally excited molecular gases

7.1  Introduction ................................................................. 108
7.2  Theoretical framework .................................................. 110
  7.2.1  Vibrational states and thermal pools ............................. 110
  7.2.2  Non-thermal rotational levels .................................... 112
  7.2.3  Pump transition rate .................................................. 113
  7.2.4  Semiclassical rate equations ...................................... 113
  7.2.5  Vibrational bottleneck .............................................. 115
7.3  OPFIR laser system: cylindrical waveguide resonator ............. 116
  7.3.1  Spatio-temporal analysis ........................................... 120
  7.3.2  Comparison to experiments ....................................... 120
  7.3.3  General analysis of laser performances .......................... 124
7.4  Conclusion ........................................................................ 128
7.A  Appendix: Rate equations and rate constants of $^{13}$CH$_3$F gas lasers ..... 128

8  Conclusions ........................................................................ 131
List of Figures

3-1 Band diagrams of a vertical cavity surface-emitting laser structure, as well as the 1D- and 2D-periodic photonic crystal slab structures illustrating zero group velocity at the Brillouin zone center. .......................... 35

3-2 Variation of $Q$ as a function of frequency for the lowest two bands above the light line, for both the infinite and finite-sized 1D periodic photonic crystal slab structures. ..................................................... 38

3-3 Total $Q$ of the two band-edge modes as a function of the 1D periodic photonic crystal slab size, $L_x$. ................................................................. 40

3-4 Output power versus input pump-rate relationships of the infinite 1D periodic (unit-cell calculations) photonic crystal surface-emitting lasers having $Q$ values 1964, 451, and 230. ................................................................. 43

3-5 Output power versus input pump-rate relationships of the finite-sized 1D periodic photonic crystal surface-emitting lasers having dimensions of $L_x$ at 20a, 40a, and 80a. ................................................................. 44

3-6 Output power versus input pump-rate relationships of the infinite 2D periodic (unit-cell calculations) photonic crystal surface-emitting lasers having $Q$ values 764, 263, and 126. ................................................................. 46

3-7 Output power versus input pump-rate relationships of the finite-sized 2D periodic photonic crystal surface-emitting lasers having dimensions of $15a \times 15a$, $25a \times 25a$, and $35a \times 35a$. ................................................................. 47

4-1 TM photonic band structure of a triangular array of dielectric rods ($\epsilon_{rod} = 12.5$) in two different background materials ($\epsilon_{bg} = 1, 11$). The rod radius $r$ is tuned to achieve an accidental Dirac-point in the Brillouin zone center. .......................... 53
4-2 Analysis of the bandedge mode spacing and $Q_\parallel$ of various finite-sized PhC cavities, consisting of a triangular array of dielectric rods embedded in air.

4-3 The band structures and corresponding $Q_\perp$ values of two $0.3a$-thick 1D periodic photonic crystal slabs, consisting of alternating high ($\epsilon_{\text{high}}$) and low dielectric constant ($\epsilon_{\text{low}}$) materials. The structures are tuned so that the bands are accidental degenerate at $\Gamma$.

4-4 Analysis of the bandedge mode spacing and quality factors of various finite-sized $0.3a$-thick 1D periodic photonic crystal slabs, consisting of alternating high ($\epsilon_{\text{high}} = 12.5$) and low ($\epsilon_{\text{low}} = 6.25$) dielectric constant materials.

4-5 TE-like band structure and corresponding $Q_\perp$ values of a GaAs-based 2D periodic photonic crystal slab on AlAs substrate, consisting of a square array of air-holes. The hole radius is tuned to achieve accidentally degeneracy of three bands at $\Gamma$.

5-1 SEM images of the fabricated photonic crystal slab. The structure is made of a 250 nm thick Si$_3$N$_4$ layer with periodic cylindrical holes on SiO$_2$ substrate.

5-2 Comparisons of the photonic crystal slab band diagrams obtained from reflectivity measurement and finite difference time domain calculations.

5-3 Simulation results for radiative quality factors of the TE- and TM-like bands near the Brillouin zone center.

5-4 $Q^{\text{total}}$ values retrieved by fitting the measured data of the TE- and TM-like bands near the Brillouin zone center.

5-5 Schematic drawings of the energy levels in organic molecules under short pulse excitation, and experimental setup in the lasing measurement of R6G dissolved in methanol placed on top of a photonic crystal slab.

5-6 Input-output energy characteristics of the organic photonic crystal slab lasing through a dark Fano resonance, under pulsed excitation.

6-1 Laser system consisting of an organic active medium embedded in a passive cavity.

6-2 (a) Output intensity and photobleaching lifetime of the organic laser as a function of the excitation intensity under continuous excitation. (b) Temporal development of lasing action of the laser system near the threshold.
6-3 Lasing threshold as a function of the optical and chemical properties in a R6G organic laser system under continuous excitation.

6-4 Photobleaching modification factor as a function of the optical and chemical properties in a R6G organic laser system under continuous excitation.

7-1 Schematic diagram of the general model used to describe the dynamics of a OPFIR molecular gas.

7-2 The OPFIR laser system considered in our numerical modeling: A cylindrical waveguide resonator of length $L_{\text{cell}}$ and radius $R$ is filled with a suitable gas that lases at the desired THz frequency, while pump power at a much higher IR frequency enters the system from the front window.

7-3 Time dependent properties of the OPFIR laser system under 10 W CW pumping at 250 and 350 mTorr.

7-4 (a) Method of threshold predictions via gain-loss balancing at 100 mTorr. (b) Same as in (a) except that the pressure is at 300 mTorr. (c) Relationship between the threshold intensity and pressure for a range of cell radii from 0.08 cm to 1 cm, as predicted from numerical model.

7-5 (a) Radial spatial variation of the excited inversion for a range of operating pressure from 50 to 450 mTorr, under CW pumping at 100 W. (b) Same as in (a) except that the transition width is also factored in to study the gain. (c) Gain dependence on pressure. (d) The optimum operating pressure and corresponding gain, as a function of the cell radius. From a molecular gas physics standpoint, results indicate that small cavities are favored for high pressure operation.

7-6 $^{13}$CH$_3$F OPFIR laser model used in this work: results of diagnostic studies and theoretical considerations restrict the degrees of freedom so that a numerically tractable, yet physically accurate model is attained.
List of Tables

6.1 Parameters of organic laser system based on R6G solution assumed for steady-state calculations. .......................................................... 105

7.1 Transitional cross sections (in Å²) of 13CH₃F. ............................... 130
Chapter 1

Overview

Light sources are indispensable to our life. There are two major kinds of sources: producing either incoherent or coherent light. The fluorescent lamp and light-emitting diodes are examples of incoherent light sources. They are commonly used for lightings, in displays, indicators, signs, and sensors. On the other hand, a laser emits coherent light. Lasers have long been established as the best possible optical sources for fundamental studies and applications requiring high field intensity, single mode operation, a high degree of coherence, a narrow linewidth and short pulses. There are many applications that require lasers of varying frequencies, powers, as well as shapes, polarizations and directions of their far-fields.

In science, the laser is a useful source in spectroscopy and microscopy, for investigating nonlinear optics phenomena and nuclear fusion. More commonly, we find them in barcode readers, laser pointers, and printers. They are also widely used for military, medical, and industrial applications. This thesis is focused on achieving new understanding of the principles and phenomena involved in the interaction of light with a variety of material systems, which will in turn guide the designs of compact lasers with feedback structures having features at the subwavelength-scale. The unique properties of optical nano-structured materials have already enabled a wide range of very important applications (e.g. in medicine, energy, telecommunications, defense) and are expected to do even more so in the future.

Since the demonstration of the first laser fifty years ago [1], controlling the spontaneous and stimulated emission of atoms, molecules or electron-hole pairs, by placing them in the proximity of complex dielectric or metallic structures has been seen as an attractive way to tailor the lasing properties of active media [2-5]. At the same time, technological
advances have enabled the control of the material structure at length-scales smaller than
the wavelength of light to create new materials (e.g. photonic bandgap crystals, or various
surface plasmon systems) whose optical properties are dramatically different than those
of any naturally occurring material. This rapid advance of nano-fabrication techniques
along with the development of novel theoretical techniques have led to the emergence of
a number of new coherent light sources, with dimensions and emission properties much
different from those found in conventional lasers [6–32]. Among these systems, the photonic
crystal slabs [33,34] have already demonstrated their significant potential to enable efficient
integrated laser sources [8,10,12,16–19,22,23,26,31]. In particular, the photonic crystal slab
lasers have not only achieved the highest surface-emitting single-mode power [35] but also
the ability to control the shapes [36], polarizations [10] and directions [31] of their far-fields.
Chapters 3, 4, and 5 discuss in more details this particular class of lasers.

This thesis begins with a discussion of the computational methods used to solve the se-
cmiclassical laser equations. In Chapter 2, we model the interaction of light with an arbitrary
complex material system, in order to achieve new understanding of the principles involved
in light-matter interaction and to theoretically analyze lasing action in these structures. In
particular, we couple the rate equations to Maxwell’s equations and implement them into the
electromagnetic model using two different theoretical techniques suitable for analyzing mi-
crostructured lasers: exact finite-difference time-domain (FDTD) calculations [9,19,37,38]
and a semi-analytic coupled-mode formalism [33,39]. Each of these offers different kinds of
insight into the system. The four-level model considered in Chapter 2 is later extended in
Chapters 6 and 7 when we consider organic and molecular gas systems, respectively.

Chapter 3 presents a theoretical analysis of lasing action in photonic crystal surface-
emitting lasers (PCSELs). PCSELs are essentially the two-dimensional (2D) versions of
the second-order distributed feedback (DFB) lasers [40], where the higher quality factor
lasing mode is selected through the symmetry mismatch to the free-space modes [41]. The
semiclassical laser equations for such structures are simulated with the two theoretical
methods described in Chapter 2. Our simulations show that, for an exemplary four-level
gain model, the excitation of dark Fano resonances featuring arbitrarily large quality factors
(due to symmetry mismatch to free-space modes) can lead to a significant reduction of the
lasing threshold of PCSELs with respect to conventional vertical-cavity surface-emitting
lasers.
In Chapter 4, we propose a larger-area single-mode PCSEL by tuning the regular lasing bandedges of quadratic dispersions in typical PCSELs to form a single accidental Dirac cone of linear dispersion at the Brillouin zone center [42, 43]. As in all laser cavities, the lasing areas of PCSELs, and hence output powers, are limited by two fundamental constraints. First, the mode spacing decreases as the cavity area increases, which promotes multi-mode lasing. Second, the distributed in-plane feedback localizes the lasing fields to individual sections, which promotes multi-area lasing. Tuning to an accidental Dirac-points not only increases the mode spacing by orders of magnitudes but also eliminates the distributed in-plane feedback, turning the periodic index-modulated cavities into equivalent Fabry-Perot-like cavities where the modes have different out-of-plane coupling losses. Both advantages promise single-mode PCSELs of larger areas and thus higher output powers. In slab devices, an additional frequency-locking phenomenon at the degenerate point, with potentially high density of states, is analyzed.

Chapter 5 demonstrates and distinguishes experimentally the existence of the dark Fano resonances first discussed in Chapter 3, at $k\approx 0$, in a macroscopic two-dimensional photonic crystal slab [44]. In particular, we fabricate a square array of holes in silicon nitride layer and perform an angular resolved spectral analysis of the various Fano resonances. We characterize the passive photonic crystal slab in terms of its resonant frequencies and radiation behaviors (quality factors) using temporal coupled-mode theory and symmetry considerations. The unique simplicity of this system, whereby an ultra-long lifetime delocalized electromagnetic field can exist above the surface and consequently easily interact with added matter, provides exciting new opportunities for the study of light and matter interaction. We then realize lasing at a dark Fano resonance of the slab structure using diluted solutions of R6G molecules as the gain medium.

In Chapter 6, we turn our attention to the organic dye lasers whose high tunability in the visible wavelengths have attracted interests for many years due to their low-cost processing, flexible choice of substrates, and large emission cross sections that can cover the spectral region from ultraviolet to the near infrared. We investigate the laser dynamics in systems of sub-wavelength photonic structures consisting of organic dye molecules. To this end, we have extended the four-level system of Chapter 2 and developed a more involved model of the levels participating in lasing to describe the interaction of organic molecules with a microstructured cavity, to achieve single-mode lasing. The formalism provides explicit analytic
expressions of the threshold and slope efficiency that characterize this class of lasers, and also the duration over which lasing action can be sustained before the dye photobleaches. Both the chemical properties of the dyes and optical properties of the cavities are considered in the model. We verified this theoretical model by comparing the numerically predicted threshold and slope efficiency values to those measured under short pulse excitations in Chapter 5. We also systematically studied the feasibility of lasing under continuous-wave excitations in optically pumped monolithic organic dye lasers. This study suggests routes to realize an organic laser that can potentially lase with a threshold of only a few $W/cm^2$. In addition to their fundamental scientific interest, this work provide formalisms that could enable the development and advancement of sub-wavelength structured organic-based light emitting and sensing devices.

Lastly, in Chapter 7, we investigate far-infrared ($\sim 0.2 - 2$ THz) laser emission from optically-pumped rotationally excited molecular gases confined in a metallic cavity [45]. This class of lasers is usually referred to as optically-pumped far infrared (OPFIR) lasers. In comparison with visible or infrared waves, terahertz radiation can penetrate into organic materials such as skin, plastics, paper, or cloth and does not cause any damage associated with ionizing radiation (e.g. X-rays) where the photon energy is high. These properties make terahertz radiation useful in packaging inspection for quality control, chemical composition analysis, and security screening. Submillimeter spatial resolution imaging and incredibly specific molecular recognition are other compelling uses for terahertz radiation. To apply terahertz radiation beyond laboratory or close range ($< 10$ m) applications, more powerful ($> 100$ mW) and efficient sources are required to see through highly attenuating obscurants, including the atmosphere itself. FIR molecular gas lasers have already shown its potential to be more powerful than other optically-driven sources [46]. The physical mechanism underlying the operation of conventional OPFIR lasers has been extensively studied in past decades [47–60]. The fundamental limitations in their performances (low efficiency, poor tunability, and limited capability of integration into a compact platform) reside in the molecular gas physics due to the so-called vibrational bottleneck. We seek to overcome these challenges through novel optical designs of the waveguide and feedback structures. To undertake this task, we generalize previous works to allow for a realistic description of the spatio-temporal dynamics characterizing the molecular collisional and diffusion processes. In particular, the model considers the active role that high-energy vibrational states play.
in the laser operation [51] (previous approaches assumed that the populations of those vibrational states were negligible). The theoretical analysis presented in this work expands the current understanding of lasing action in OPFIR lasers and, thus, could contribute to the development of a new class of far-infrared and terahertz sources able to operate efficiently at room temperature. The advent of quantum cascade lasers to replace CO$_2$ pump lasers may combine to produce truly compact submillimeter-wave laser sources in the near future.
Chapter 2

Methods for analyzing lasing action

This chapter discusses the computational methods used to solve the semiclassical laser equations. We begin by modeling the interaction of light with an arbitrary material system, in order to achieve new understanding of the principles involved in light-matter interaction and to theoretically analyze lasing action in these structures. In particular, we couple the rate equations to Maxwell’s equations and implement them into the electromagnetic (EM) model using two approaches: exact finite-difference time-domain (FDTD) calculations \[9,19,37,38\] and a semi-analytic coupled-mode formalism \[33,39\]. Each of these offers different kinds of insight into the system. Before proceeding with the description of the electrodynamic techniques in Sec. 2.2 and 2.3, we first present the general theoretical framework from which these techniques are developed.

2.1 General framework

We model a dispersive Lorentz active medium using a generic four-level atomic system \[9,19,61\]. The population density in each level is given by \(N_i\) \((i = 0, 1, 2, 3)\). Maxwell’s equations for isotropic media are given by \(\partial \mathbf{B}(r, t)/\partial t = -\nabla \times \mathbf{E}(r, t)\) and \(\partial \mathbf{D}(r, t)/\partial t = \nabla \times \mathbf{H}(r, t)\), where \(\mathbf{B}(r, t) = \mu \mu_0 \mathbf{H}(r, t)\), \(\mathbf{D}(r, t) = \epsilon \epsilon_0 \mathbf{E}(r, t) + \mathbf{P}(r, t)\) and \(\mathbf{P}(r, t)\) is the dispersive electric polarization density that corresponds to the transitions between two atomic levels, \(N_1\) and \(N_2\). The vector \(\mathbf{P}\) introduces gain in Maxwell’s equation and its time evolution can be shown \[62\] to follow that of a homogeneously broadened Lorentzian oscillator driven by the coupling between the population inversion and the external electric
field. Thus, $\mathbf{P}$ obeys the equation of motion

$$\frac{\partial^2 \mathbf{P}(\mathbf{r}, t)}{\partial t^2} + \Gamma_m \frac{\partial \mathbf{P}(\mathbf{r}, t)}{\partial t} + \omega_m^2 \mathbf{P}(\mathbf{r}, t) = -\sigma_m \Delta N(\mathbf{r}, t) \mathbf{E}(\mathbf{r}, t)$$  \hspace{1cm} (2.1)$$

where $\Gamma_m$ stands for the linewidth of the atomic transitions at $\omega_m = (E_2 - E_1)/\hbar$, and accounts for both the nonradiative energy decay rate, as well as dephasing processes that arise from incoherently driven polarizations. $E_1$ and $E_2$ correspond to the energies of $N_1$ and $N_2$, respectively. $\sigma_m$ is the coupling strength of $\mathbf{P}$ to the external electric field and $\Delta N(\mathbf{r}, t) = N_2(\mathbf{r}, t) - N_1(\mathbf{r}, t)$ is the population inversion driving $\mathbf{P}$. Positive inversion is attained when $\Delta N(\mathbf{r}, t) > 0$, in which case the medium is amplifying; when $\Delta N(\mathbf{r}, t) < 0$, the medium is absorbing. In order to model realistic gain media, only conditions favorable to the former are considered.

The atomic population densities obey the rate equations

$$\frac{\partial N_3(\mathbf{r}, t)}{\partial t} = R_p N_0(\mathbf{r}, t) - \frac{N_3(\mathbf{r}, t)}{\tau_{32}}$$  \hspace{1cm} (2.2)$$

$$\frac{\partial N_2(\mathbf{r}, t)}{\partial t} = \frac{1}{\hbar \omega_m} \mathbf{E}(\mathbf{r}, t) \cdot \frac{\partial \mathbf{P}(\mathbf{r}, t)}{\partial t} + \frac{N_3(\mathbf{r}, t)}{\tau_{32}} - \frac{N_2(\mathbf{r}, t)}{\tau_{21}}$$  \hspace{1cm} (2.3)$$

$$\frac{\partial N_1(\mathbf{r}, t)}{\partial t} = -\frac{1}{\hbar \omega_m} \mathbf{E}(\mathbf{r}, t) \cdot \frac{\partial \mathbf{P}(\mathbf{r}, t)}{\partial t} + \frac{N_2(\mathbf{r}, t)}{\tau_{21}} - \frac{N_1(\mathbf{r}, t)}{\tau_{10}}$$  \hspace{1cm} (2.4)$$

$$\frac{\partial N_0(\mathbf{r}, t)}{\partial t} = -R_p N_0(\mathbf{r}, t) + \frac{N_1(\mathbf{r}, t)}{\tau_{10}},$$  \hspace{1cm} (2.5)$$

where $\pm \frac{1}{\hbar \omega_m} \mathbf{E} \cdot \frac{\partial \mathbf{P}}{\partial t}$ are the stimulated emission rates. For $\Delta N(\mathbf{r}, t) > 0$, the plus sign corresponds to radiation while the minus sign represents excitation. $R_p$ is the external pumping rate that transfers electrons from the ground state to the third excited level, and is proportional to the incident pump power. $\tau_{ij}$ is the nonradiative decay lifetimes from level $i$ to $j$ ($i > j$) so that the energy associated with the decay term $\frac{N_i}{\tau_{ij}}$ is considered to be lost. Since the total electron density $N_{\text{tot}} = \sum N_i(\mathbf{r}, t)$ is conserved in the rate equations, the active medium modeled this way becomes saturable at high values of $R_p$.

Lasing action in this class of active media is obtained as follows. Electrons are pumped from the ground-state level $N_0$ to $N_3$ at a rate $R_p$. These electrons then decay nonradiately into $N_2$ after a short lifetime $\tau_{32}$. By enforcing $\tau_{21} \gg (\tau_{32}, \tau_{10})$, a metastable state is formed at $N_2$ favoring a positive population inversion between $N_2$ and $N_1$ (i.e. $\Delta N > 0$), which are separated by energy $\hbar \omega_m$. In this regime, a net decay of electrons to $N_1$ occurs through
stimulated emission and nonradiative relaxation. Lastly, electrons decay nonradiatively and quickly to \( N_0 \). Lasing occurs for pumping rates beyond a threshold \( R_{th} \), once sufficient inversion (gain) is attained to overcome the total losses in the structure. In reality, some of these \( N_i \) levels may stand for clusters of closely spaced but distinct levels, where the relaxation processes among them are much faster than that with all other levels.

2.2 Finite-difference time-domain simulations of active media

The finite-difference time-domain method [37] can be used to numerically solve for the optical response of the active material. Similar to the derivation of Yee [63], Maxwell's equations are approximated by the second order center differencing scheme so that both three dimensional (3D) space and time are discretized, leading to spatial and temporal interleaving of the electromagnetic fields. Pixels of the Yee lattice are chosen to be smaller than the characteristic wavelength of the fields. Moreover, this uniform space grid (i.e. \( \Delta x = \Delta y = \Delta z \), where \( \Delta x \), \( \Delta y \) and \( \Delta z \) are the space increments in the \( x \), \( y \) and \( z \) directions) should also be made fine enough so that the optical structures in consideration are well-represented. For numerical stability, Von Neumann analysis places an upper bound on the size of the time step, \( \Delta t \leq S\Delta x/c \), where \( c \) is the speed of light. The Courant factor \( S \) is typically chosen to be \( \frac{1}{2} \) to accommodate 3D simulations.

Following Yee, we evolve the \( E \) and \( H \) fields at alternate time steps. For simplicity, we show explicitly the set of discretized equations implemented for a one dimensional (1D) setup assuming a non-magnetic and isotropic medium, and denote any functions of space and time as \( F^n(i) = F(i\Delta z, n\Delta t) \). \( H \) is first updated as

\[
B_{y}^{n+1/2}(i + \frac{1}{2}) = B_{y}^{n-1/2}(i + \frac{1}{2}) - \frac{\Delta t}{\Delta z} \left[ E_{x}^{n}(i + 1) - E_{x}^{n}(i) \right] \quad (2.6)
\]

\[
H_{y}^{n+1/2}(i + \frac{1}{2}) = \frac{1}{\mu_o}B_{y}^{n+1/2}(i + \frac{1}{2}) \quad (2.7)
\]

Next, we update the polarization density \( P \) at \( n + 1 \) from the two previous instances of \( P \), and the previous \( N_i \) and \( E \) according to Eq. (2.1). Note that components of \( P \) reside at the
same locations as those of $E$.

$$
P_x^{n+1}(i) = (1 + \Gamma_m \Delta t/2)^{-1} \left\{ \left(2 - \omega_m^2 \Delta t^2 \right) P_x^n(i) + (\Gamma_m \Delta t/2 - 1) P_x^{n-1}(i) - \Delta t^2 \sigma_m \left[ N_2^n(i) - N_1^n(i) \right] E_x^n(i) \right\} \quad (2.8)
$$

We can then use these updated $H$ and $P$ values to retrieve $E$ at $n + 1$:

$$
D_x^{n+1}(i) = D_x^n(i) - \frac{\Delta t}{\Delta z} \left[ H_y^{n+1/2}(i + \frac{1}{2}) - H_y^{n+1/2}(i - \frac{1}{2}) \right] \quad (2.9)
$$

$$
E_x^{n+1}(i) = \frac{1}{\epsilon(i) \epsilon_o} \left[ D_x^{n+1}(i) - P_x^{n+1}(i) \right] \quad (2.10)
$$

Lastly, $N_i$ at $n + 1$ requires $N_i$ at $n$ and both the previous and updated $E$ and $P$ values at $n$ and $n + 1$. Since the population densities of the four levels are interdependent in Eq. (2.2) to (2.5), they must be solved simultaneously by setting up the following system of equations:

$$
\tilde{A} N^{n+1}(i) = \tilde{B} N^n(i) + C \quad (2.11)
$$

where

$$
N^{n+1}(i) = \begin{bmatrix} N_0^{n+1}(i) \\ N_1^{n+1}(i) \\ N_2^{n+1}(i) \\ N_3^{n+1}(i) \end{bmatrix}, \quad N^n(i) = \begin{bmatrix} N_0^n(i) \\ N_1^n(i) \\ N_2^n(i) \\ N_3^n(i) \end{bmatrix}, \quad C = \begin{bmatrix} 0 \\ -E P \\ E P \\ 0 \end{bmatrix},
$$

$$
E P = (2 \hbar \omega_m)^{-1} \left\{ \left[ E_x^{n+1}(i) + E_x^n(i) \right] \times \left[ P_x^{n+1}(i) - P_x^n(i) \right] \right\},
$$

$$
\tilde{A} = \begin{bmatrix} 1 + e_1 & -e_2 & 0 & 0 \\ 0 & 1 + e_2 & -e_3 & 0 \\ 0 & 0 & 1 + e_3 & -e_4 \\ -e_1 & 0 & 0 & 1 + e_4 \end{bmatrix}, \quad \tilde{B} = \begin{bmatrix} 1 - e_1 & e_2 & 0 & 0 \\ 0 & 1 - e_2 & e_3 & 0 \\ 0 & 0 & 1 - e_3 & e_4 \\ e_1 & 0 & 0 & 1 - e_4 \end{bmatrix},
$$

$$
e_1 = \frac{\Delta t \tau_0}{2}, \quad e_2 = \frac{\Delta t}{2 \tau_{10}}, \quad e_3 = \frac{\Delta t}{2 \tau_{21}}, \quad e_4 = \frac{\Delta t}{2 \tau_{32}}.
$$

$\tilde{A}$ and $\tilde{B}$ are tensors that couple the populations in the four atomic levels, and updated $N_i$ values at $n + 1$ in Eq. (2.11) can be computed by inverting $\tilde{A}$. Note that the atomic population density $N_i$ is a scalar and depends generally on all three components of $E$ and $P$. The above cycle is repeated at each time step until steady state is reached, allowing the
full temporal development of the laser mode to be tracked.

In this FDTD scheme with auxiliary differential equations for \( P \) and \( N_i \), all physical quantities of the active material are tracked at all points in the computational domain and at all times. The numerical results computed in this \textit{ab initio} way are exact apart from discretization of space-time, and allow for nonlinear interactions between the media and the fields. In our simulations, the electric, magnetic and polarization fields are initialized to zero except for background noise while the total electron density is initialized to the ground state level. The computational domain is truncated with Bloch periodic boundary conditions or perfectly matched layers, which are artificial absorbing material designed so that the computational grid’s boundaries are reflectionless in the limit \( \Delta z \to 0 \) [64]. The resolution is consistently set to 20 for every lattice constant \( a \) throughout the thesis, where we checked that the relative differences in frequency and \( Q \) values between the 20 pixels per \( a \) case and the 40 pixels per \( a \) case is less than 5%.

2.3 Coupled-mode theory formalism applied to lasing media

Coupled-mode theory (CMT) [33,39] has been extensively used to study a broad range of different problems in photonics, both in the linear and nonlinear regimes [65–71]. Here, for the first time to our knowledge, we extend this class of analysis to the case of micro-photonic structures that include active media.

Consider first an arbitrary gain medium embedded in an EM cavity with a single lasing mode\(^1\). The power input to this system, \( P_{in} \), can be seen as the result of the work performed by the current induced in the system by the active media, \( J(r,t) \), against the electric field of the cavity, \( E(r,t) \). Noticing that \( J(r,t) \) actually comes from the temporal variation of the polarization density, i.e. \( J(r,t) = \partial P(r,t)/\partial t \) [see definition of \( P \) in Eq. (2.1)], \( P_{in} \) can be written as [72]

\[
P_{in} = \frac{1}{2} \text{Re} \left\{ \int \text{d}r \left[ \frac{\partial P(r,t)}{\partial t} \right] E^*(r,t) \right\} \tag{2.12}
\]

Now, we assume that the spatial and temporal dependence in both the electric field and the polarization density can be separated as \( E(r,t) = E_0(r)a(t)\exp(-i\omega_c t) \) and \( P(r,t) = \frac{1}{2} \text{Re} \left\{ \int \text{d}r \left[ \frac{\partial P(r,t)}{\partial t} \right] E^*(r,t) \right\} \)

\(^1\)Although the generalization of the approach described here to the case where more than one mode is lasing is straightforward, for simplicity, we only consider the single-mode case in this paper.
\[ E_0(r)P(t) \exp(-i\omega_m t), \] respectively. \( E_0(r) \) is the normalized cavity mode profile where
\[ \int \text{dr} \epsilon_0 n^2(r) |E_0(r)|^2 = 1, \]
and \( a(t) \) is the corresponding slowly-varying wave amplitude normalized so that \( |a(t)|^2 \) is the energy stored in the resonant mode [39]. \( \omega_c \) and \( \omega_m \) stand for the resonant frequency of the cavity and the considered atomic transition [see Eq. (2.1)], respectively.

If we further assume that all the power emitted by that system is collected by a waveguide evanescently coupled to the cavity, and that \( \omega_c = \omega_m \), using first order perturbation theory in Maxwell’s equations, one can show from energy conservation arguments [39] that the temporal evolution of the electric field amplitude \( a(t) \) is governed by the equation

\[
\frac{da(t)}{dt} = - \left( \frac{1}{\tau_{1O}} + \frac{1}{\tau_{ex}} \right) a(t) + \xi_1 \left[ i \omega_m P(t) - \frac{dP(t)}{dt} \right],
\]

(2.13)

where \( \tau_{ex} \) and \( \tau_{1O} \) are, respectively, the decay rates due to external losses (mainly absorption and radiation losses) and due to the decay into the waveguide. The confinement factor
\[ \xi_1 = (1/2) \int_A \text{dr} |E_0(r)|^2, \]
where \( A \) denotes the active part of the structure, accounts for the fact that only the active region drives the temporal evolution of the cavity mode amplitude [as seen in Eq. (2.13)].

We now turn to the analysis of the polarization amplitude \( P(t) \). Within the first-order perturbation theory approach we are describing here, one can obtain a simple first-order differential equation from Eq. (2.1) for the temporal evolution of \( P(t) \) by making three main assumptions: (i) we assume that the linewidth of the atomic transition is much smaller than its frequency, i.e. \( \Gamma_m \ll \omega_m \); (ii) we apply the rotating-wave approximation (RWA), i.e. we consider just the terms that oscillate as \( \exp(\pm i\omega_m t) \); and, (iii) we apply the slowly-varying envelope approximation (SVEA) [62], i.e. we assume \( d^2P(t)/dt^2 \ll \omega_m dP(t)/dt \). For the structures that we will consider in the next chapter, these approximations give good agreement between CMT and FDTD.

Thus, the equation of motion for \( P(t) \) is given by

\[
\frac{dP(t)}{dt} + \frac{\Gamma_m}{2} P(t) = - \frac{i\sigma_m}{2\omega_m} a(t) \langle \Delta N(t) \rangle \]

(2.14)

Here \( \langle \Delta N(t) \rangle \) is defined as the population inversion \( \Delta N(r, t) \) averaged over the mode profile.
in the gain region of the system

\[ \langle \Delta N(t) \rangle = \frac{\int_A dr \ |E_0(r)|^2 \Delta N(r, t)}{\int_A dr \ |E_0(r)|^2} \quad (2.15) \]

Notice also that in Eq. (2.14), the definitions of \( \sigma_m \) and \( \omega_m \) are the same as those given in Eq. (2.1).

Finally, by projecting Eqs. (2.2)–(2.5) onto the cavity mode profile intensity \( |E_0(r)|^2 \) over the active region \( A \), following a derivation similar to that given above and after some straightforward algebra, one can obtain the following set of equations governing the time evolution of the average population densities \( \langle N_i(t) \rangle \) (using the same definition of average as in Eq. (2.15), and with \( i=0,...,3 \))

\[
\frac{d\langle N_3(t) \rangle}{dt} = R_p \langle N_0(t) \rangle - \frac{\langle N_3(t) \rangle}{\tau_{32}} \tag{2.16}
\]

\[
\frac{d\langle N_2(t) \rangle}{dt} = \frac{1}{4\hbar} \xi_2 \left\{ a(t) \left[ iP^*(t) + \frac{1}{\omega_m} \frac{dP^*(t)}{dt} \right] + c.c. \right\} + \frac{\langle N_3(t) \rangle}{\tau_{32}} - \frac{\langle N_2(t) \rangle}{\tau_{21}} \tag{2.17}
\]

\[
\frac{d\langle N_1(t) \rangle}{dt} = -\frac{1}{4\hbar} \xi_2 \left\{ a(t) \left[ iP^*(t) + \frac{1}{\omega_m} \frac{dP^*(t)}{dt} \right] + c.c. \right\} + \frac{\langle N_2(t) \rangle}{\tau_{21}} - \frac{\langle N_1(t) \rangle}{\tau_{10}} \tag{2.18}
\]

\[
\frac{d\langle N_0(t) \rangle}{dt} = -R_p \langle N_0(t) \rangle + \frac{\langle N_1(t) \rangle}{\tau_{10}} \tag{2.19}
\]

where the parameter \( \xi_2 \) is given by \( \xi_2 = (2 \int_A dr \ |E_0(r)|^4)/(\int_A dr \ |E_0(r)|^2) \). Thus, starting from the knowledge of the electric field profile of the resonant cavity \( E_0(r) \), and their corresponding decay rates, one can compute all the relevant physical quantities characterizing lasing action in any structure just by solving the linear system of first-order differential equations given by Eqs. (2.13),(2.14),(2.15)–(2.19). In particular, once such system of equations have been solved, the total emitted power can be easily computed from \( P_e(t) = 2|a(t)|^2/\tau_{10} \).

The formalism presented here also allows us to explicitly retrieve analytic expressions for the lasing threshold and the slope in the region near threshold, with the spatial contents of the setup entirely embedded in \( \xi_1 \) and \( \xi_2 \). For the four-level system considered, assuming \( \tau_{10}, \tau_{32} \ll \tau_{21} \) and that \( 1/\tau_{21} \gg R_p \) at steady state, the threshold population inversion is

\[
\langle \Delta N \rangle_{th} = \frac{\Gamma_m}{\sigma_m \tau_{tot} \xi_1} \tag{2.20}
\]

where \( \tau_{tot} \) is defined so that \( 1/\tau_{tot} = 1/\tau_{10} + 1/\tau_{ex} \) and that all time dependences have been
dropped since we are considering the steady state. Correspondingly, the pumping threshold is found to be

\[ R_{th}^p = \frac{\Gamma_m}{\sigma_m \tau_{tot} \xi_1 \tau_{21} \langle N_{tot} \rangle} \]  \hspace{1cm} (2.21)

where \( \langle N_{tot} \rangle = \Sigma_{i=0}^{3} \langle N_i \rangle \). Eq. (2.21) may be related to the threshold power by \( P_{th} = R_{th}^p \hbar \omega \int_A d\mathbf{r} \langle N_{tot} \rangle \). Finally, we define the slope in this paper as the ratio of the emitted power to the pumping rate. Our model predicts that

\[ \frac{dP_e}{dR_p} = \frac{4 \hbar \omega_m \langle N_{tot} \rangle \xi_1}{\xi_2} = \langle N_{tot} \rangle \hbar \omega_m \frac{\left( \int_A d\mathbf{r} |E_0(\mathbf{r})|^2 \right)^2}{\int_A d\mathbf{r} |E_0(\mathbf{r})|^4} \]  \hspace{1cm} (2.22)

Steady state predictions provided by Eq. (2.20), (2.21), (2.22) match with results commonly derived in textbooks [73] for a similar four-level system, in the case where the following are assumed in our present model: (i) spatially uniform field (ii) only the loss channel related to the cavity’s Q is present, i.e. \( \tau_{tot} = \tau_{TO} \) (iii) the gain medium fills the entire cavity (iv) \( \sigma_m = \epsilon_m \epsilon_0 \lambda^2 \omega_m / 4 \pi^2 \tau_{spont} \) as derived in [62], where \( \tau_{spont} \) is the radiative spontaneous lifetime of the lasing transition (i.e. between \( N_1 \) and \( N_2 \) in our four-level system).
Chapter 3

Lasing action in photonic crystal slabs enabled by Fano resonances

We describe the passive properties of the photonic crystal surface-emitting lasers (PCSELS) and compare them to those of conventional vertical-cavity surface-emitting lasers (VCSELS). We then present a theoretical analysis of lasing action in PCSELS. The semiclassical laser equations for such structures are simulated with the two theoretical methods described in Chapter 2. Our simulations show that, for an exemplary four-level gain model, the excitation of dark Fano resonances featuring arbitrarily large quality factors can lead to a significant reduction of the lasing threshold of PCSELS. In addition to their fundamental interest, these findings may affect further engineering of active devices based on photonic crystal slabs.

3.1 Introduction

The unique properties of photonic crystals (PhCs) to achieve simultaneous spectral and spatial electromagnetic (EM) mode engineering can be exploited in two different ways. On one hand, one can take advantage of the extremely high $Q/V_{\text{mode}}$ microphotonic cavities ($Q$ and $V_{\text{mode}}$ being the corresponding quality factor and the modal volume, respectively) that can be introduced in PhCs simply by inducing local variations in the geometry or the dielectric constant of an otherwise perfectly periodic structure [33, 74]. Alternatively, one can benefit from the multidirectional distributed feedback effect occurring at frequencies close to the band-edges in a defect-free PhC slab; this enables coherent lasing emission.
and polarization control over large areas [10]. In this work, we focus on the latter class of structures, usually referred to as photonic crystal surface-emitting lasers [26].

One of the most intriguing aspects of periodic PhC slabs is the presence of guided resonances, the so-called Fano resonances [33,34,75]. The physical origin of these resonances lies in the coupling between the guided modes supported by the slab and external plane waves, this coupling being assisted by the Bragg diffraction occurring in the considered structures due to the periodic modulation of their dielectric constant. Importantly, in the case of an infinitely perfectly periodic PhC slab, and essentially due to symmetry reasons [41], some of these Fano resonances are completely de-coupled from the external world (i.e. their $Q$-factor diverges despite lying above the light line). As we show later in the chapter, in actual finite-size PhC slabs, these dark Fano resonances retain some of the properties of their infinite periodic counterparts and, thus, in the limit of large size PhC slabs, can display arbitrarily large $Q$ values. Since these dark modes typically have photon lifetimes much longer than those of other modes, we expect them to dominate the lasing properties of PCSELs.

The purpose of this chapter is to analyze theoretically how lasing action in PCSELs is influenced by the presence of the dark Fano resonances, using the finite-difference time-domain method and semi-analytical coupled-mode theory of Chapter 2. Each of these techniques offers different kinds of insight into the systems. Our results suggest that the physical origin of the low lasing thresholds observed in actual PCSELs, compared to conventional vertical-cavity-surface-emitting lasers (VCSELs) [76], can be explained in terms of dark Fano states. In comparison with previous work in this area [10,12,22,26,31], the findings reported in this chapter expand the current understanding of lasing action in PCSELs, which, to our knowledge, has hitherto been explained only in terms of the low group velocity band-edge modes supported by this class of lasers.

3.2 Passive properties

3.2.1 Band diagrams of infinite periodic structures

We begin by studying the dispersion relations of the three PhC lasing structures shown in the insets of Fig. 3-1: a 1D cavity structure with 1D periodicity, a 2D slab structure with 1D periodicity, and a 3D slab structure with 2D periodicity (Fig. 3-1(a), (b) and
(c) respectively). In all three cases, the corresponding dispersion relations were computed through FDTD calculations by setting up a unit cell of the PhC and imposing perfectly matched layer (PML) absorbing boundary conditions on the top and bottom surfaces of the computational domain. Bloch periodic boundary conditions on the electric fields were imposed on the remaining surfaces perpendicular to the slabs.

Figure 3-1(a) illustrates the band diagram of a structure resembling a conventional VCSEL [76], which extends uniformly to infinity in the x and z directions. In this system, a one-wavelength thick cavity with \( n = 3.55 \) (e.g. as in InGaAsP) is enclosed by 25 and 30 bilayers of quarter-wave distributed Bragg reflectors (DBRs) on the top and bottom sides of the structure, respectively. These dielectric mirrors consist of alternate layers of material with \( n = 3.17 \) and \( n = 3.51 \) (e.g. as in InP and the InP-based lattice-matched InGaAlAs, which offers a relatively larger refractive index contrast of \( \Delta n = 0.34 \) at 1.55 \( \mu \text{m} \) wavelength; allowing broadband, high reflectivity and low penetration depth DBRs to be attained with fewer layers). Pink shaded regions represent the continuum of bands guided in the DBRs, while the red line represents the air light line that separates the modes that are propagating in air from those that are evanescent in air. Only transverse magnetic (TM) modes with electric field oriented along the z direction are considered.

The lowest guided mode [green line in Fig. 3-1(a)] is bounded by the light line of the \( n = 3.55 \) center layer (not shown) and that of the multilayer cladding (bottom edge of the lower continuum region). Thus, this mode is guided within the cavity layer via total internal refraction, just as in regular dielectric waveguide slab, with no means of coupling to air. It is the portion of the second mode [blue line in Fig. 3-1(a)] which lies above the air light line that is useful for laser operation. In fact, it is most often desirable to operate at the frequency that corresponds to \( k_x = 0 \) (the \( \Gamma \) point) so that the power is vertically emitted through the surface in the longitudinal (y) direction. This mode resides in the lowest photonic bandgap of the periodic claddings and, therefore, is trapped within the cavity layer by the high reflectivities (> 96%) of the DBRs. From our calculations, we find that \( Q \), which measures the radiation loss of the VCSEL in the y direction, is 7500 at \( k_x = 0 \) and may generally be increased further by adding more bilayers of the claddings. Thus, VCSEL structures similar to the one described here, resemble a conventional laser cavity in which the eigenmodes are formed in the longitudinal direction due to feedback from the dielectric mirrors and in which the number of the modes increases with the cavity.
thickness. Notice that the group velocity \( (v_g = d\omega/dk_x) \) is near zero for small values of \( k_x \). This not only provides good lateral modal confinement, but also maximizes the wave-matter interaction inside the cavity.

Fig. 3-1(b) and (c) render the dispersion relations of *air-bridge* type PhC slabs with 1D corrugation and punctured 2D square lattice of air cylinders respectively. These PhC slabs can support Fano resonances; the resonances form when periodic air perturbations, introduced in an otherwise homogeneous slab, enable the coupling between the guided modes supported by the slab and the external radiation continuum. The strength of this coupling is measured by \( Q \) of the slab structures. One major difference between these PhC slabs and VCSEL-like structures is that in the former, light confinement occurs in the in-plane periodic directions due to Bragg diffractions and in the out-of-plane direction due to index guiding. It is this presence of index guiding that limits the photon lifetime at frequencies above the air light line, leading to far-field radiation. Since discrete translational symmetries exist due to in-plane periodicity, the projected band diagrams are plotted with respect to the lateral wave vectors along the irreducible Brillouin zone. We shall briefly examine the geometries of the two slab structures separately, before drawing the similarities between them when operated as band-edge mode lasers.

The 2D PhC slab sketched in the inset of Fig. 3-1(b) consists of a 0.3a-thick \( n = 3.17 \) slab with a set of 1D periodic grooves along the \( x \)-direction. These grooves are 0.15a deep and 0.1a wide, and extend uniformly in the \( z \) direction. Only modes with electric field oriented along \( z \) are considered. On the other hand, the PhC slab shown in inset of Fig. 3-1(c) consist of a 0.3a-thick \( n = 3.17 \) slab punctured with a 2D square lattice of circular air cylinders in the lateral directions, with both depth and radius being equal to 0.25a. In this case, only transverse-electric-like (TE-like) modes, with the electric fields primarily horizontal near the center of the slab, are excited. As in the case of the VCSEL, the modes above the light line at \( \Gamma \) are the most desirable for lasing since they allow the power to be coupled vertically out of the slab surface. Moreover, in this structure, the zero in-plane group velocity facilitates formation of standing waves, as in any conventional cavity, leading to in-plane feedback of the eigenmodes. Note that a VCSEL, on the other hand, has the same direction of periodicity, feedback, and power emission. In fact, in the finite size slab devices that we will consider next, \( \Delta k_|| \neq 0 \) so that the dispersion curves near \( \Gamma \) may be well approximated by the second order Taylor expansion, in which case, \( v_g \) becomes directly
Figure 3-1: Band diagrams of 1D, 2D, 3D systems, illustrating zero group velocity at $k_{||} = 0(2\pi/a)$. The light lines $\omega = ck_{||}$ (red) separate the modes that are oscillatory ($\omega > ck_{||}$) in the air regions from those that are evanescent ($\omega < ck_{||}$) in air. (a) TM band diagram of a 1D system: Cavity enclosed by 25 and 30 bilayers (on top and below, respectively) of quarter-wave distributed Bragg reflectors. Pink shaded region represents a continuum of bands corresponding to the guided modes in the DBRs. Green line is the fundamental mode guided via total internal refraction while blue line is the mode guided within the band gap of the DBRs. Only modes with electric field oriented along $z$ direction are considered. Inset shows the VCSEL structure extending uniformly to infinity in the $x$ and $z$ directions, with a 1-$\lambda$ thick $n = 3.55$ cavity layer (green). Alternate red and blue layers of the reflectors correspond to $n = 3.17$ and $n = 3.51$ respectively. (b) Band diagram of a 2D system: $n = 3.17$ slab of height 0.3$a$ with 1D periodic grooves that are 0.15$a$ deep along $y$ and 0.1$a$ wide along $x$. Only modes with electric field oriented along $z$ direction are considered. Inset shows the structure, which is periodic in the $x$ direction and extends uniformly in the $z$ direction. (c) Band diagram of a 3D system: $n = 3.17$ slab of eight 0.3$a$ with square lattices of circular air cylinders whose depth and radius are 0.25$a$. Only TE-like modes are considered. Inset shows the slab structure, which is periodic in $x$ and $y$ directions.
proportional to the curvature of the bands. Flat dispersion curves having high density of photonic states and low $v_g$ are favorable for enhancing light-matter interaction, which is essential for lasing to take place.

The first set of modes at $\Gamma$ are ideal for orthogonal out-of-plane surface emission lasers [12]. These are the two band-edge modes shown in Fig. 3-1(b) and the four band-edge modes shown in Fig. 3-1(c). The former corresponds to the phase matching condition $k^d_x = k^i_x + qK$, where $k^d_x$ and $k^i_x$ are the diffracted and incident wave vectors respectively, $K = 2\pi/a$ is the Bragg grating vector, and we only consider $q = 1$ to ensure vertical outcoupling. All other higher lying frequency modes result in additional out-of-plane emission directions at oblique angles from the slab surfaces. For the corrugated slab, the phase matching conditions in the reciprocal space also implies that the waves traveling in the $+x$ direction are coupled to those in the $-x$ direction within each unit cell, forming an in-plane feedback mechanism similar to a 1D cavity. These lateral standing waves are in turn coupled into $y$ because the Bragg condition is also satisfied along the slab normal to enable out-of-plane surface emission. For the slab shown in Fig. 3-1(c), phase matching at $\Gamma$ couples waves in the four equivalent $\Gamma - X$ directions of a unit cell to the waves emitting in $z$. Here, the main feedback mechanism is provided separately by waves traveling in the $\pm x$ and $\pm y$ directions. Further coupling of waves between these orthogonal directions is facilitated by higher order waves traveling in the $\Gamma - M$ directions (see inset of Fig. 3-1(c) for the definitions of directions in the reciprocal space of a square lattice). Due to the ease of fabrication resulting from the connected nature of the defect-free lattice, as well as other advantages mentioned at the beginning of this section, PhC slab structures hold great potential as laser devices. The key is its ability to excite a single lateral and longitudinal mode over a large 2D lasing area, as a result of the multidimensional distributed feedback mechanism described above. Intuitively, we may treat each unit cell as an individual cavity in-sync with its neighbors to produce coherent laser oscillations, and properties of the lasing mode may be controlled simply by tuning the design of each lattice cell. This approach has been experimentally realized to control the polarization of the lasing mode [10].

In this work, we focus on another property of the PhC slab that allows it to operate as a high $Q$, low-threshold laser: the existence of band-edge modes with infinite photon lifetime (i.e. no means of coupling out of the slab). This phenomenon occurs for the lower band edge in Fig. 3-1(b) and for the singly degenerate modes in 2D periodic PhC slabs, corresponding
to the two lowest band-edge modes at $\Gamma$ in Fig. 3-1(c). The absence of radiative components at these points in the band diagram is a result of in-phase superpositions of the forward and backward traveling waves, with in-plane electric field vectors adding destructively. This same feature can be explained using the symmetry mismatch existing between the guided modes in the PhC slab and the diffracted radiation field in air [41]. We shall reinforce these arguments in the next section based on the electric field profiles of the radiation components. Infinite $Q$ above the air light line can only be achieved in PhC slabs; this property is absent in VCSELs, or conventional microcavity structures that use high reflectivity mirrors for mode trapping.

In order to study the mode trapping capabilities of the slab structures in PCSELs, we first examine in detail the corrugated slab design. This 2D design, though analytically and computationally less demanding, encompasses the same essential physics as a 3D PhC slab realizable in experiments, which we will also study at the end of this section.

Fig. 3-2(a) presents the $Q$ of the two bands above the light line at small values of $k_x$, plotted against frequency, in the vicinity of the bandgap for the slab structure shown in Fig. 3-1(b). We see from the figure that the two band-edge modes differ drastically. The $Q$ of the lower frequency mode diverges rapidly as $k_x \to 0$, while that of the next-ordered band remains finite. This is clearly illustrated by the electric field profiles in the unit cell, depicted in the two leftmost panels of Fig. 3-2(c) for the lower (left) and upper (right) band-edge modes. The unbounded $Q$ mode, whose radiative electric field component is antisymmetric about the groove, interferes destructively with itself in the far-field, resulting in no net outcoupling to air. For $k_x$ away from $\Gamma$, this symmetry mismatch is lost, and $Q$ decreases rapidly but remains large. On the other hand, the second mode is symmetric and vertical emission out of the slab is possible. Note that despite this leakage, most of the electric field is confined within the slab to form a standing wave pattern, producing a signature of Fano resonances. Apart from mode symmetries, the resonances in the slabs are also influenced by the size of the grooves. Results for 1D periodic grooves with depth $0.05a$, $0.1a$ and $0.15a$ are compared in Fig. 3-2(a). Consistent with predictions from the perturbation theory [33], the bandgap decreases with the grooves size while $Q$ increases, approaching the slab waveguide limit of infinity when no grooves are present.
Figure 3-2: (a) Variation of $Q$ as a function of frequency for the lowest two bands above the light line for the infinite slab structure illustrated in Fig. 3-1 (b), as well as two other similar designs where the depth of the grooves are reduced to 0.05a and 0.1a. (b) Variation of $Q$ as a function of frequency for the infinite slab (red lines), and slabs that are finite in the $x$ direction (but remain uniform and infinite in the $z$ direction) with length $L_x$. Depth of the grooves is 0.05a for all slabs considered in (b) and (c). (c) First two insets illustrate the mode profiles of the lower and upper band-edges of the 2D infinite slabs, where only a period of the slab in the $x$-$y$ plane is shown. The lower band edge mode is anti-symmetric about the groove while the upper band edge mode is symmetric. The photonic crystal slab is outlined in green and electric field pointing into the page is depicted with positive (negative) values in red (blue). The two insets on the right show the band-edge modes of the 20a finite slabs. Near the slab's center, the top (bottom) profile shares the same symmetry relative to the groove as the infinite slab's lower (upper) band edge mode.
3.2.2 Finite periodic structures

The symmetry of the lower band-edge mode, which forbids outcoupling, is exact only for the infinite (periodic) structure. In any finite system, the photon lifetime is large but finite. Fig. 3-2(b) shows the $Q$ factor, as a function of frequency, for finite slabs with lateral sizes ranging from 20 to 320 periods. These results were obtained from FDTD calculations, with the boundary of the computational domain padded with absorbing boundary conditions (PMLs) to mimic the behavior of a slab in free space. A couple of key observations are in order: (i) The lower band-edge mode of the finite PhC slab no longer possesses an unbounded $Q$, owing to the fact that an additional loss channel is opened up: energy can now leak from the sides of the slab. This can be observed in the top right panel of Fig. 3-2(c) for a 20$a$ long PhC slab. These lateral losses dominate in the lower band-edge mode. The bottom right panel of Fig. 3-2(c) shows the symmetric mode, where both vertical and lateral power emission appears equally dominant. It is thus no surprise that the net $Q$ of the lower band-edge mode remains higher than that of the symmetric mode [see Fig. 3-2(b)]. (ii) The $Q$ of the lasing structure increases with the number of periods, so the lasing threshold correspondingly decreases. We shall quantify the losses in Fig. 3-3, as functions of the number of periods. (iii) The resonant frequencies of the upper band-edge modes behave differently in the finite and infinite slabs. $k_x$ and the group velocity increases with frequency for the upper band. In the finite system, this leads to a higher radiation leakage from the sides and hence, a decrease in $Q$. In the infinite system, there are no lateral losses and so, $Q$ increases with frequency near the band-edge due to the corresponding reduction in out-of-plane radiation losses. For the lower band-edge, mode symmetry considerations ensure that $Q$ remains a maximum for both the infinite and finite slabs.

Next, we quantify the $Q$ values of the corrugated slab in order to understand how the lateral size of the device, $L_x$, affects the outcoupling of Fano resonances. Fig. 3-3 compiles the total $Q$ ($Q_{tot}$) of the two band-edge modes presented in Fig. 3-2(b) for PCSEL structures having 0.05$a$ deep grooves for $L_x$ ranging from 20$a$ to 320$a$. In order to operate the device at typical optical communication wavelength ($\sim 1.55 \mu m$), we set $a = 675$ nm here and in subsequent results. Since a larger PhC slab provides a longer confinement time, $Q$ increases with $L_x$ for both symmetric and anti-symmetric modes. The anti-symmetric mode has higher $Q$, due to its reduced vertical emission, as already observed in Fig. 3-
Figure 3-3: Total $Q$ of the two band-edge modes for the finite PhC slab punctured with 0.05$a$ deep grooves, and having lateral size, $L_x$, ranging from 20 to 320 unit cells. Green lines are the fitted curves using the relationships described in the text and the horizontal line indicates $Q$ value of the corresponding infinite slab (for the symmetric mode case).

2(c). For $L_x > 100 \mu m$, the total $Q$ of both modes tends towards that of their infinite counterpart [see Fig. 3-2(b)]: $Q_{\text{sym}}$ saturates at 1964, whereas $Q_{\text{anti-sym}}$ is unbounded. Therefore, the anti-symmetric mode holds great potential for low-threshold laser operation. Using approximate analytic relationships that govern $Q$'s dependence on $L_x$ (unique for each mode), curves fitted to the calculated data are also plotted in Fig. 3-3. We specify these relationships in the following paragraph.

To better explore the potential of the PhC slab as a vertical emission laser, we decompose $Q_{\text{tot}}$ into two $Q$ values governing the in-plane ($Q_{\parallel}$) and orthogonal out-of-plane ($Q_{\perp}$) directions. The former is a measure of lateral losses from the sides of the slabs while the latter indicates the degree of vertical emissions. They are related by $1/Q_{\text{tot}} = 1/Q_{\parallel} + 1/Q_{\perp}$.

From $Q_{\parallel} = \omega \tau_{\parallel}/2$ ($\tau_{\parallel}/2$ is the photon lifetime before escaping from the sides), it can be shown that near $\Gamma$, $Q_{\parallel}$ scales approximately with the finite slab's size as $C_1 L_x^2$, where $C_1$ is a constant independent of $L_x$. This scaling may be derived by first quadratically approximating the band near the band-edge as $\omega \propto k_{\parallel}^2$ so that the $v_g = d\omega/dk_{\parallel} \propto k_{\parallel}$. In addition, taking the limit at $\Delta k_{\parallel} \Delta x = C$, where $C$ is a constant and $\Delta x = L_x$ here, it may be concluded that $\Delta k_{\parallel}$ scales as $1/L_x$. This sets the characteristic scale for $k_{\parallel}$ and hence, $v_g \propto 1/L_x$. Thus, together with the distance $L_x$ the photon travels, $\tau_{\parallel}$ scales as
$L^2_z$. Since both band-edge modes possess low group velocity and thus relatively large lateral photon confinement, and experience the same structural interfaces, $Q_\parallel$ behaves in the same manner for both modes. The same does not apply to $Q_\perp$, where modal symmetry mismatch considerations may act to impede outcoupling. To obtain an approximate scaling of $Q_{\text{tot}}$ with $L_x$, we assume $Q_{\text{anti-sym}}$ to remain much larger than $Q_{\text{anti-sym}}$ while $Q_{\text{sym}}$ to be a finite value independent of $L_x$ but related to the groove size. Hence, $Q_{\text{tot}}^{\text{anti-sym}} \sim C_2 L_x^2$ and $Q_{\text{tot}}^{\text{sym}} \sim Q_{\text{periodic}}^{\text{sym}}/(1 + C_3/L_x^2)$, where $Q_{\text{periodic}}^{\text{sym}}$ is the value for the symmetric mode of the corresponding infinite slab. $C_2$ and $C_3$ are constants independent of $L_x$. Curve fitting results shown for $Q_{\text{tot}}$ are made using these relationships and a reasonably good match is achieved. It now becomes clear that great variance of the photon lifetimes for the two modes in Fig. 3.3 arises purely from their $Q_\perp$. We conclude that, for small $L_x$, the PhC slab behaves as an in-plane emitting device; to excite enough of the band-edge effects and achieve a vertical out-of-plane emitter with large lasing area, it is critical that the dimensions containing the periodicity be made sufficiently large.

### 3.3 Lasing

Thus far, we have based our analysis on the properties of the passive dielectric structure. Let us now consider the effects of adding gain; specifically, the four-level gain medium described in Chapter 2. Although we will focus on the particular case of four-level gain media, we expect the lasing properties of the PCSELs to be influenced primarily by the EM properties of the passive dielectric structure, rather than the microscopic details of the gain mechanism. Hence, our results should be broadly applicable to any active device describable by semiclassical laser theory. The specific effects of optimizing lasing action will depend on the gain medium. Thus, as we show below, for a four-level gain medium, it leads to an arbitrary reduction of the lasing threshold; whereas in a semiconductor laser, it may lead to lasing thresholds close to the limit set by the transparency condition.

For simplicity, we assume that the gain is uniformly distributed within the dielectric (the effects of non-uniform gain is outside the scope of this chapter, but the present numerical techniques can treat it effectively). For the decay lifetimes, we take $\tau_{10} = \tau_{32} = 5 \times 10^{-14}$ s and $\tau_{21} = 5 \times 10^{-12}$ s (so a metastable state can form at $N_2$). For the coupling constant, we take $\sigma_m = 1 \times 10^{-4}$ C$^2$/kg (this value was obtained assuming that the Purcell effect is
negligible); for the total electron density, we take \( N_{\text{tot}} = 5 \times 10^{23} \text{ m}^{-3} \). These values are realistic, and similar to those used in Ref. [9]. For each structure, we choose a different value of the gain center \( \omega_m \), in order to select the mode that we wish to lase; in the FDTD calculations, this frequency is set to the frequency of the corresponding passive cavity mode. The gain linewidth \( \Gamma_m \) is taken to be 0.002 \((2\pi c/a)\), which is sufficiently narrow to avoid exciting neighboring modes.

### 3.3.1 Infinite periodic structures

We first compute the lasing properties of the infinite slab. The computational domain is similar to the one in Fig. 3-1(b), with periodic boundary conditions along the left and right boundaries and PML absorbers along the top and bottom boundaries. Fig. 3-4 shows the resulting plot of output power versus \( R_p \), where the threshold corresponds to that defined in Eq. (2.21) of Chapter 2. Three different structures, with groove depths of 0.05\( a \), 0.1\( a \), and 0.15\( a \), are simulated; the groove width is kept at 0.1\( a \), and slab thickness at 0.3\( a \). The filled circles in this plot are the results of FDTD calculations (time-stepping until steady-state laser operation was achieved); the solid lines are the CMT predictions, with parameters fitted from separate FDTD calculations of the passive structure’s Fano resonance frequency, electric field mode profile, and \( Q \).

For each of the calculations in Fig. 3-4, the gain center \( \omega_m \) is situated at the resonance frequency of the symmetric mode. The anti-symmetric mode is neglected in this analysis because it does not allow power emission in an infinite structure. For groove depths of [0.05\( a \), 0.1\( a \), 0.15\( a \)], we have \( \omega_m = [0.435, 0.444, 0.456] \times (2\pi c/a) \) and \( Q \) values [1964, 451, 230]; see Fig. 3-2(a). The laser threshold is found to be inversely proportional to \( Q \); physically speaking, higher input pump rates are needed to overcome larger losses. Moreover, the three structures exhibit very similar rates of growth of output power, \( dP/dR_p \) [as defined in Eq. (2.22) of Chapter 2]. As we will later explain, this is not true for finite structures.

The agreement between FDTD and CMT is very good. In particular, CMT predicts that the output power grows linearly with \( R_p \) above the lasing threshold, and the FDTD results are very close to linear. The match remains excellent for \( R_p \) as much as an order of magnitude above the lasing threshold. The CMT is particularly useful for \( R_p \) near threshold, where FDTD computations are very time-consuming due to the temporal turn-on delay.
Figure 3-4: Output power versus $R_p$ relationships of the 2D infinite slab described in Fig. 3-1(b), for three depths of the air grooves at 0.05a, 0.1a, and 0.15a (width remains at 0.1a), with corresponding $Q$ values 1964, 451, and 230 respectively. Both semi-analytic predictions from CMT (solid lines) and FDTD (filled circles) steady-state calculations are plotted for the upper band-edge mode at $k_x = 0(2\pi/a)$. There is good agreement between the semi-analytic and calculated values. The threshold is higher for the lower-$Q$ PhC slab which clearly suggests that higher pumping rates are needed to overcome systems with higher losses.

before lasing action begins. For larger $R_p$, the results begin to deviate; the influence of the gain media on the fields can no longer be taken to be linear, so second order corrections to CMT are required and the lasing modes are no longer accurately described by the modes of the linear (passive) cavity. Hence, FDTD calculations should be applied for large $R_p$ above threshold. This shows that the CMT model that we have developed greatly complements the FDTD approach.

### 3.3.2 Finite periodic structures

Fig. 3-5 shows $R_p$ versus total power output obtained by FDTD and CMT in finite slabs (i.e. the PML absorber is now placed along all four boundaries of the computational domain). Three different slab widths are used: $L_x = 20a$, 40a, and 80a. We fix the groove depth at 0.05a, with all other parameters kept the same as in Fig. 3-4.

The left-hand plot in Fig. 3-5 shows the upper band-edge modes, while the right-hand plot shows the anti-symmetric lower band-edge. Readers are referred to Fig. 3-2(b) or the
Figure 3-5: Output power versus $R_p$ relationships of the 2D finite slabs described in Fig. 3-2(b) for three dimensions of $L_x$ at 20a, 40a, and 80a. Size of the grooves is fixed at 0.05a x 0.1a. (a) Higher-frequency symmetric modes with corresponding $Q$ values 179, 413, and 925. (b) Lower-frequency anti-symmetric modes with corresponding $Q$ values 231, 749, and 3243. Both semi-analytic predictions from CMT (solid lines) and FDTD (filled circles) steady-state calculations are shown. Insets plot the same data in linear scale for $R_p$ values near threshold. In addition, (b) also shows the SALT (dashed lines) results for the 20a and 40a slabs. Good agreements between the three methods are observed. Slope of the lines changes with $L_x$ (see text) while the right plot confirms that the anti-symmetric mode has the largest $Q$ in the finite system that is available for lasing.

figure caption for the $Q$ values and frequencies of these modes. As expected, the structures with larger $L_x$ have lower lasing thresholds, due to stronger diffraction of the waves, which is needed for better feedback and modal confinement. The band-edge effects in finite PhC slabs depend upon the degree of overlap between the lasing modes and the periodic dielectric, as well as the lateral sizes in their periodic planes [26]. The close proximity of the gain medium to the air grooves in our setup ensures that the former condition is well met, so that band-edge mode laser operation can be achieved for $L_x$ of as little as 20a. It is also noted from the two plots that the lower band-edge modes give rise to lower thresholds than the upper band-edge modes, for slabs of equal $L_x$. Moreover, the slopes $dP/dR_p$ are different. The large 80a slab, with the largest lasing area, emits the most power and therefore exhibits the highest slope. This can clearly be seen from the insets of Fig. 3-5(a) and (b), where linear plots of the same data are shown for values near the thresholds.

In order to confirm the FDTD and CMT findings, we have also carried out calculations using the steady-state ab-initio laser theory (SALT) [21,25,77]. Unlike the FDTD calcula-
tions, the SALT is a frequency-domain method and does not face FDTD's computational limitations where many time steps are required to bring the laser to its steady state, especially near threshold. Thus, it is possible to apply much finer spatial discretization. The power output calculated by the SALT is shown by the dashed curves in Fig. 3-5. $R_p^{th}$ from the SALT differs from the FDTD result by $\sim 9\%$, a very acceptable deviation considering the difference in resolution between the two calculations\(^1\), and shows a similar decrease with $L_x$. The power slope was found to increase approximately linearly with $L_x$. Apart from the aforementioned $9\%$ difference in $R_p^{th}$, these results are in good agreement with FDTD, and in particular are in slightly better agreement than CMT for large $R_p$. For more details on how the SALT is formulated and applied to the PhC slabs in this work, the readers are referred to Ref. [78].

3.4 2D periodic slab lasers

We now turn our attention to the 3D slab system illustrated in the inset Fig. 3-1(c), consisting of a square lattice of air cylinders. The slab thickness is $0.3a$, while the radius and depth of the air cylinders are $0.15a$. This design operates based on the same principles as the simpler 1D periodic grooves design, so that the physical concepts explored previously may be equally applied in this case. In Fig. 3-6, the magnetic and electric field profiles of TE-like excitations are provided for the four modes at $\Gamma$, two being non-degenerate (two lowest frequency modes) and the remaining pair is degenerate. Analogous to the antisymmetric modes that exist in corrugated slabs, the non-degenerate modes of such infinite periodic slabs have infinite photon lifetime, which again may be attributed to mode symmetry mismatch with the radiative continuum [34]. At $\Gamma$ of the square lattice, its irreducible representation may either be 1D (singly-degenerate) or 2D (doubly-degenerate), and possesses the full symmetry of the lattice. Such symmetries enforced upon the singly-degenerate modes ensure that their in-plane radiative components cancel. This can be seen from the electric field mode profiles in Fig. 3-6(a) and (b), where the $E_x$ components are of opposite directions relative to the air cylinder. The same holds for $E_y$. Hence, no coupling to air is observed. On the other hand, vertical radiation of the electric field occurs for the doubly-degenerate modes in Fig. 3-6(c) indicating low finite $Q$ values. We apply the CMT approach

\(^1\)On the other hand, the CMT results agree very well with FDTD results because the CMT parameters were fitted from passive FDTD calculations performed with the same spatial resolution
Figure 3-6: (a) Magnetic and electric field profiles of the first singly-degenerate mode at \( \Gamma \) in a unit cell of the 0.3\( a \)-thick 3D PhC slab structure shown in Fig. 3-1(c), partially punctured with square lattice of air cylinders having height and radius 0.15\( a \). The PhC slab is outlined in green. Top panels depict the lateral cuts along the xy-plane with magnetic field pointing into the page and electric field pointing to the left, where positive (negative) values are in red (blue). Bottom panels are cuts along the yz-plane with magnetic field pointing downwards and electric field pointing into the page. These results are for TE-like modes. \( E_y \) (not shown) has the same profile as \( E_x \), except rotated 90° about z-axis. (b) Same as in (a) but for the second singly-degenerate mode. (c) Same as in (a) but for the doubly-degenerate mode. Its counterpart at the same frequency has the magnetic field profile rotated 90° about z-axis. (d) Output power versus \( R_p \) relationships retrieved from CMT is plotted for air cylinders with radius 0.3\( a \), 0.4\( a \), and 0.5\( a \), for the doubly-degenerate mode presented in (c). Their respective frequencies are 0.449, 0.457, and 0.466(2\( \pi c/a \)) with \( Q_s \) equal 764, 263, and 126.

To calculate the power output from a unit cell for the degenerate mode at three radii of the air cylinders shown in Fig. 3-6(d). As in the corrugated slab, \( Q \) increases for smaller air cylinders leading to lower threshold pump rate. Practical considerations with regards to size of the structural periodic perturbations include the ease of fabrication as dimensions scale down, and also the need for close proximity to the gain layers for enhancement of the band-edge effects.

Lastly, we examine the finite size 3D slab in Fig. 3-7. Having already verified the CMT predictions with FDTD method, the former is again adopted to calculate the power output of finite 0.3\( a \)-thick PhC slabs with air cylinders 0.15\( a \) deep and diameter 0.3\( a \). Three sizes of the PhC region are studied: 15\( a \times 15 a \), 25\( a \times 25 a \) and 35\( a \times 35 a \). In order to excite PhC states and to model realistic conditions in similar PhC lasers operating with an optically or electrically pumped central area, the 2D PhC region has to be surrounded by un-pumped...
Figure 3-7: Top: Magnetic and electric field profiles corresponding to the singly-degenerate mode in finite $15\alpha \times 15\alpha$ PhC slab structure described in Fig. 3-6(a). The PhC slab is outlined in green. Top two panels depict the lateral cuts along the xy-plane with magnetic field pointing into the page and electric field pointing to the left, where positive (negative) values are in red (blue). Bottom panels are cuts along the yz-plane with magnetic field pointing downwards and electric field pointing into the page. These results are for TE-like modes. $E_y$ (not shown) has the same profile as $E_x$, except rotated $90^\circ$ about z-axis. Bottom: Output power versus $R_p$ relationships retrieved from CMT. Frequencies and $Q$ values of the $15\alpha \times 15\alpha$, $25\alpha \times 25\alpha$, and $35\alpha \times 35\alpha$ PCSEL structures are $0.431$, $0.433$, $0.435(2\pi c/\alpha)$, and $64$, $178$, $385$, respectively.
regions. This is achieved in our simulations by truncating the finite size PhC in air and extending the uniform dielectric slab into the PML, which surround the whole computational domain. The lasing mode considered is that of the first singly-degenerate mode shown in Fig. 3-6(a) and may be compared to the field profiles presented in Fig. 3-7 for the PhC slab of size 15a x 15a. The slope [as defined in Eq. (2.22)] and threshold pump rate improves for the larger PhC, consistent with what we would have expected, while the PCSEL remains single-mode. The primary losses for the small-sized PhC considered here is through the lateral leakage into the absorbing boundaries. Further note that magnitudes of the output power, and hence slope, are significantly higher than those found for the corrugated slabs in Fig. 3-5. This can be understood as a consequence of an additional dimension present in the current calculations. We confirm that the singly degenerate modes for these finite slabs have higher Qs than the doubly-degenerate modes and hence, should be the ones within the first set of frequencies at Γ that, in practice, are selected for lasing. Such mode selection (singly-degenerate) should be possible owing to the distinct frequencies and field profiles of the four modes at Γ, and its lower pumping rate requirements. We also note here that upon successful coupling to the desired laser modes, Q may further be increased by enhancing the confinement geometrically: (i) in the lateral directions by employing PhC heterostructures, and (ii) in the out-of-plane direction via the addition of DBRs on top or below the PhC slab [79].

3.5 Conclusion

We have shown that the lasing action in PCSELS originates from Fano resonances. Of particular interest for lasing are dark Fano resonances. We used two different theoretical techniques to study these systems. We have seen that in actual finite-size structures, the large Q factors displayed by the dark states lead to a significant reduction of the corresponding lasing threshold with respect to conventional VCSELS. In addition, our calculations suggest that, for input pump rates close to the threshold, PCSEL structures emit most of their lasing power in the plane of periodicity rather than in the vertical direction. However, notice that this lasing power can be directed into the out-of-plane direction simply by perturbing the symmetry of dark Fano resonances [80]. We believe the findings reported here provide further physical insight into lasing action in PCSELS and will help designing active devices
based on this class of systems. Finally, it should also be mentioned that spontaneous emission has not been included in our simulations as it does not affect the physics explored and the conclusions of this work. This leads to no output power detected below the threshold pump rate $R_{th}$. We simply note here that spontaneous emission may be represented in our gain medium by introducing noise-like dipole sources [9].
Chapter 4

Larger area single-mode photonic crystal surface-emitting lasers enabled by an accidental Dirac-point

We tune the regular lasing bandedges of quadratic dispersions in the photonic crystal surface-emitting lasers (PCSELs) to form a single accidental Dirac cone of linear dispersion at the Brillouin zone center. This not only increases the mode spacing by orders of magnitudes, but also eliminates the distributed in-plane feedback to enable single-mode PCSELs of larger area thus higher output power. The advantages of using accidental Dirac cones are systematically evaluated through two-dimensional in-plane calculations and confirmed by three-dimensional simulations of slab devices. When the out-of-plane radiation is present, an additional frequency-locking phenomenon at the degenerate point is analyzed.

4.1 Introduction

Higher power single-mode on-chip lasers with good beam qualities are always in demand in applications. While the edge emitting sources (DFBs) suffer from catastrophic optical damage at their facets, surface-emitting sources (VCSELs) are usually limited by their small cavity sizes. In both examples, the single mode is selected by one-dimensional feed-
backs structures. Utilizing two-dimensional distributed feedbacks, surface emitters have achieved broad-area single-mode operations [12,81]. In particular, the PCSELs have not only achieved the highest surface-emitting single-mode power [35] but also the ability to control the shapes [36], polarizations [10] and directions [31] of their far-fields. PCSELs are essentially the two-dimensional (2D) versions of the second-order distributed feedback (DFB) lasers [40], where the higher quality factor lasing mode is selected through the symmetry mismatch to the free-space modes [41]. However, the lasing areas of PCSELs are limited by two fundamental constraints. First, the mode spacing decreases as the cavity area increases, which promotes multi-mode lasing. Second, the distributed in-plane feedback localizes the lasing fields to individual sections, which promotes multi-area lasing. In this work, we tune the regular lasing bandedges of quadratic dispersions to form accidental Dirac cones [42,43] of linear dispersions. This not only increases the mode spacing by orders of magnitudes but also eliminates the distributed in-plane feedback, turning the periodic index-modulated cavities into equivalent Fabry-Perot-like cavities where the modes have different out-of-plane coupling losses. Both advantages promise single-mode PCSELs of larger areas thus higher output powers. In the following two sections, we systematically evaluate the advantages of using accidental Dirac cones through 2D calculations and provide consistent results of 3D slab devices. In slab devices, an additional frequency-locking phenomenon at the degenerate point, with potentially high density of states (DOS), is analyzed. Although we only consider passive photonic crystals (PhCs) in this work, their modal properties and dispersions remain the same in systems with uniform gain or loss.

4.2 In-plane analysis

4.2.1 2D PhCs (unit-cell calculations)

A Dirac cone is a special dispersion relation in the band structure where the dispersion is linear and the DOS vanishes at the Dirac point. It is well-known that pairs of Dirac cones can exist in PhCs [82,83]. However, a single Dirac cone at the Brillouin zone center (Γ) is ideal for both vertical emission and single-mode lasing in a PCSEL. This single Dirac cone can form when the PhC is geometrically tuned so that a singly-degenerate band is accidentally degenerate with a pair of doubly-degenerate bands at Γ [42,43]. When this happens, two of the three bands form an isotropic [84] Dirac cone and the other one is flat.
Figure 4-1: TM photonic band structure of a triangular array of dielectric rods ($\varepsilon_{\text{rod}} = 12.5$) in two different background materials ($\varepsilon_{\text{bg}} = 1, 11$). The rod radius $r$ is tuned so that the doubly-degenerate modes (with $E_1$ symmetry in the $C_6\nu$ point group) are accidentally degenerate with a singly-degenerate $A_1$ mode (red band) at $\Gamma$. (a) The band structure and DOS of the high dielectric contrast PhC. The modal profiles of the three accidentally degenerate modes at $\Gamma$ are depicted with electric field pointing into the page and having positive (negative) values in red (blue). (b) The band structure of the low dielectric contrast PhC.
This accidental Dirac point can exist in PhCs of square or triangular lattices consisting of either dielectric-rod or air-hole arrays, with either high or low index contrasts. In Fig. 4-1, we obtained the single accidental Dirac cone in a triangular array of dielectric rods by tuning the rod radius \( r \), where the lattice constant is denoted by \( a \). The calculations were performed with a unit-cell using the MIT Photonic-Bands package [85]. The dielectric contrast of the PhC is high (12.5 : 1) in Fig. 4-1(a) and low (12.5 : 11) in Fig. 4-1(b). The resulting linear dispersion of the singly-degenerate mode (red band) has DOS that vanishes linearly with frequency at the accidental Dirac point, as shown in the right plot of Fig. 4-1(a). This corresponds to large mode spacings near the bandedge and a high spontaneous emission coupling factor [86]. Modal profiles of the three accidentally degenerate modes at \( \Gamma \) are also illustrated in Fig. 4-1(a). Only the symmetry of the singly-degenerate mode is mismatched with the free space modes [41] and so, has lower out-of-plane radiation losses (higher \( Q_\perp \)) than the other two modes. Thus, the bandedge modes of this singly-degenerate band are the only lasing candidates within the spectral range shown in the insets of Fig. 4-1(a) and (b). From now on, we only consider the modes of this singly-degenerate band for the rest of the chapter.

### 4.2.2 Finite-sized 2D PhC cavities

To quantify the benefits of having linear dispersions at the lasing bandedges, we first compare the bandedge modes of two finite-sized PhCs. One of them has a linear dispersion at the accidental Dirac point while the other has a quadratic dispersion. The PhC analyzed is a triangular array of dielectric rods (\( \epsilon_{\text{rod}} = 12.5 \)) in air with dimension \( L \) shown in Fig. 4-2(a). We altered the band dispersions of the PhC by varying the rod radius \( r \). The dispersion relations for three values of \( r \) are illustrated in Fig. 4-2(b). A linear dispersion is formed when \( r = 0.184a \) while quadratic dispersions are formed at the remaining two radii. In Fig. 4-2(c), the quality factors \( Q_\parallel \) of the bandedge modes are plotted as a function of the frequency for a cavity of \( L = 300a \) with linear dispersion (top plot) and a cavity of \( L = 40a \) with quadratic dispersion (bottom plot). These results were calculated using the finite-difference time-domain (FDTD) method [38] with perfectly matched layer boundary regions. The 300a PhC cavity at the accidental degeneracy is found to have the same mode spacing (between the first two bandedge modes) as that of the 40a cavity even though the former is 50 times larger in area. Moreover, \( Q_\parallel \) of the first bandedge mode in the 300a cavity
Figure 4-2: (a) A finite-sized PhC cavity consisting of a triangular array of dielectric rods \((\varepsilon_{\text{rod}} = 12.5)\) embedded in air with dimension \(L = 40a\). (b) The high \(Q_\bot\) band dispersions of three PhCs with different rod radii. At \(\Gamma\), the red band has a linear dispersion while the green and blue bands have quadratic dispersions. (c) Bandedge modes of two finite-sized PhC cavities of \(L = 300a\) with linear dispersion (top plot) and of \(L = 40a\) with quadratic dispersion (bottom plot). Insets show the mode profiles with electric field pointing into the page and having positive (negative) values in red (blue). (d) The upper plot illustrates the bandedge mode spacing \((\Delta \omega/\omega_0)\) where \(\omega_0\) is the bandedge frequency) as a function of the cavity area. The lower plot illustrates \(Q_\parallel\) of the bandedge mode as a function of the cavity area. The filled circles and squares correspond to the results shown in (c).
(linear dispersion) is an order of magnitude smaller than that in the 40a cavity (quadratic dispersion). We note that the low $Q_\parallel$ implies a weak in-plane feedback and low localization effects. Another distinction between the cavities with linear and quadratic dispersions in Fig. 4-2(c) is that the modes of the top plot (linear dispersion) have similar $Q_\parallel$ values while the modes of the bottom plot (quadratic dispersion) have $Q_\parallel$ values that are decreasing with frequency. Nonetheless, in both cases, the first bandedge modes having the highest $Q_\perp$ values will lase with the lowest thresholds, because $Q_\parallel$ values increasing with the cavity sizes will not be the main loss mechanism in large cavities.

In the upper plot of Fig. 4-2(d), we compare the mode spacing as a function of the PhC area for linear and quadratic dispersions at the bandedge. The mode spacing of a finite-sized PhC can be estimated from the dispersion relation using the periodic boundary condition (PBC) to relate the dimension $L$ to the in-plane wavevector $k$. The period of the PBC is approximated to be $2L$ and $L$ for the first and second bandedge mode based on their mode profiles shown in Fig. 4-2(c); the monopole-like mode has half a period within $L$ while the dipole-like mode has one period inside $L$. The corresponding wavevectors for these two modes are $k_0 = \pi/L$ and $2k_0 = 2\pi/L$ under this approximation. The bandedge mode spacing is then the frequency difference between these two $k$-points illustrated in Fig. 4-2(b). Using this approach, the mode spacing ($\Delta \omega$) of a linear dispersion is found to be inversely proportional to $L$ ($\Delta \omega = \pi \beta / L$) while the mode spacing of a quadratic dispersion is inversely proportional to $L^2$ ($\Delta \omega = 3\pi^2 \alpha / L^2$). $\beta$ is the linear slope and $\alpha$ is the quadratic curvature of the dispersions near the bandedge, and $c$ is the speed of light in air. These semi-analytical expressions described above are verified by calculations of actual finite-sized PhC cavities in Fig. 4-2(d). The results clearly indicate that by tuning to a linear dispersion at accidental degeneracy, the mode spacing can be made much larger than that in a typical PCSEL with quadratic dispersion. For instance, at $L = 400a$, the mode spacing at the accidental point (red line) is at least 60 times larger compared to the PhC detuned from it (blue line). We note that this increase in mode spacing becomes arbitrarily large as the area increases. Equivalently, for the same mode spacing, the PhCs with a linear dispersion can be made much larger in area than those with quadratic dispersions. In Fig. 4-2(d), the cavity size is increased by more than two orders of magnitudes when $r$ is tuned from 0.26a to 0.184a while maintaining the same mode spacing ($\Delta \omega/\omega_0 = 1 \times 10^{-4}$).

In the lower plot of Fig. 4-2(d), we compare the in-plane feedback as a function of the
PhC cavity area for linear and quadratic dispersions at the bandedge. We quantify the in-plane feedback strength by the in-plane quality factor $Q_\parallel = \omega_0 \tau_\parallel$, where $\tau_\parallel (\propto L/v_g)$ is the photon lifetime in the PhC cavity [78] and $v_g = d\omega/dk$ is the group velocity. $v_g$ is a constant when the dispersion is linear, and is proportional to $k$ when the dispersion is quadratic. The above analysis using the PBC finds that $k$ scales as $1/L$. Hence, $Q_\parallel$ should scale with $L$ when the dispersion is linear and $L^2$ when the dispersion is quadratic. In Fig. 4-2(d), $Q_\parallel$ calculated from finite-sized cavities agree well with the above trends, except for small structures whose modes are of $k$ values too far away from $F$ to follow the quadratic functions. Physically, the linear increase of $Q_\parallel$ with $L$ implies that the distributed in-plane feedback in a typical PCSEL is completely eliminated at the accidental degeneracy. In other words, the PCSEL behaves like a 2D Fabry-Perot cavity where feedback only comes from its end mirrors. However, unlike typical Fabry-Perot cavities where all the modes have the same $Q$ values, the proposed PCSELs can still select the first bandedge mode to lase due to its highest $Q_\parallel$ value. In real devices, fabrication imperfections can cause field localization effects, but those effects will be much reduced when the bandedge is of linear dispersion.

4.3 Slab analysis

The previous section considered 2D PhCs where out-of-plane radiation loss does not exist. In this section, we consider PhC slab structures with symmetric air claddings and open boundaries in the vertical directions. To investigate how the previous analysis of accidental degeneracy at $\Gamma$ differs in PhC slabs, we first study the simpler 1D PhC slabs in Fig. 4-3 and 4-4, and extend the analysis to 2D PhC slabs in Fig. 4-5. The calculations in this section are performed with the finite-element method (FEM) using the commercially available software COMSOL.

4.3.1 1D periodic slabs (unit-cell calculations)

It is known that linear dispersions at $\Gamma$ exist in 1D PhCs at the quarter-wave-stack (QWS) condition where the two bands are accidentally degenerate. We find that similar features exist in 1D PhC slabs when the bandedges are tuned to be degenerate, except for a caveat: the two bands remain degenerate even away from $\Gamma$. One such example is considered in Fig. 4-3(a), where the band diagram of the two lowest bandedge modes and their corre-
Figure 4-3: The top inset shows an unit-cell of a 0.3a-thick 1D PhC slab consisting of alternating high ($\varepsilon_{\text{high}}$) and low dielectric constant ($\varepsilon_{\text{low}}$) materials. The width of the block with $\varepsilon_{\text{high}}$ is tuned so that the bands are accidental degenerate at $\Gamma$. Only modes with electric field pointing into the page are considered. (a) The band structure of the PhC slab with $\varepsilon_{\text{high}} = 12.5$ and $\varepsilon_{\text{low}} = 6.25$. The top plot shows the corresponding $Q_L$ values. (b) The band structure of the PhC slab with $\varepsilon_{\text{high}} = 12.5$ and $\varepsilon_{\text{low}} = 11$. The shaded regions are the radiation-loss linewidths of the two bands centered along the green dashed lines.

The corresponding $Q_L$ values are plotted. An accidental degeneracy is achieved by tuning the width $w$ in the PhC unit-cell shown at the top of Fig. 4-3. The green dashed lines are the uncoupled linear dispersions that one would expect in a 1D PhC (with no radiation loss) at the QWS condition. However, in slabs, the two bands are frequency-locked when their in-plane wavevectors $k_x$ are less than the $k_{\text{critical}}$ value indicated in Fig. 4-3(a). The flat band region occurs when the frequencies of the two modes are close enough so that their respective radiation-loss linewidths ($\Delta\omega_\perp = \omega/Q_\perp$) overlap in frequency. Physically, $k_{\text{critical}}$ marks the point beyond which the two modes no longer interact via the out-of-plane radiation-loss channel. For the bandedge modes to have large mode spacings and low in-plane feedback, we would like to keep $k_{\text{critical}}$ small [e.g. Fig. 4-3(b)] so the first bandedge mode can operate outside this flat-band region.

We model the frequency-locking behavior of the 1D PhC slab at $\Gamma$ using the coupled-wave theory, similar to the analysis for the second-order DFB lasers discussed in Ref. [87]. Periodic index modulation in the PhC slab couples the forward propagating waves ($k_x = 2\pi/a$)
to a out-of-plane radiating wave \( (k_x = 0) \) in first-order diffraction, and to a backward propagating wave \( (k_x = -2\pi/a) \) in second-order diffraction. Higher diffraction orders carrying little energy are neglected. These three terms of the electric field are plugged into the Maxwell equations, and the dispersion relation at accidental degeneracy can be derived as

\[
\frac{\Delta \omega_g}{v'_g} = -i \frac{\Delta \omega'_\perp}{2v'_g} \pm \sqrt{k_x^2 - \left(\frac{\Delta \omega'_\perp}{2v'_g}\right)^2}.
\] (4.1)

Here, \( \Delta \omega_g \) is the departure of the frequency from the accidental degenerate frequency, \( \Delta \omega'\perp = \omega/Q'_\perp \) is the linewidth of out-of-plane radiation-loss when \( k_x > k_{\text{critical}} \), and \( v'_g \) is the slope of the linear dispersion (green dashed line). The two dispersion curves merge when the real part of \( \Delta \omega_g \) is zero, which is the case when \( k_x < k_{\text{critical}} = \Delta \omega'_\perp/2v'_g \). \( k_{\text{critical}} \) is thus inversely proportional to both \( Q'_\perp \) and the slopes of the linear dispersions. On the other hand, the imaginary part of \( \Delta \omega_g \) is related to the linewidth of out-of-plane radiation-loss.

At \( k_x = 0 \), Eq. (4.1) predicts a mode with infinite \( Q_\perp \) and another with quality factor half of the \( Q_\perp \) value outside \( k_{\text{critical}} \). This trend is consistent with the \( Q_\perp \)-plots in Fig. 4-3. We note that the above analysis remains valid in active systems with uniform gain or loss which simply adds an imaginary term to Eq. (4.1).

To verify the analytical model in Eq. (4.1) and demonstrate how the size of the flat-band region can be controlled, we increase \( e_{\text{low}} \) of the 1D PhC slab in Fig. 4-3(a) from 6.25 to 11. The resulting band diagram is shown in Fig. 4-3(b) where the same frequency-locking behavior exists, except that the \( k_{\text{critical}} \) value now becomes 30 times smaller. This reduction in \( k_{\text{critical}} \) by more than an order of magnitude is due to the roughly 30 times increase in the \( Q_\perp \) of both bands when the dielectric constant is made larger.

The frequency-locking phenomenon described above, where accidentally-touched band dispersions merge further through radiation losses, can also be found in system of periodic gain-loss modulations [88]. However, our analysis does not apply to symmetry-protected band degeneracies, such as the linear Dirac and the quadratic degeneracies at K in Fig. 4-1(a). Those dispersion relations do not alter unless their underlining symmetries are broken [89]. Although the symmetry-protected Dirac points are more robust than the single accidental one under the perturbation of loss, gain, refractive index or structural changes, they do not possess the benefits of single-mode operation nor vertical emission.
4.3.2 Finite-sized 1D periodic slabs

We extend the unit-cell analysis of the 1D PhC slab in Fig. 4-3(a) to study finite-length slabs with dimension $L_x$ shown at the top of Fig. 4-4\textsuperscript{1}. In finite slab structures, radiation losses in both the in-plane ($x$) and out-of-plane ($y$) directions are present. The total quality factor $Q_{\text{tot}}$ is calculated from the FEM and is related to the in-plane and out-of-plane radiation losses by power conservation: $1/Q_{\text{tot}} = 1/Q_{||} + 1/Q_{\perp}$. To obtain $Q_{||}$ and $Q_{\perp}$ from $Q_{\text{tot}}$, the time-averaged power density are integrated along the boundaries of a box enclosing the PhC slab, and the fraction of the total escaped power in the $x$- and $y$-directions is found.

The mode spacings and $Q$ values of the first bandedge modes are plotted as a function of $L_x$ in Fig. 4-4(a) and (b) respectively. $Q_{\text{tot}}$ of the first and second bandedge modes are also compared in Fig. 4-4(c). In our previous analysis using the PBC, we related $k_x = \pi/L_x$ to the first bandedge mode. Using this relationship, we can relate the $k_{\text{critical}}$ value in Fig. 4-3(a) to the corresponding dimension $L_x^{\text{critical}}$ indicated in Fig. 4-4. $L_x^{\text{critical}}$ separates the three plots into two regions. When $L_x < L_x^{\text{critical}}$, the first bandedge mode lies outside the flat band region in Fig. 4-3(a) where the dispersion is linear. This is confirmed in Fig. 4-4(a) and (b) where the mode spacing of the first bandedge mode is observed to scale with $L_x^{-1}$ while its $Q_{||}$ value scales with $L_x$. However, the $Q_{\text{tot}}$ values of the first and second bandedge modes are similar, making single-mode selection difficult. When $L_x > L_x^{\text{critical}}$, the $Q_{\text{tot}}$ values between the first and second bandedge modes are prominent facilitating single-mode selection. However, in this case the first bandedge mode lies in the flat band region of Fig. 4-3(a) where its mode spacings decreases sharply and its $Q_{||}$ value increases at a rate faster than $L_x^2$ as the cavity size increases. Consequently, in order to avoid the disadvantages on both sides away from $L_x^{\text{critical}}$, a cavity size close to $L_x^{\text{critical}}$ would be an ideal operation point for a single-mode PCSEL of the largest area.

We also investigated how structural disorders might affect the bandedge mode spacing and in-plane feedback at the accidental degeneracy by introducing disorders to both the locations and width ($w$) of the high-indexed blocks in the finite-sized PhC slabs. Ten different disordered cavities were calculated for each $L_x$, where the disorders were generated with Gaussian distributions of standard deviation 0.02a. The statistical results are represented

\textsuperscript{1}Since a large $k_{\text{critical}}$ corresponds to a small $L_x^{\text{critical}}$, the slab structure in Fig. 4-3(a) with a large $k_{\text{critical}}$ value is not ideal for large area single-mode operations of PCSELS. It is used in our analysis because the features near its $k_{\text{critical}}$ point can be probed with a small finite-sized slab (i.e. $L_x \approx 55a$), allowing the full PhC structure to be numerically simulated with modest computational resources.
Figure 4-4: The top inset shows a finite-sized $0.3a$-thick 1D PhC slab of dimension $L_x$ consisting of alternating high ($\epsilon_{\text{high}} = 12.5$) and low ($\epsilon_{\text{low}} = 6.25$) dielectric constant materials. Band diagram of its unit-cell counterpart is shown in Fig. 4-3(a), where the bands at $\Gamma$ are accidentally degenerate. (a) The mode spacing between the first and second bandedge modes as a function of the PhC slab length. (b) $Q_\parallel$ and $Q_\perp$ of the first bandedge mode as a function of the PhC slab length. (c) The total quality factors $Q_{\text{tot}}$ of the first (red diamond) and second (blue diamond) bandedge modes plotted as a function of the PhC slab length.
Figure 4-5: Band structure (TE-like modes near Γ) of a GaAs-based 2D PhC slab on AlAs (n = 3) substrate. A square array of air-holes with radius 0.285a is patterned in the top 0.467a-thick GaInP (n = 3.22) layer. Sandwiched between GaInP and the substrate is a homogeneous 1a-thick GaAs (n = 3.516) layer. Bottom inset shows 2 × 2 unit-cells of the considered PhC slab. The hole radius is tuned so that the pair of doubly-degenerate modes (blue bands) is accidentally degenerate with a singly-degenerate mode (red) at Γ. The corresponding Q-plot above shows that $Q_\perp$ diverges only for the red band.

by the error bars in Fig. 4-4; the bars are centered at the mean values and the height of the bars are two times of the standard deviations. Important, the linear trends in both mode spacings and $Q_\parallel$ below $L_x^{\text{critical}}$ are preserved.

### 4.3.3 2D periodic slabs (unit-cell calculations)

Lastly, we consider accidental degeneracy in a realistic 2D PhC GaAs-based slab on AlAs substrate, achieved by tuning the air-hole radius. Fig. 4-5 illustrates its band structure near Γ. Similar frequency-locking behaviors are observed: a $k_{\text{critical}}$ exists so that the bands form a 2D flat plane for $|k| < k_{\text{critical}}$ and becomes linear as $|k|$ increases. Single-mode PCSEL operation is again possible because $Q_\perp$ diverges only for one band (red). As before, $k_{\text{critical}}$ is related to the mode coupling via the radiation-loss channel in air and can similarly be tuned to be large or small. Here, without any optimizations, $L_x^{\text{critical}}$ is roughly 2500a. This corresponds to a cavity area more than six times larger than some of the largest PCSEL areas reported [12,26].

As a passing remark, we point out that the flat-band regions of the band diagrams (near Γ) can potentially have high DOS. This is confirmed in Fig. 4-4 where the mode
spacing reduces abruptly and the $Q_{\text{tot}}$ increases as the length of the PhC slab grows beyond $L_x^{\text{critical}}$. Moreover, we expect the three-fold band degeneracies in 2D PhC slabs to further enhance the DOS. While we seek to make the flat-band region tiny for larger-area single-mode PCSELs, a large $k$-region of flat bands providing large DOS can be very useful for the enhancement of light-emitters [90] and light-matter interactions [91].

4.4 Conclusion

We have demonstrated that, compared to typical PCSELs with quadratic bandedge dispersions, the formation of accidental Dirac cones of linear dispersions at $\Gamma$ not only increases the mode spacing by orders of magnitudes but also eliminates the distributed feedback in-plane. This overcomes two of the fundamental limitations to attaining larger-area single-mode lasers. We also found that, at the degenerate point in the passive slabs, the bandedge dispersions are locked into flat bands through the coupling of out-of-plane losses. To achieve large area single-mode PCSELs, we would like to engineer the flat-band region to be small. We believe that our findings are beneficial towards realizing higher-power single-mode lasers.
Chapter 5

Experimental realization of photonic crystal surface emitting lasers

In this chapter, we first characterize the passive macroscopic photonic crystal slabs in terms of their resonant frequencies and quality factors at $k \approx 0$. In particular, Sec. 5.1 distinguish experimentally the existence of the dark Fano resonances discussed in Chapter 3. In Sec. 5.2, we realize lasing at these dark Fano resonances in slab structures using solutions of R6G molecules as the gain medium. The experimental lasing results (i.e. thresholds and slope efficiencies) are verified with the organic lasing models of Chapter 6.

5.1 Unique high-Q optical resonances in macroscopic photonic crystal slabs

We demonstrate and distinguish experimentally the existence of dark Fano resonances at $k \approx 0$ in a macroscopic two-dimensional photonic crystal slab. We fabricate a square array of holes in silicon nitride layer and perform an angular resolved spectral analysis of the various Fano resonances. We elucidate their radiation behavior using temporal coupled-mode theory and symmetry considerations. The unique simplicity of this system whereby an ultra-long lifetime delocalized electromagnetic field can exist above the surface and consequently easily interact with added matter, provides exciting new opportunities for the study of light and
matter interaction.

5.1.1 Introduction

The realization of high quality factor cavities in photonic crystals has led in the past two decades to experimental observations of novel physical phenomena in both fundamental and applied research [10,33,74,75,78,92–98]. Modes supported by such cavities fall into two categories: 1) pure modes with infinite lifetimes that lie outside the light cone and 2) resonant modes with finite lifetimes that lie within the light cone and consequently can couple to radiation modes. A proposed surprising exception to the latter involves dark Fano resonances of a macroscopic two-dimensional periodic photonic crystals slab, whose lifetimes are predicted to approach infinity as their crystal wavevector, $k$, approaches zero within the light cone [34,41,99]. The only possibility for these dark Fano resonances to completely decouple from the continuum of free-space modes is by mismatching their symmetries. It is the periodic nanostructure that determines the symmetry of the modes and the macroscopic large area that enables their approaching-to-infinity lifetime. In this work, we employ a centimeters square photonic crystal slab to demonstrate and distinguish $k=0$ non-degenerate Fano resonances with quality factors as high as $10^4$ that extend over $10^8$ unit cells. The photonic crystal, fabricated using interference lithography, consists of a square lattice array of holes in Si$_3$N$_4$ layer with periodicity of 320 nm. Through angle-resolved spectral measurements and temporal coupled-mode theory, we determined the resonances’ quality factors and the various physical mechanisms that govern their value. Using symmetry considerations, we elucidate the behavior of the different resonances at $k=0$. The physical origin of Fano resonances in PhC slabs lies in the coupling between the guided modes supported by the slab and external plane waves, which occurs because of the periodic modulation of the dielectric constant. Typically all these Fano resonances have long lifetimes or high quality factors ($Q$), but there is a subset of them whose $Q$'s have been proposed to approach infinity. In theory, in a perfect infinite periodic PhC slab, due to symmetry considerations, a subset of Fano resonances at $k=0$ have been predicted to completely decouple from the external world with infinite radiative quality factor ($Q_{\text{rad}}$) despite lying within the light cone [34,41,99]. For $k$ near zero, these unique guided resonances have ultra-long (but finite) lifetime, providing an efficient means to couple light in and out of the slab. In practice due to the finite size of any experiment, the incoming and outgoing beams always include wavevectors.
with $k>0$, and hence the resonance lifetime is finite. Although this very unique behavior of Fano resonances in PhC slabs has been discussed theoretically [34,41,99,100], experimental verification of high-Q Fano resonances near $k=0$ over a macroscopically large area has yet to be demonstrated. The key challenge in observing these resonances is that in practical structures, in addition to limits imposed by material absorption, fabrication imperfections partially break the crystal symmetry which results in coupling of these Fano resonances to radiating modes. In addition, the mode itself needs to extend over a macroscopic area in order to support high $Q_{rad}$, posing a significant fabrication challenge.

5.1.2 Measured band diagram

Realizing high quality-factor resonances in photonic nano-structures requires both the careful consideration of the bulk material properties and the sub-wavelength structure geometry. Material absorption sets the upper bound of the attainable quality factor, while the structure geometry can be optimized to minimize scattering due to surface roughness and non-uniformities of the periodic structure. A favorable candidate for achieving high quality factor resonances in the visible is a slab of $\text{Si}_3\text{N}_4$ deposited on top of microns thick oxide layer of a silicon wafer [101]. With refractive index of 2.02, $\text{Si}_3\text{N}_4$ provides sufficient index contrast with the $\text{SiO}_2$ below and air or fluids on top. We fabricated large area square
lattice PhC (see Appendix 5.A) with periodicity of 320 nm and unit cell consisting of a 55 nm deep, 160 nm in diameter cylindrical hole in a 250 nm thick Si$_3$N$_4$ layer (Fig. 5-1). Uniform periodic patterns were obtained on samples as large as 3 cm$^2$. We performed optical characterization of the PhC slab using a supercontinuum laser source at small incident angles, $\theta$, measured from the normal to the PhC plane towards the x-axis (see Appendix 5.B). The reflection spectra as a function of angle, for two orthogonal pump polarizations are presented in Fig. 5-2(a) and (d) revealing eight energy bands. To corroborate these results we used finite difference time domain simulation [38] to calculate the modes of the PhC (see Appendix 5.C). Figure 5-2(c) and (f) show the dispersion curves of the eight lowest energy bands along the $\Gamma$-X line ($k(\Gamma) = [0, 0].(2\pi/a)$, $k(X) = [0.5, 0].(2\pi/a)$, $k = [k_x, k_y]$ and $k_x = (\omega/c)\sin\theta$). The four lower frequencies bands are TE-like (numbered 1-4) and the four higher frequencies are TM-like (numbered 5-8). The presented $E_z$ component of all eight modes are calculated at the center of the Si$_3$N$_4$ layer at $k = [0.01, 0].(2\pi/a)$. The calculated resonant wavelengths are shifted by not more than $\pm 0.5\%$ from the measured spectra, well within the uncertainty of the measured periodicity or the value of the refractive index. Exception to that is the TE-like mode number 2 in Fig. 5-2(a) that appear to be very faint (almost missing): we explain the cause for this later.

It is evident from the measured spectral reflectivity of Fig. 5-2(a) and (d) that the incident beam may excite different modes of the PhC depending on its polarization. This can be understood from symmetry considerations: exciting the PhC slab with a source of one type of symmetry results in coupling to the modes of the same type of symmetry only. Note that moving away from $\Gamma$ to X the symmetry group changes from $C_{4v}$ to $C_{1h}$ [102], reducing the number of irreducible representations from 5 to 2. Mirror reflection operation around the x-axis leaves the modes of one irreducible representation unchanged, while the modes of the other irreducible representation are altered by a factor of -1. We can determine the symmetry of each mode by examining the mode profile of its $E_z$ component as shown in Fig. 5-2(c) and (f). Modes 1, 2, 4, and 6 are altered by a factor -1 under mirror reflection operation around the x-axis and hence excited by $E_y$ polarized source, while modes 3, 5, 7, and 8 are unchanged under the same operation and hence excited by $E_x$ polarized source.
Figure 5-2: Band diagrams of the PhC obtained from reflectivity measurement and finite difference time domain (FDTD) simulation. Reflectivity measurements of the PhC with (a) $E_y$ and (d) $E_x$ polarized beam. The inset shows a schematic of the experimental setup. (b), (e) A slice of the reflectivity spectrum at 1.8°. (c), (f) Band diagram of the eight lowest energy modes (measured at the $\Gamma$ point) of the PhC obtained from FDTD simulation. The four lower frequencies modes (numbered 1-4) are TE-like and the four higher frequencies (numbered 5-8) are TM-like. Modes excited externally by odd (even) polarized source with respect to the $x$-axis are colored purple (green); other modes are shown with gray dashed lines. Their $E_z$ field profiles at the center of the Si$_3$N$_4$ layer at $k = [0.01, 0](2\pi/a)$ are also shown. Contour of the hole is shown with black dashed circle. The inset depicts a schematic of the unit computational cell used in the numerical calculation. By applying periodic boundary conditions, the simulated structure becomes periodically infinite.
5.1.3 Measured quality factors

Figure 5-3 depicts the calculated $Q_{\text{rad}}^{\text{total}}$ of these eight bands. It reveals that while the doubly-degenerate (at $\Gamma$) bands 3, 4 and 6, 7 have finite $Q_{\text{rad}}^{\text{total}}$ at $k \approx 0$, the singly-degenerate (at $\Gamma$) bands 1, 2, 5, and 8 have $Q_{\text{rad}}^{\text{total}}$ that go to infinity when approaching $k = 0$. This can be qualitatively understood from symmetry arguments. As mentioned earlier, a mode at the $\Gamma$ point belongs to one of five irreducible representations of the $C_{4v}$ point group [34,102]. One of the irreducible representations is doubly degenerate and has the same symmetry as free-space modes, while the rest are all singly degenerate and are completely decoupled from free-space modes. As a result, $Q_{\text{rad}}^{\text{total}}$ of these four singly-degenerate modes at the $\Gamma$ point should be infinite despite lying within the light cone, while the doubly-degenerate modes have finite $Q_{\text{rad}}^{\text{total}}$. As we move away from $\Gamma$ to $X$ the point group becomes $C_{1h}$ and doubly-degenerate modes split into two. The two irreducible representations of the $C_{1h}$ point group share symmetry with the free-space modes and therefore $Q_{\text{rad}}^{\text{total}}$ become finite for all resonances, as is evident from the calculation.

To gain a deeper insight into the physics of the measured resonances, we developed a semi-analytical temporal coupled-mode theory model (see Appendix 5.D) that accounts
for the presence of guided leaky resonances in the Si$_3$N$_4$ layer [33, 34]. We excited the
model with an incident source propagating from the top and impinging onto the Si$_3$N$_4$ layer
resonant cavity. From first-order perturbation to Maxwells equation, energy conservation
considerations, and neglecting second-order effects, we attained the following expression for
the reflectivity of our sample:

$$|r_{phc}|^2 = \left| \frac{r_d - \gamma_{tol} \cdot (\gamma_{tol} r_d + \gamma_{SiO_2} t_d)}{i(\omega - \omega_0) + \gamma_{tol}^2/2 + \gamma_{SiO_2}^2/2 + 1/\tau_{loss}} \right|^2 \quad (5.1)$$

$r_d$ and $t_d$ are the complex reflection and transmission coefficients of the sample without the
square lattice of cylindrical air holes. $\gamma_{tol}$ and $\gamma_{SiO_2}$ are the coupling strengths of the resonant
mode to the top environment and the SiO$_2$ layer respectively, and can be related to the
quality factors by $\gamma_{SiO_2}^2 = \omega_0/Q_{SiO_2}^2$ and $\gamma_{tol}^2 = \omega_0/Q_{tol}^2$. From Eq. (5.1), it becomes obvious
that there exist two temporal pathways: $r_d$ represents the direct transmission and reflection
processes of the uniform stack, and the second term represents the guided resonances excited
within the Si$_3$N$_4$ layers whose energy leaks into the far-field. It is the superposition of the
two physical processes that contribute to the typical narrow Fano line shapes superimposed
on a Fabry-Perot-like background that are observed in the reflectivity spectra of Fig. 5-2(b)
and (e). We fitted Eq. (5.1) to the measured spectra (see Appendix 5.D) and obtained the
corresponding $Q^{total}$, defined as $1/Q^{total} = 1/Q_{rad}^{total} + 1/Q_{loss}^{total}$, where $Q_{loss}^{total}$ includes losses
from both material absorption and scattering due to fabrication imperfections. The results
are summarized in Fig. 5-4, with an example of a fitted Fano resonance curve for the data
measured at $0.8^\circ$ of band 5. A complementary approach that also provides further intuitive
understanding to calculate the reflection from such a structure was proposed by Pottage
et. al. [103].

Figure 5-4 reveals a clear distinction between the singly-degenerate (modes 1, 2, 5, and 8)
and the doubly-degenerate (modes 3, 4, 6 and 7) modes at small angles. While the measured
value of $Q^{total}$ increases when approaching $k=0$ for modes 1, 5, and 8, the doubly-degenerate
modes have decreasing or fixed values. We note that although $Q^{total}$ as high as $10^4$ are
observed, the calculated $Q_{rad}^{total}$ (Fig. 5-3) of the singly-degenerate modes are much greater
at small angles, suggesting that close to $k=0$ the resonant energy decay is dominated by
absorption and incoherent scattering from fabrication imperfections ($Q^{total} \approx Q_{loss}^{total} \approx 10^4$),
both of which could be significantly reduced by improving the fabrication process. On the
Figure 5-4: $Q^{\text{total}}$ values retrieved by fitting Eq. (5.1) to the measured data. Insets show the reflectivity spectra of leaky mode 5 measured at three angles (0.1°, 0.4°, and 0.8°). The right inset depicts an example of the curve fitting process discussed in the text. Note the distinct higher quality factors of the singly-degenerate modes close to zero angle (i.e. zero wave vector).

other hand, the four low-$Q$ bands 3, 4 and 6, 7 in Fig. 5-4 have $Q^{\text{total}}$ values that are comparable to the calculated $Q^{\text{rad}}$ and smaller than $Q^{\text{total}}$. Indeed, FDTD calculations of the resonant mode show that the energy confinement is approximately unchanged within the plotted range of angles, suggesting that $Q^{\text{scat}}$ is relatively constant in the considered range of angles. Apart from limiting the values of $Q^{\text{total}}$ and hence the linewidth of the resonant lineshapes, the presence of relatively large scattering loss and absorption compared to far-field radiation near normal incidence leads to reduced resonant amplitudes. Conversely, the decrease of $Q^{\text{rad}}$ away from the normal provides a better match between $Q^{\text{scat}}$ and $Q^{\text{total}}$, which leads to an increase in the height of the features. This is consistent with Eq. (5.1), and also explains why band 2 appears only weakly in the measurement results shown in Fig. 5-2(a). Unlike other high $Q^{\text{total}}$ modes whose values decrease rapidly away from the $\Gamma$ point, the $Q^{\text{rad}}$ of the missing TE-like band 2 remains high (Fig. 5-3) for most angles, resulting in small reflectivity amplitudes which are harder to detect.
5.1.4 Conclusion

We experimentally differentiate and demonstrate the existence of a special class of resonances in PhCs with quality factors that could, in principle, approach infinity despite lying within the light cone. These non-degenerate Fano resonances are delocalized modes that decouple from the light cone states at $k=0$ due to symmetry considerations. A clear distinction between these modes and degenerate Fano resonances with finite $Q^{\text{total}}$ at the $\Gamma$ point is presented. With future improved fabrication that decreases the roughness and non-uniformities of the PhC slab, the current observed quality factors of $\sim 10^4$ can be significantly enhanced. The experimental realization of this mode has several important consequences: 1) the strongly enhanced field close to the PhC surface and the simple access to it provides a new platform for the study of light and matter interaction; 2) it offers an easy-to-fabricate structure that supports delocalized modes with ultrahigh quality factors; 3) it can be shown from coupled mode theory [104] that up to 50% of external radiation can be coupled to these strongly confined modes in symmetric PhC slabs, when one ensures that the Q-matching condition between the radiative life-time, and the absorptive life-time is satisfied; and 4) despite the macroscopically large area resonator, only a few high-Q modes are supported within a fairly broad frequency range. The delocalized nature of this mode is particularly important in applications where the interaction of an enhanced electric field with a macroscopic volume of matter can dramatically improve the performance of the process, such as in bimolecular sensing and organic light emitting devices. Furthermore, the realization of this novel resonance could enable the enhancement and the demonstration of new physical phenomena in laser physics, energy conversion, nonlinear optics, and optical filters.

5.2 Low-threshold organic laser realized with photonic crystal slabs

5.2.1 Introduction

The top surface of the PhC slab of Sec. 5.1, patterned with a sub-wavelength periodic square array of holes, supports Fano resonances with wavefunctions extending above it. The delocalized nature of these resonances, their long lifetimes, and the altered spectral density of states (DOS) of the structure result in a dramatically modified spectral and spatial radia-
tion pattern of the organic molecules when compared to their free space emission [105]. For molecules that are placed close to the surface, sharp spectral features in their fluorescence spectra are observed, with enhancement of the differential radiated power. The enhancement mechanisms could modify the lasing threshold of such system due to two main effects. First, the excitation field is enhanced near the surface of the PhC due to on-resonance excitation. This enables substantial absorption of the pump within a thin layer of diluted molecules near the PhC surface. The second contribution originates from the enhanced emission of the molecules into the lasing mode when compared to their free-space emission with similar modal volume. This enhancement can be introduced phenomenologically into the lasing rate equation through the spontaneous emission coupling factor $\beta$ (see Sec. 6.2.3 of Chapter 6) which is classically defined as the ratio between the emission rate into the laser mode and the total emission rate. The power threshold of laser systems is typically inversely proportional to $\beta$ and hence can be reduced in cases where the emission rate into the laser mode is enhanced while the total rate remains almost constant. In the following, we show that the enhanced excitation and high-$Q$ factor of a dark Fano resonance enables lasing of a 100 nm thin layer of diluted organic dye molecule solution with a threshold intensity slightly below 2 kW/cm$^2$ under pulse excitation.

5.2.2 Lasing measurements and comparisons with model

The lasing experiments with 1 mM R6G organic molecules on top of the PhC slab were conducted with the setup illustrated in Fig. 5-5(b) (see also Appendix 5.E). The pump is a 532 nm second harmonic of a 5 ns excitation width ($\tau_{\text{pulse}}$) collimated Nd:YAG laser at 10 Hz repetition rate with beam diameter of 1 mm. Figure 5-5(a) shows the typical four-level energy levels in the molecules, where the molecular population in each level is labeled as $N_i$ ($i = 0, 1, 2, 3$). In this study, we have disregarded the influence of the triplet states since the lifetime to cross over from the singlet states are at least an order of magnitude longer than the excitation pulse width. The threshold pulse energy can be approximated by Eq. (6.7) of Chapter 6, with $F_p$ set to 1 and assuming that self-absorption losses due to excitation of molecules to the higher $S_2$ singlet states are negligible:

$$U_{\text{thr}}^{\text{pulse}} \approx \frac{hf_p\tau_{\text{pulse}}}{\eta} \times \frac{1/\tau_{\text{spont}} + 1/\tau_{21}}{\beta\Gamma_s/\tau_{\text{spont}}}.$$  (5.2)
Figure 5-5: (a) Schematic drawing of energy levels of the organic molecules under short pulse excitation. Physical processes illustrated include the excitation to higher energy levels due to the absorption of pump photon, non-radiative decay, and radiative transition to lower energy levels. (b) Schematic drawing of the experimental setup in the lasing measurement of R6G dissolved in methanol placed on top of the PhC slab. The grey substrate is the macroscopic PhC slab. The orange spheres are schematic drawing of the R6G molecules in solution. The pump source is a pulsed laser with short excitation width of 5 ns at a repetition rate of 10 Hz. The blue surface represents the equal energy density surface of the pump-mode we are coupling into. Lasing output was recorded using a high-resolution spectrometer placed close to the normal of the PhC.

$\eta$ is the fraction of the pump energy the dye absorbs to excite the molecules to the $S_1$ states, $f_p$ is the pump frequency, $\Gamma_s$ is the confinement factor of the lasing mode in the organic medium, and $\tau_{\text{spont}}$ and $\tau_{\text{21}}$ are the radiative and non-radiative relaxation lifetimes of the excited molecules. $1/\tau_{\text{loss}}'$ is the total loss-rate of the lasing photons due to cavity losses and absorptions in the organic medium.

The active PhC is optically excited with $\lambda_{\text{pump}} = 532$ nm, incident at $\phi = 10.02^\circ$ for on-resonance pumping. The total absorption of the excitation energy is about 11.7% (see Appendix 5.F). Narrow emission line was observed at $\lambda_{\text{ase}} \approx 580$ nm first and then $\lambda_{\text{ase}} \approx 575$ nm corresponding to two of the singly-degenerate TM-like modes near the $\Gamma$-
Figure 5-6: Input-output energy characteristics of the organic PhC slab lasing through the longer wavelength singly-degenerate TM-like mode in Sec. 5.1 under pulsed pumping. The solid lines are analytic predictions from our lasing model while red circles are energies measured using the spectrometer. Green circles are data measured with a power meter. The jump in output power clearly indicates the onset of lasing. Lower inset shows the same results in linear scale, where the output grows linearly with the pump energy beyond threshold. Top inset is the measured power spectrum of emission from the PhC slab at normal incidence below (blue) and above (red) the lasing threshold. Single-mode lasing is attained near the threshold.

Point, where both resonances exist well within the R6G’s emission spectrum. At 580 nm, \( Q_{\text{tot}} \) was measured to be \( 8.3 \times 10^3 \) (see Sec. 5.1), \( Q_{\text{rad}} \) was calculated to be \( 1.9 \times 10^5 \) at the \( k \)-value corresponding to the divergence angle of the lasing beam (\( \sim 0.1^\circ \)), and \( \Gamma_s \) is 6%. The pumping spot size was measured to be more than a millimeter in diameter, while the observed lasing spot size was approximately 250 \( \mu \)m. This discrepancy between the pumping and lasing areas may arise due to long range non-uniformity of the structure that limits the size of the largest coherent area, and also because the higher power within the central core of the pump’s Gaussian beam profile lases first, before the outer less intense region.

Plugging information of electronic transitions in R6G and parameters of the PhC cavity above into the full lasing model of Appendix 6.A in Chapter 6 [which includes the higher
excited singlet states not shown in Fig. 5-5(a)], the pulse energy input-output curve is plotted against the measured data in Fig. 5-6. The jump in the log-log plot clearly indicates the onset of lasing. The same results in linear scale is shown in the right inset, where the output energy grows linearly with the pump energy beyond threshold. The left inset is the emission spectra of the molecules near normal incidence when pumped below (blue) and above (red) the threshold. It shows that single-mode lasing at 580 nm is achieved at 130 nJ even though the existence of another high-Q mode near 575 nm (the other singly-degenerate TM-like mode) is detected below the threshold. Both the theoretically predicted threshold and the slope efficiency match well with the measured data within experimental errors. In particular, the measured threshold energy is about 100 nJ corresponding to an intensity of 1.8 kW/cm², and the quantum slope efficiency is 0.09%. We attribute the low measured threshold of R6G (despite a weak lasing mode confinement of 6%) to (i) the resonant coupling of the pump into the PhC slab leading to 11.7% absorption of the pump energy, (ii) high-Q factors of the dark Fano resonances, and (iii) relatively high spontaneous emission coupling rate into the lasing mode ($\beta \approx 5 \times 10^{-5}$) which is achieved due to the strong enhancement in spectral DOS compared to free space [105]. On the other hand, the low slope efficiency may be attributed primarily to the small fraction of lasing power emitted into the far field because of the relatively high $Q_{\text{rad}}$ (i.e. $0.4 \times Q_{\text{tot}}/Q_{\text{rad}} \approx 1.75\%$)\(^1\).

5.2.3 Conclusion

We demonstrate lasing of a thin layer (100 nm) of diluted organic dye molecule solution with threshold intensity slightly below 2 kW/cm² under pulse excitation. Through coupled mode theory calculation and $Q$-matching arguments in Appendix 5.F, we showed that it is the enhancement of the excitation electric field close to the surface of the PhC slab that enables the efficient absorption of the excitation photons in a diluted system. The ability to simply introduce organic molecules or colloidal nanoparticles to a surface to interact with delocalized resonant modes of the considered nano-structured cavity enables new opportunities in optical molecular sensing and surface light emitting devices, as already evident from the lasing results.

\(^1\)After accounting for the fact that only 40% of the total radiated energy leaks out the top PhC surface and couples to the detector.
5.A Appendix: Fabrication

Silicon nitride layer was grown by LPCVD on top of 6 µm thermally grown SiO₂ layer on a silicon wafer (from Lionix). Using prism coupler, we measured propagation losses in the Si₃N₄ layer to be less than 0.3 dB/cm at 632 nm. The fabrication process of the PhC pattern started by depositing a trilayer resist stack, which consists of a negative photoresist (PR) layer, an SiO₂ intermediate layer, and an antireflection coating (ARC). The thickness of the ARC layer was optimized to minimize back reflection to the PR layer during lithography. The PhC pattern was produced by the interference lithography (IL) system using a 325 nm He/Cd laser. In the IL system, the laser beam is split into two and then interfered to form a standing wave with a period of \( \lambda/(2 \sin \theta) \). A second exposure with the sample rotated by 90° relative to the first exposure defines the two-dimensional grid pattern on the PR layer. The period of the pattern is determined by the angle of incidence, while the diameter of the hole is determined by the exposure dose. After the exposure, the sample was developed in a commercial PR developer. Pattern transfer from the PR layer through the SiO₂ and ARC layers to the Si₃N₄ layer was achieved with reactive ion etching (RIE). CF₄ gas was used to etch the SiO₂ and the Si₃N₄ layers, and He/O₂ gas was used to etch the ARC layer. Due to the anisotropic characteristic of RIE, vertical and smooth sidewalls were produced as well as relatively small lateral roughness compared to wet etched samples. The average period of the pattern and the average hole diameter both had standard deviation of 6 nm. Uniform periodic patterns were obtained on samples as large as 3 cm². For the optical characterization however a smaller sample was needed due to size constrain of the cell used to mount the sample. Thus all measurements shown in this chapter were obtained from 0.7 x 0.7 cm² sample.

5.B Appendix: Optical setup (passive system)

We performed optical characterization of the PhC slab using a supercontinuum laser source (SuperK Compact, NKT Photonics) with spot size of 2 mm at small incident angles, measured from the normal to the PhC plane. The PhC slab was placed in a precision liquid cell containing toluene (n = 1.49), representing a potential future incorporation of the slab into fluidic systems for organic lasers and sensing applications. The liquid cell was mounted on a precision motorized rotating stage (ESP300, Newport) with a resolution of 0.01°. A
schematic of the experimental setup is shown in the inset of Fig. 5-2(a). Passing through a beam splitter, the laser beam is back-reflected from the PhC slab at an angle set by the horizontally rotating stage and collected by a spectrometer with a resolution of 0.03 nm (HR4000, Ocean optics). The polarization of the randomly polarized incoming beam was set using a polarizer to be either $E_x$ (horizontal) or $E_y$ (vertical). Figure 5-2(a) and (d) depict the reflection spectra measured for each of the polarizations at 61 different angles between $0^\circ$ and $2^\circ$ (measured from the normal towards the x-axis).

5.C Appendix: Computation

The computational cell consisted of a 250 nm thick Si$_3$N$_4$ with refractive index of $n = 2.02$ on top of an infinitely thick layer of SiO$_2$ ($n = 1.45$). Hole with depth of 55 nm and diameter of 160 nm was introduced in the center of the top surface of the 250 nm thick Si$_3$N$_4$ layer and was filled with refractive index of $n = 1.49$ corresponding to toluene. Periodic boundary conditions in the in-plane directions were applied with periodicity set to 320 nm. A schematic drawing of the computational cell is shown in the inset of Fig. 5-2(c). The realization of the infinitely thick layers was achieved by burying the out-of-plane boundaries into perfectly matched layer (PML) [38] while leaving enough space between the PML layer and the Si$_3$N$_4$ boundary. In the real structure, the SiO$_2$ and toluene are each a few micrometers thick making the evanescent tail of the guided resonance field negligible at their outer boundaries, therefore justifying modeling effectively infinitely thick layers in the numerical calculations. We used the Harminv tool of MEEP [38] to calculate the resonant frequencies of the structure and their radiative quality factors.

5.D Appendix: Coupled-mode theory

We assume that the energy of the resonance is allowed to decay exponentially over time into one of the following four channels: (i) far-field radiation into toluene, (ii) far-field radiation into the SiO$_2$ layer, (iii) material absorption in the Si$_3$N$_4$ layer, and (iv) incoherent scattering losses due to fabrication disorder. Coupling into each of these channels can be quantified by correspondingly defining the following quality factors: (i) $Q^\text{tol}_\text{rad} = \omega_0\tau^\text{tol}_\text{rad}/2$, (ii) $Q^{\text{SiO}_2}_\text{rad} = \omega_0\tau^{\text{SiO}_2}_\text{rad}/2$, (iii) $Q^{\text{abs}}_\text{loss} = \omega_0\tau^{\text{abs}}_\text{loss}/2$, and (iv) $Q^{\text{scat}}_\text{loss} = \omega_0\tau^{\text{scat}}_\text{loss}/2$, where $\omega_0$ is the resonant frequency in consideration and $\tau$ is the lifetime over which the field decays by $e^{-1}$. 79
As such, the total radiation into the far-field is characterized by $1/Q_{\text{rad}}^{\text{total}} = 1/Q_{\text{rad}}^{\text{tot}} + 1/Q_{\text{SiO}_2}^{\text{tot}}$, while $1/Q_{\text{loss}}^{\text{total}} = 1/Q_{\text{loss}}^{\text{abs}} + 1/Q_{\text{loss}}^{\text{scat}}$ represents the total loss in the system. At 632 nm, using prism coupling to the Si$_3$N$_4$ layer of the bare wafer, $Q_{\text{loss}}^{\text{abs}}$ of the Si$_3$N$_4$ layer was measured to be higher than $10^5$ (includes scattering losses of the bare wafer). This provides us with an estimate of an upper bound on $Q_{\text{loss}}^{\text{abs}}$ of the PhC slab; $Q_{\text{loss}}^{\text{abs}}$ could be lower than that if the surface was contaminated with absorptive material during the fabrication process.

In order to retrieve the $Q_{\text{total}}^{\text{tot}}$ from the measured data, we first fit the smooth background without the narrow line shapes to retrieve $r_d$ and $t_d$ by curve-fitting the measurement results without the Fano resonances to an analytic formula of a homogeneous slab with an effective refractive index [34]. This accounts for the smooth undulating background in the reflectivity spectrum, as can be seen from the fitted blue line in the right inset of Fig. 5-4. Next, Eq. (5.1) is fitted to the desired Fano resonance of the measured data (red circles in the right inset of Fig. 5-4) using nonlinear least squares method to attain the values of $\gamma_{\text{tot}}^{\text{total}}$, $\gamma_{\text{SiO}_2}$, $\gamma_{\text{loss}}^{\text{tot}}$ and $\omega_0$.

5.E Appendix: Optical setup (lasing system)

The spontaneous emission range for the R6G 590 is between 540 nm and 590 nm. The fluorescence information of the PhC was collected with the spectrometer (HR4000, Ocean optics) having resolution of 0.03 nm, and aligned close to the normal direction because we were mainly interested in the dark Fano resonance along the normal direction of the PhC slab. The dimension of the aperture in the spectrometer is about 5 µm in X direction and 2 mm in Y direction corresponding to an acceptance angle of approximately 1°. By tuning the position of the spectrometer’s observer head with a XYZ stage, we were able to detect different emission angles along $\Gamma - X$ direction or $\Gamma - M$ direction.

5.F Appendix: Fraction of pump power absorbed, $\eta$

Typically in nano-structured resonances, the active volume of the organic material that interacts with the resonance is small (compared to the wavelength), and so, the absorption of the incident excitation beam is only a small fraction of the total incident power in most cases. However, with resonant cavities at the excitation wavelength the interaction lifetime can be orders of magnitude higher than in free space and hence the absorption can be strongly
enhanced. Using coupled mode theory and applying the $Q$-matching arguments [104], the fraction of incidence power ($P_{\text{in}}^\lambda_{\text{pump}}$) absorbed by the molecules at resonance within a $d_{\text{eff}}^\text{pump}$-thick layer above the PhC slab extended by the resonant wavefunctions can be shown to be:

$$\eta = \frac{P_{\text{resonance}}}{P_{\text{in}}^\lambda_{\text{pump}}} = \frac{2\lambda_{\text{pump}}}{\pi n} \frac{\Gamma_p (Q_{\text{tot}}^\text{pump})^2}{Q_{\text{rad}}^\text{pump}} \times N_0 \sigma_{\text{abs}}$$  \hspace{1cm} (5.3)

where $\lambda_{\text{pump}}$ is the pump wavelength, $n$ is the refractive index of the liquid, $Q_{\text{rad}}^\text{pump}$ and $Q_{\text{tot}}^\text{pump}$ are the radiative and the total quality factors of the resonance mode that the excitation beam is coupled to, $\Gamma_p$ is the fraction of pump mode energy within the volume of the organic molecules, $\sigma_{\text{abs}}$ is the absorption cross-sections of molecules at the excitation wavelength and $N_0$ is the number density of molecules. Note that all quantities in Eq. (5.3) can be found either by FDTD simulations or reflection measurements of the passive structure.

The effective thickness $d_{\text{eff}}^\text{pump}$ of the pump mode is defined as the thickness of the layer on top of the PhC surface where the energy density in the region is $(1 - e^{-2})$ of the total energy density beyond the top surface of the PhC. Here, we get $d_{\text{eff}}^\text{pump} \approx 100$ nm. Furthermore, $Q_{\text{rad}}^\text{pump} = 1.6 \times 10^4$ through FDTD simulation; $Q_{\text{tot}}^\text{pump} = 3100$ obtained from reflection measurements. $\sigma_{\text{abs}} = 3.8 \times 10^{-20} m^2$ for R6G and $N_0 = 6 \times 10^{23} m^{-3}$ at 1 mM; $\Gamma_p = 6\%$ from FDTD simulation; $n = 1.33$ for methanol; $\lambda_{\text{pump}} = 532nm$; therefore, $Q_{\text{abs}}^\text{pump} = 2\pi n / \lambda_{\text{pump}} \sigma_{\text{abs}} N_0 \Gamma_p = 1.1 \times 10^4$. From Eq. (5.3), $\eta = P_{\text{resonance}} / P_{\text{in}}^\lambda_{\text{pump}} \approx 11.7\%$, meaning the absorption from the 100 nm evanescent tail of the Fano resonance mode is about 11.7\% for on-resonance coupling. Note that $Q_{\text{rad}}^\text{pump}$ above has taken into the fact that only 40\% of the total radiated energy leaks out of the top PhC surface and couples to the pump.
Chapter 6

Threshold and dynamics behavior of organic nanostructured lasers

We investigate the laser dynamics in systems of sub-wavelength photonic structures consisting of organic dye molecules. To this end, we have developed a comprehensive theoretical framework based on time-dependent perturbation theory able to accurately describe the interaction of organic molecules with a micro-structured cavity to produce single-mode lasing. The formalism provides explicit analytic expressions of the threshold and slope efficiency that characterize this class of lasers, and also the duration over which lasing action can be sustained before the dye photobleaches. Both the chemical properties of the dyes and optical properties of the cavities are considered in the model. We verified this theoretical model by comparing the numerically predicted threshold and slope efficiency values to those measured in experiments under short pulse excitations in Chapter 5. We also systematically studied the feasibility of lasing under continuous-wave excitations in optically pumped monolithic organic dye lasers. This study suggests routes to realize an organic laser that can potentially lase with a threshold of only a few $W/cm^2$. In addition to their fundamental scientific interest, this work provide formalisms that could enable the development and advancement of sub-wavelength structured organic-based light emitting and sensing devices.

6.1 Introduction

Organic dye lasers with high tunability in the visible wavelengths have attracted interests for many years due to their low-cost processing, flexible choice of substrates, and large emis-
sion cross sections that can cover the spectral region from ultraviolet to the near infrared. Electrically pumped organic semiconductor lasers are desirable from a practical standpoint but their realizations have remained elusive due to their high thresholds which is hard to attain with the large losses at the electrical contacts, low charge-carrier mobility in organic materials, and efficient exciton annihilation process in solid-state organic media [106–110]. A compromise to the above is the hybrid electrically pumped organic lasers [111,112]. These lasers are optically pumped by small electrically driven inorganic diode lasers and thus, do not suffer from the disadvantages of the direct electrical pumping scheme pointed out above. While optically pumped organic lasers have been widely demonstrated [113,114], lasing is only possible with high peak power excitation sources of short pulses; no continuous-wave (CW) operated organic lasers has been demonstrated without liquid dye circulation. A motivation of this work is to explore the feasibility of achieving low-threshold lasing in a continuous optically pumped monolithic organic laser, where lasing action is sustained over a time scale longer than all decay lifetimes in the organic medium. To achieve this, one would need to (i) lower the thresholds of existing organic lasers to levels that commensurate with most commercial applications (typically few W/cm²) [115], (ii) minimize the photobleaching effects in the organic media, and (iii) ensure thermal management of the devices. In this work, we assumed that the thermal effects produced by localized heating of the dyes under excitations are minimized through careful design of the devices [116,117]. We focus instead on optimizing the chemical properties of the dyes and optical properties of the cavities to achieve (i) and (ii), assuming a photobleaching rate determined by the system chemical composition and packaging. To do so, it is critical to develop a complete theoretical understanding of the physical mechanisms underlying lasing action in organic lasers. The purpose of the chapter is to present such an analysis. Specifically, we introduce a theoretical framework that extends previous studies on organic dyes [118,119] in order to account for the full multi-level rate equations, the photobleaching of dyes, and the enhancement effects of microcavities due to the modification of the density of states (akin to the Purcell effect). The formalism provides explicit analytic expressions of the threshold and slope efficiency that characterize this class of lasers, as well as the duration over which lasing action can be sustained before the dye photobleaches. In particular, using a solution of R6G dye molecules as the gain medium, our model suggests routes to attain a low-threshold organic laser that can potentially lase at a few W/cm². We expect our findings to open a
path to the realization of cheap and compact organic sources based on the hybrid electrical pumping scheme, which will in turn open up several potential applications in the visible spectrum.

The chapter is organized as follows: Section 6.2 (and Appendix 6.A) discusses the general theoretical model used throughout this work. Section 6.2.3 focuses on the lasing actions in organic lasers under pulse excitations, where results predicted by the theoretical model are verified with measurements in a pulse laser experiment consisting of R6G dye solution on top of a photonic crystal slab (see Chapter 5). Section 6.3 presents a detailed analysis of lasing action in organic lasers under CW excitations. Finally, in Section 6.4 we provide a set of conclusions of this work.

6.2 Theoretical framework

As a general energy-level model of an organic molecule, we consider the three lowest singlet and two lowest triplet states. Figure 6-1 illustrates the energy level diagram and the different energy transfer mechanisms that form the basic structure of our model. Often, a four-level gain model of only the two lowest singlet states ($S_0$ and $S_1$) suffices in analyzing systems where the influences of the triplet states are minimized, either by pulsed excitations with widths much shorter than the intersystem crossing lifetimes [120] or by fast circulation of the dye solutions [117,121]. In this work, we considered the full energy-level model shown in Figure 6-1 to identify the key parameters (from both molecular and optical standpoints) that can be optimized in order to simultaneously decrease the thresholds, increase the slope efficiencies, and reduce the photobleaching rates under continuous pumping in monolithic organic lasers. We would like to point out that while the model described here represents fluorescing molecules dispersed in amorphous environment, it can be simply adjusted to other configurations such as solid-state Alq$_3$:DCM (based on Förster energy transfer) and crystalline organic molecules by modifying the model of energy levels. For demonstration in this work, the model is applied to the case of optically-pumped lasing systems consisting of R6G in solution under pulsed and CW excitations.

The various processes in the gain model are described via the rate equations (see Appendix 6.A) which are presented in a form that is convenient for numerical analysis, and yet allow for direct relations to experimental parameters. The most general description of an
Figure 6-1: Laser system consisting of an organic active medium embedded in a passive cavity. Optical properties of the laser cavity are the spontaneous emission enhancement factor $F_p$, spontaneous emission coupling factor $\beta$, and total quality factor $Q_{\text{tot}}$. The energy-level structure of the considered organic medium comprise of the three lower singlet states of $S_0$, $S_1$, and $S_2$ and two lowest triplet states of $T_1$ and $T_2$. Red (blue) solid vertical arrows are optically-induced transitions at the lasing (excitation) wavelength whereas dashed vertical arrow is the radiative decay of molecules (spontaneous emission). Wavy arrows are non-radiative transitions. Intersystem crossing of molecules from the singlet to triplet states is characterized by the decay lifetime $\tau_{\text{isc}}$ while the subsequent decay of triplets back to the ground singlet state is characterized by $\tau_l$. Photo-bleaching in the organic medium is modeled as the loss of molecules from the triplet state at a rate of $1/\tau_{\text{bleach}}$. 
energy level would include terms representing the optical transitions that are characterized by their absorption and emission cross-sections, as well as the radiative and non-radiative relaxations that are characterized by their lifetimes. Optical transitions of molecules to the upper states act as resonant absorbers of the excitation and emitted light while those to the lower states result in the stimulated emission of light, producing lasing photons when the pump power is above the threshold. On the other hand, radiative relaxation of molecules to the ground singlet state leads to the spontaneous emission of light. The non-radiative processes considered are the internal conversions due to rotational and vibrational relaxations, the intersystem crossing of the molecules from the excited singlet state $S_1$ to the triplet state $T_1$, as well as their subsequent de-excitations back to the ground state $S_0$. Since $T_1$ is energetically closer to $S_1$ than $S_0$, the transition rate between $S_1$ and $T_1$ is typically faster than that between $T_1$ and $S_0$, resulting in build-up of the triplets population. High occupancies in the triplet states have three detrimental effects on lasing action: the population inversion becomes limited due to the smaller number of molecules in the singlet states, the fraction of the excitation light absorbed by the triplet states is increased, and dye absorption of the stimulated light is enhanced due to the overlapping emission and triplet-absorption spectrum. Our treatment allows for the modifications of the intersystem crossing rates between the singlet and triplet states in order to reduce the effects of such triplet excited-state absorption. In organic dye solution for example, quenchers can be used to decrease the decay lifetime of the triplets [122,123]. Photobleaching is also included in our analysis and is modeled as the loss of the molecules in the triplet state [124,125] at a rate that is dependent on the excitation conditions (i.e. pump power and the presence of oxygen).

The organic gain model described above is coupled to the phenomenological rate equation describing the population of the lasing photons. These photons are produced by stimulated (above the threshold) and spontaneous emission, and at the same time, are resonantly re-absorbed by the organic media. In addition, the photons within the laser cavity are also lost due to radiation into the far-field, material absorption in actual devices, and scattering from fabrication disorders. Using the organic laser model of Fig. 6-1 and the notations in Appendix 6.A, lasing action is obtained as follows: dye molecules are primarily excited from $N_0$ to $N_3$ at a rate proportional to the pump power $P_{sre}$. The excited molecules then decay nonradiatively into $N_2$ after a lifetime $\tau_{32}$. A net decay of molecules from $N_2$ to $N_1$
occurs through stimulated and spontaneous emission, as well as nonradiative relaxation. Some of the molecules in \( N_2 \), however, decay to the triplet state of \( N_4 \) and are trapped if \( \tau_{\text{sc}} \ll \tau_1 \). The resulting reduction in the number of molecules in the singlet states leads to a reduced population inversion, which implies a lower gain. Moreover, a high population of \( N_4 \) will also increase the lasing photon absorption and the number of molecules that are permanently lost through photobleaching. Molecules that do make it into \( N_1 \) then decay nonradiatively to \( N_0 \) with a lifetime \( \tau_{10} \). If the pumping rate exceeds the net decay rate of molecules in \( N_2 \), then a population inversion between \( N_2 \) and \( N_1 \) can be easily achieved because \( N_2 \) depopulates at a slower rate compared to \( N_1 \) and \( N_3 \) (i.e. \( \tau_{21} \gg \tau_{10} \approx \tau_{32} \)) to form a metastable state. Note that dye absorptions of the emitted lasing photons also occur in the singlet states where the molecules in \( S_0 \) and \( S_1 \) are optically excited to \( S_1 \) and \( S_2 \) respectively. Lasing occurs beyond a threshold \( P_{\text{thr}} \) when the population inversion between \( N_2 \) and \( N_1 \) leads to a gain that is sufficiently large to overcome the total losses in the dye and cavity.

### 6.2.1 Sub-wavelength structured cavity effects

Additionally, we consider here the sub-wavelength structured cavity effects on the excited state lifetime emission of the molecules. The optical density of states (DOS) is typically modified in such structures and leads to modification of the spontaneous emission when compared to their free-space emission, which requires consideration in the rate equations. We quantify this modification to the spontaneous emission rate centered near the lasing frequency \( f_l \) by the factor \( F_p \). This inclusion in the model allows us to study its impacts on the onset of lasing in novel nanostructured cavity designs. The enhanced spontaneous emission rate at \( f_l \) in the presence of a suitably designed cavity for such purpose is

\[
\gamma_{\text{cav}}^{f_l} = \frac{\gamma_{\text{cav}}^{f_l}}{\gamma_{\text{all modes}}^{f_l}} \times \frac{\gamma_{\text{all modes}}^{\text{bulk}}}{\gamma_{\text{all modes}}^{\text{bulk}}} \beta \times F_p \times \frac{1}{\tau_{\text{spont}}} \tag{6.1}
\]

where \( \beta \) is the spontaneous emission coupling factor, \( \gamma_{\text{all modes}}^{\text{bulk}} = 1/\tau_{\text{spont}} \) is the total emission rate of the bulk organic media, \( \gamma_{\text{all modes}}^{\text{cav}} \) is the total emission rate in the presence of a cavity and \( \gamma_{\text{cav}}^{f_l} \) is the emission rate into the lasing mode. In regular large cavity where no enhancement of the spontaneous emission exists, \( F_p = 1 \).
6.2.2 Continuous-wave lasing system (steady-state analysis)

We analytically solve for the coupled rate equations of Eq. (6.8) to (6.18) in steady state, and extend the results to accommodate pulse excitations in Sec. 6.2.3.

An analytic threshold expression, that accounts for all relevant chemical properties of the organic molecules and optical properties of the cavities, can identify the relative importances of the various parameters in lowering the threshold under practical constraints. These constraints include the upper limit placed on the quality factor of a cavity, the relative intersystem crossing lifetimes $\tau_t/\tau_{isc}$, and the highest concentration of organic dye that one can use before its quantum yield reduces. Similarly, an expression for the lifetime of an organic dye before it photobleaches will allow one to better design a system to prolong the duration over which lasing action is possible. We separate the steady-state analysis into two parts: (i) predictions of the threshold and slope efficiency assuming no photobleaching, (ii) prediction of the time the dye takes to photobleach.

Threshold

The relatively slow photobleaching process can be ignored in the predictions of thresholds since $\tau_{bleach}$ is typically a few milliseconds (in air saturated solution at room temperature) [124,126] while the next longest lifetime is $\tau_t$, which varies between $10^{-7}$ to $10^{-4}$ seconds (in R6G) depending if a solid-state or solution is used, or if a quencher is added. In other words, the effects of photobleaching only kick in after the lasing actions begin. Under such assumption, the total molecular population is conserved in our model (i.e. $dN_{tot}/dt = 0$) such that the gain will become saturable at high pump rate. In this work, we confined to the regime near the lasing threshold where the effects of the gain medium on the field remain linear [78]. We also assumed $N_{tot} = N_{den}^{tot} \times V = N_0 + N_2 + N_4$ since molecules in the other levels are depleted due to their fast relaxation rates, and this assumption breaks down only at very high excitation power. The steady state threshold power $P_{thr}^{src}$ is then found by extrapolation of the linear input-output power curve to zero output:

$$\frac{\eta S_0 S_1 P_{thr}^{src}}{h f_p} = \frac{(F_p/\tau_{spont} + 1/\tau_{21} + 1/\tau_{isc})/\tau'_{loss}}{F_p/\beta V T_s/\tau_{spont} - (1 + \tau_t/\tau_{isc})/N_{tot}^{den} \cdot \tau''_{loss}} \times V \tag{6.2}$$

where $1/\tau'_{loss} = 1/\tau_{loss} + v g_{abs}^{S_0 S_1} \Gamma_s N_{tot}^{den}$ and $1/\tau''_{loss} = 1/\tau_{loss} + v g_{abs}^{S_0 S_1} \Gamma_s N_{tot}^{den}$ are the total loss-rates of the lasing photons due to cavity losses and absorptions in the organic
medium. In deriving Equation 6.2, $\beta \Gamma_s$ is assumed to be much less than one and the relative magnitudes of the self-absorption cross sections are assumed to follow that in Table 6.1 for R6G in solution. Since $\sigma_{\text{abs}}^{S_1S_2} \approx \sigma_{\text{abs}}^{T_1T_2}$, the total photon loss rates in both the $S_1S_2$ and $T_1T_2$ states may be represented by $1/\tau''_{\text{loss}}$ in our equations. We also note that the $\beta V$ quantity is regarded to be volume-independent because $\beta$ generally scales as the inverse of $V$. From the denominator of Equation 6.2, it becomes clear that lasing can only be realized in systems that satisfy

$$\frac{F_p \beta \Gamma_s N_{\text{tot}}}{\tau_{\text{spont}}} > \frac{(1 + \tau/\tau_{\text{isc}})}{\tau''_{\text{loss}}}.$$  

The above relationship implies that in laser systems where the intrinsic losses are higher than the maximal gain available, lasing cannot be attained regardless of the pump power. To overcome this, one should strive to increase the concentration of the organic media (while taking care to maintain the same quantum yield), increase the confinement and $\beta$ factors through optimizing the cavity designs, and reduce the losses in both dyes and cavities.

**Efficiency**

The quantum efficiency of the laser is proportional to the linear slope of the input-output power relationship above threshold. It can be expressed as

$$\eta_{\text{lase}} = \frac{\text{no. of emitted lasing photons}}{\text{no. of absorbed pumping photons}} = \frac{\Delta P_{\text{out}}}{\Delta P_{\text{src}}} \cdot \frac{f_p}{f_t} = \frac{\eta_{S_0S_1} F_p \beta V \Gamma_s / \tau_{\text{spont}} - (1 + \tau/\tau_{\text{isc}}) / N_{\text{tot}} \tau''_{\text{loss}}}{\tau_{\text{rad}} \eta_{S_0S_1} F_p \beta V / \tau_{\text{spont}} \tau_{\text{loss}} + v_0 \sigma_{\text{abs}}} \left[ 1/\tau_{\text{loss}} + (1 + \tau/\tau_{\text{isc}}) / \tau''_{\text{loss}} \right].$$  

The numerator in the second fraction of Eq. (6.4) is equivalent to the denominator of the threshold expression in Eq. (6.2), consistent with the well-known fact that a low-threshold lasing system also has a high slope efficiency. In fact, if we replace the second fraction in Eq. (6.4) with the total photon-loss lifetime of the laser system $\tau_{\text{loss}}$, a simple relationship for the slope efficiency can be found: $\Delta P_{\text{out}} / \Delta P_{\text{src}} \propto \eta_{S_0S_1} \tau_{\text{loss}}^{\text{rad}} / \tau_{\text{rad}} \approx \eta_{S_0S_1} F_{\text{rad}} / \tau_{\text{loss}}^{\text{rad}}$. In other words, the laser’s slope efficiency depends on both the fraction of pump power that is absorbed by the organic medium and the fraction of the emitted power that is radiated into the far-field. It is, however, independent of the quantum yield in the gain medium and its volume.
Photobleaching

Next, we include the photobleaching channel in $N_4$ through which a molecule can become permanently lost and not return to the singlet states to further participate in stimulated emission. In this case, the total molecular population is no longer conserved but reduces according to $dN_{\text{tot}}/dt = -N_4/\tau_{\text{bleach}}$, where $\tau_{\text{bleach}}$ can be markedly different depending on the environment of the dyes (see Appendix 6.A). This sets the time scale $\tau_{\text{system}} = \tau_{\text{bleach}}N_{\text{tot}}/N_4$ beyond which the dye photobleaches and the emitted signal is permanently quenched. Note that $N_4$ and $N_{\text{tot}}$ are time-dependent quantities. However, to obtain an approximate analytic expression for the lasing lifetime (under CW operations) at pump powers above the threshold, the steady-state populations of $N_4$ and $N_{\text{tot}}$ under the condition in $(i)$ are used. We checked the validity of such an approximation by comparing the numerically predicted photobleaching rates using the full rate equations with the analytically predicted ones, and found their comparison to be good (illustrated in Sec. 6.3).

As a passing remark, we point out that our model predicts $\tau_{\text{bleach}}$ to decrease linearly with the excitation power when operating below the lasing threshold. This linear trend of the bleaching rate with pump power is consistent with the experimental observations in Ref. [116].

The photobleaching lifetime upon lasing (a volume-independent quantity) can be expressed as

$$
\tau_{\text{bles}}^{\text{lase}} = \tau_{\text{bleach}} \times F_{\text{lase}}^{\text{bleach}} \\
\approx \tau_{\text{bleach}} \times \frac{N_{\text{den}} \cdot T_s \cdot \Gamma_s^{\text{loss}}}{\tau_t / \tau_{\text{isc}}} \times \left[ \frac{F_p \cdot 2V}{\tau_{\text{spont}}} - v_g \sigma_{\text{abs}} T_1 T_2 \left( 1 + \frac{\tau_t}{\tau_{\text{isc}}} \right) \right].
$$

(6.5)

Beyond this lifetime, the dye photobleaches and lasing shut down. $F_{\text{lase}}^{\text{bleach}}$ is the modification to $\tau_{\text{bleach}}$ afforded by the specific design of the laser system; it depends on the dye concentration and intersystem crossing lifetime ratio ($\tau_t / \tau_{\text{isc}}$), as well as the cavity’s quality factor, confinement factor, spontaneous emission coupling factor and $F_p$. Again, the relative values of the absorption cross-sections in Table 6.1 are assumed in Eq. (6.5). The quantum efficiency of dye bleaching may similarly be derived as

$$
q_{\text{bleach}} = \frac{\text{no. of photobleached molecules}}{\text{no. of absorbed pumping photons}} \approx \frac{N_{\text{tot}} h f_p}{\tau_{\text{system}} \gamma S_0 S_1 P_{\text{src}}}.
$$

(6.6)

$^1$Note that $\tau_{\text{lase}}^{\text{bleach}} > 0$ so long the systems considered can realize lasing [i.e. Eq. (6.3) is satisfied].
Note that \( q_{\text{bleach}} \) is independent of the organic concentration. Its reciprocal \( 1/q_{\text{bleach}} \) provides an estimate of the number of times a molecule is recycled from \( S_0 \) to \( S_1 \) and back to \( S_0 \) before it is lost. Since \( \tau_{\text{system}}^{\text{bleach}} \propto 1/P_{\text{src}} \) as noted above, \( q_{\text{bleach}} \) in Eq. (6.6) is also independent of the excitation power (below the threshold).

6.2.3 Pulsed lasing system

Most lasing experiments involving organic gain materials are excited by short pulses [113, 114, 127], except for systems where liquid dye circulation exist [117, 121]. The key difference of the analysis in this section to that of the steady-state is that the undesirable triplet influences can now be minimized when the excitation pulse-width \( \tau_{\text{pulse}} \) is shorter than the intersystem crossing lifetime \( \tau_{\text{isc}} \) required by the singlet excitons to decay into the triplet states. This decoupling between the singlet and triplet states can be represented in our analytic model by setting \( \tau_{\text{isc}} \) to large values so that \( N_{\text{tot}}^{\text{pulse}} = N_0 + N_2 \) at all times and no photobleaching takes place within the short lifespan of the lasing pulse signal. Physically, a large \( \tau_{\text{isc}} \) implies a quasi steady-state regime where the pulse length is assumed to be longer than all decay lifetimes of the molecules but remains shorter than \( \tau_{\text{isc}} \). We note that the self-absorption of the lasing photons in \( S_1S_2 \) is sometimes neglected because the population of \( S_0 \) is much greater than that of \( S_1 \) so that \( N_0\sigma_{\text{abs}}^{S_0S_1} \) is the more dominating photon loss term in Eq. (6.18). As a result, a four-level gain model of the lower singlet states usually suffices in capturing the main lasing mechanisms. In this work, we included the photon absorption in \( S_1S_2 \) (since \( \sigma_{\text{abs}}^{S_1S_2} \gg \sigma_{\text{abs}}^{S_0S_1} \) in some dyes such as R6G) and show that it plays a non-diminishing role on the threshold values when \( 1/\tau_{\text{loss}} \) in \( 1/\tau''_{\text{loss}} \) becomes relatively small (i.e. \( 1/\tau''_{\text{loss}} \approx v_0\sigma_{\text{abs}}^{S_1S_2}\Gamma_{s}N_{\text{tot}}/V \)). Using the parameters of our lasing experiments conducted in Sec. 5.2 of Chapter 5 (and Ref. [105]), this happens when \( Q_{\text{tot}} = 2\pi f_{\text{thr}}\tau_{\text{loss}} > 5 \times 10^5 \) which is a physically realizable value [128].

By setting \( \tau_{\text{isc}} \) to infinity in Eq. (6.2), the threshold pulse energy in the quasi steady-state regime is approximated as

\[
\frac{E_{\text{thr}}}{\text{pulse}} = \frac{E_{\text{thr}}}{\text{pulse}} \approx \frac{h_f}{\eta_{S_0S_1}} \frac{\tau_{\text{pulse}}}{\tau_{\text{spont}}} \cdot \frac{1}{\eta_{S_0S_1}^{S_0S_1} \Gamma_s} \frac{1}{N_{\text{det}}^{S_0S_1}} \cdot V. \tag{6.7}
\]

In most pulsed lasing systems, \( \tau_{\text{pulse}} \) and \( \tau_{\text{spont}} \) are in fact both on the order of a few nanoseconds and so, the quasi steady-state approximation applied in Eq. (6.7) leads to
an underestimation of the threshold pulse energy. For instance, in the lasing experiment described in Sec. 5.2 of Chapter 5, Eq. (6.7) differs from the threshold obtained through solving the full model (with pulse input of $\tau_{\text{pulse}} = 5$ ns) by a factor of 0.6. Nonetheless, Eq. (6.7) is still useful in identifying the key molecular and photonic parameters needed to lower the lasing threshold. Similar to the steady-state regime of Eq. (6.3), lasing can only take place in systems where the maximal potential gain is greater than the total cavity and molecular losses where $F_p \beta \Gamma s N_{\text{tot}}/\tau_{\text{spont}} > 1/\tau_{\text{loss}}''$. Unlike in Eq. (6.3), these losses are no longer influenced by the triplet states.

6.3 Results and discussion

In this section, we analyze the steady-state operation of the R6G organic laser in terms of its threshold and the duration over which lasing action can be sustained before the dye photobleaches. The optical and chemical properties of the system in Eq. (6.2) and (6.5) are varied and their impacts on the lasing performances are studied. The parameter values used in our examples are listed in Table 6.1 for R6G in solution.

We begin the analysis by verifying the steady-state analytic expressions derived in Sec. 6.2. In particular, the analytic solutions are compared to the numerical solutions of the full model. Figure 6-2(a) illustrates the output intensity $I_{\text{out}}$ (red curve) and photobleaching lifetime (blue curve) of the laser as a function of the excitation intensity $I_{\text{src}}$. $I_{\text{out}}$ plotted corresponds to the quasi steady-state values before the laser emission is quenched [see Figure 6-2(b)]. In this example, $\tau_0$ is assumed to be 10 $\mu$s so that $\tau_0/\tau_{\text{sec}} = 100$ and the threshold is found to be near 1 W/cm$^2$. By plotting the results against $\eta I_{\text{src}}$ in Fig. 6-2(a), we have implicitly assumed that all the incident power is absorbed by the dye. When $\eta < 1$, the threshold intensity would have to be scaled by $1/\eta$. For instance, if only 10% of the incident power is absorbed by the dye laser, then, the threshold becomes 10 W/cm$^2$ instead. We also found that in this example, the quantum slope efficiency of the laser (calculated from the linear input-output intensity plot beyond threshold) is roughly 40%. This matches the prediction of Eq. (6.4) and is also consistent with the simple relationship of $q_{\text{lase}} = \Delta P_{\text{out}} f_p/\eta \Delta P_{\text{src}} f_1 \approx P_{\text{rad}}^{\text{eff}}/P_{\text{loss}}^{\text{eff}}$ presented in Sec. 6.2. Specifically, $P_{\text{rad}}^{\text{eff}}/P_{\text{loss}}^{\text{eff}} = 50\%$ for the passive cavity considered in Table 6.1 and further reduction to 40% arises when the absorption losses in the dye's triplet states are included.
Next, we examine the photobleaching lifetime of the dye system under continuous excitations. From Fig. 6-2(a), \( \tau_{\text{bleach}} \) decreases linearly with the excitation intensity below threshold and clamps at its threshold value upon lasing. To understand this behavior, we first compute the bleaching quantum efficiency in Eq. (6.6) and find \( q_{\text{bleach}} = 5 \times 10^{-5} \). This implies that every molecule in the dye laser can be recycled to produce roughly \( q_{\text{lase}} / q_{\text{bleach}} = 8 \times 10^3 \) emitted photons before it is lost in \( N_4 \). When the pump rate is high, the molecules are recycled through the \( S_0S_1 \) states at a faster rate; each molecule reaches its maximum emission of \( 8 \times 10^3 \) photons in a shorter time period. Hence, the photobleaching rate increases linearly with the excitation intensity. However, once lasing is attained, the additional pumping power above threshold is channeled entirely into the coherently oscillating cavity mode and \( \tau_{\text{bleach}} \) clamps at its threshold value of \( \tau_{\text{lase}}^{\text{bleach}} \) [Eq. (6.5)].

We note that \( q_{\text{bleach}} \) can be reduced by two orders of magnitude to \( 5 \times 10^{-7} \) if an appropriate triplet quencher for R6G is used to reduce \( \tau_{l}/\tau_{\text{lase}} \) from 100 to 1. The agreement between the analytic and numerical solutions is excellent in Fig. 6-2(a) except for the \( \tau_{\text{lase}}^{\text{bleach}} \) value which differs by a factor of \( \sim 1.5 \). The mismatch occurs because the analytic solutions considered the steady-state \( N_4 \) and \( N_{\text{tot}} \) values (without photobleaching) even though they
are in fact time-dependent quantities (see Sec. 6.2). Nonetheless, this discrepancy remains smaller than a factor of 2 in all the examples considered and does not affect the conclusions drawn in this work.

Figure 6-2(b) shows the temporal development of the lasing action in the same laser for three $\eta I_{src}$ values near the threshold. Below threshold at $\eta I_{src} = 0.5 \text{ W/cm}^2$, the spontaneous emission corresponds to roughly one noise photon. When $\eta I_{src}$ is increased to 0.9 $\text{W/cm}^2$, the laser is turned on initially between $10^{-8} \text{ s}$ to $10^{-5} \text{ s}$ where the output intensity reaches more than three orders of magnitude above that of the noise photon. However, the laser shuts down at $10^{-5} \text{ s}$ and the output decreases by two orders of magnitude to a quasi steady-state level before the dye photobleaches. The initial turn-on occurs when $I_{src}$ is high enough to overcome the total losses in the singlet states. As the triplet states begin to populate, the associated absorption losses and trapping of molecules eventually quench the singlets inversion. At higher pumping rate of $\eta I_{src} = 1.6 \text{ W/cm}^2$, the laser is turned on and the stimulated emission of light is sustained even after the triplet excite state absorption kicks in. In all the three cases above, dye emission ceased after $10^{-1} \text{ s}$ due to the molecular losses in $N_4$. Moreover, a series of very narrow spikes whose amplitudes die down with time can be observed upon turn on of the laser. These phenomena of spiking and relaxation oscillations are characteristics of laser systems in which the recovery time of the excited state population inversion is longer than the total decay time of the lasing mode [62].

In Fig. 6-3 and Fig. 6-4, five parameters of the laser system are varied and their effects on the threshold ($\eta I_{src}^{\text{thr}}$) and corresponding photobleaching modification factor ($F_{\text{bleach}}^{\text{lase}}$) are studied. These results are plotted using Eq. (6.2) and (6.5) respectively. For illustrative purpose, the five parameters selected in this study are the total quality factor of the passive cavity ($Q_{tot}$), intersystem crossing lifetime ratio ($\tau_t/\tau_{nc}$), dye concentration, confinement factor ($\Gamma_s$), and the spontaneous emission enhancement factor ($F_p$). Practical values of $Q_{tot}$ range from few hundreds to tens of millions [128] and we expect the threshold intensity to decrease with increasing $Q_{tot}$. A reduced pumping intensity for lasing will in turn lengthen the time it takes for the dye to photobleach. In the following analysis, we varied $Q_{tot}$ with each of the other four parameters above to understand their effects on the laser performances.
6.3.1 Intersystem crossing lifetime ratio \( (\tau_t/\tau_{isc}) \)

**Fig. 6-3(a) and Fig. 6-4(a)**

In Eq. (6.2) to (6.5), the ratio \( \tau_t/\tau_{isc} \) plays an important role in affecting the lasing performances. Specifically, a high \( \tau_t/\tau_{isc} \) will not only increase the lasing threshold but also decrease the slope efficiency and lasing lifetime \( \tau_{bleach} \). These observations can be understood from the accompanying build-up in the triplet state population, \( N_4 = \tau_t/\tau_{isc} \times N_2 \), through which high exciton- and photon-losses occur. Hence, it is desirable that the organic gain media have small values of \( \tau_t/\tau_{isc} \). In R6G, triplet quenchers which affect \( \tau_t \) are readily available and by simultaneously changing the environment of the molecules from solid to solution, \( \tau_t/\tau_{isc} \) can be varied over four orders of magnitudes from 1 to \( 10^4 \). The threshold plot illustrates a cut-off beyond which lasing shut down as \( \tau_t/\tau_{isc} \) increases. This happens when the triplets influence on the system is so high that its associated absorption and exciton annihilation process overwhelmed the stimulated emission of laser light. In other words, Eq. (6.3) cannot be satisfied. Note that high values of the lifetime ratio beyond \( 10^3 \) are typical in solid-state organic materials and based on our results, are major impediments to the attainment of low-threshold CW organic solid-state lasers. On the other hand, as \( \tau_t/\tau_{isc} \) decreases, the triplet states become less populated and the threshold values saturated towards that of a short-pulse system where only the energy transfers in the singlet states are relevant (see Sec. 6.2.3). Unlike the threshold values, the photobleaching factor in Fig. 6-4(a) does not saturate but continue to increase with declining \( \tau_t/\tau_{isc} \). This trend is directly related to the reduced population of the \( N_4 \) state, where loss of the molecules takes place in our model, when \( \tau_t/\tau_{isc} \) is small. In our example, a threshold of \( \eta_{thr}^{src} = 0.7 \text{ W/cm}^2 \) and photobleaching factor of \( F_{bleach}^{lase} = 1125 \) is predicted for the considered CW R6G laser when \( Q_{tot} = 5 \times 10^4 \) and \( \tau_t/\tau_{isc} = 1 \).

6.3.2 Dye concentration

**Fig. 6-3(b) and Fig. 6-4(b)**

In diluted R6G with concentration below \( 5 \text{ mM} \), the quantum yield is high (i.e. \( q_{dye} = 0.9 \)) and \( \tau_{spont} \) is approximately 5 ns. As the concentration increases beyond \( 10 \text{ mM} \), both the quantum yield of the dye and its spontaneous emission lifetime begin to decrease [129]. This reduction in \( \tau_{spont} \) originates from the close proximity among molecules in concentrated
Figure 6-3: Lasing threshold ($\eta I_{thr}^{r}$) as a function of the optical and chemical properties in an organic laser system under continuous excitation. $\eta$ is the fraction of the excitation power absorbed by the dye. The parameter values used are listed in Table 6.1 for R6G in solution, except for the plot in (d) where $\tau_t$ is assumed to be 10 $\mu$s so that $\tau_t/\tau_{isc} = 100$ instead of 1. In particular, the total quality factor of the passive cavity ($Q_{tot}$) is varied with the intersystem crossing lifetime ratio ($\tau_t/\tau_{isc}$) in (a), the dye concentration in (b), the confinement factor ($\Gamma_s$) in (c), and the spontaneous emission enhancement factor ($F_p$) in (d).
Figure 6-4: Photobleaching modification factor ($F_{\text{bleach}}$) as a function of the optical and chemical properties in a organic laser system under continuous excitation. The parameter values used are listed in Table 6.1 for R6G in solution, except for the plot in (d) where $\tau_1$ is assumed to be $10 \, \mu s$ so that $\tau_1/\tau_{\text{isc}} = 100$ instead of 1. In particular, the total quality factor of the passive cavity ($Q_{\text{tot}}$) is varied with the intersystem crossing lifetime ratio ($\tau_1/\tau_{\text{isc}}$) in (a), the dye concentration in (b), the confinement factor ($\Gamma_s$) in (c), and the spontaneous emission enhancement factor ($F_p$) in (d).
solutions which in turn lead to higher collision rates. In this example, we model all decay lifetimes in the molecules to decrease at the same rate as $\tau_{\text{spont}}$ at high concentration levels. From the threshold plot, we found that a minimum dye concentration is required for lasing to take place [i.e. Eq. (6.3) is satisfied] when the cavity losses are high; dilute organic solutions cannot provide sufficient gain to overcome the total losses of the system. On the other hand, concentrated R6G solution above $10 \text{ mM}$ is also undesirable because $q_{\text{dye}}$ begins to decrease causing the threshold to rise rapidly. Physically, a low $q_{\text{dye}}$ implies only a small fraction of the molecules excited into $N_2$ contributes to the emission of light. This leaves $(5 - 10) \text{ mM}$ to be the optimal concentration to operate the laser with the lowest threshold. In Fig. 6-4(b), the photobleaching factor is observed to increase with concentration (up to $10 \text{ mM}$) for all values of $Q_{\text{tot}}$. This falls directly from the fact that a highly concentrated solution consists of more molecules (assuming the same volume of dye) able to participate in the excitation and de-excitation processes, and be recycled through the $S_0S_1$ states before they become quenched from photobleaching.

6.3.3 Confinement factor ($\Gamma_s$)

Fig. 6-3(c) and Fig. 6-4(c)

A higher confinement factor leads to larger gain in the system for the same excitation level, and hence, the threshold decreases with increasing $\Gamma_s$. This reduction in the threshold excitation intensity in turn increase the photobleaching factor, as observed in Fig. 3-4(c), and equivalently slows down the photobleaching rate. However, the decrease in the threshold and photobleaching rate is dampened by the enhanced self-absorption of the dye at higher $\Gamma_s$, and hence, only a gradual improvement of the laser performances with increasing $\Gamma_s$ is observed. The above description is applicable when $\Gamma_s > 0.1$ for the range of $Q_{\text{tot}}$ considered. When $\Gamma_s < 0.1$, the laser performances deteriorate rapidly.

6.3.4 Spontaneous emission enhancement factor ($F_p$)

Fig. 6-3(d) and Fig. 6-4(d)

In this example, $\tau_t$ is assumed to be $10 \mu s$ so that $\tau_t/\tau_{\text{sc}} = 100$ instead of 1 as considered in Table 6.1. $F_p$ is the enhancement to the spontaneous emission rate of the bulk organic dye ($1/\tau_{\text{spont}}$) in the presence of a cavity. It can be high in microcavities ($F_p = 75$) with
6.4 Conclusion

We have presented a comprehensive theoretical analysis of lasing action in systems of subwavelength photonic structures consisting of organic dye molecules. To this end, we have developed a theoretical framework based on time-dependent perturbation theory able to accurately describe the interaction of organic molecules with a micro-structured cavity to produce single-mode lasing. Both the chemical properties of the dyes and optical properties of the cavities are considered in the model. The formalism provides explicit analytic expressions of the threshold and slope efficiency that characterize this class of lasers, and also the duration over which lasing action can be sustained before the dye photobleaches. To illustrate this theoretical framework, we have analyzed in detail the case of lasing action from optically pumped monolithic R6G organic dye lasers. Our simulations have shown that for this system, with exemplary parameters listed in table 6.1, a threshold $\eta T_{\text{thr}}$ of a
few W/cm² can be achieved. In addition to their fundamental scientific interest, this work provide formalisms that could enable the development and advancement of sub-wavelength structured organic-based light emitting and sensing devices.

6.A Appendix: Rate equations and parameter values of organic lasing model

In this appendix, we first present the set of the spatially-averaged coupled rate equations that are used to model the organic gain medium described by the processes shown in Fig. 6-1. As a general energy-level model of an organic molecule, we consider the three lowest singlet and two lowest triplet states. We then relate the optical transition terms of our model to the experimentally-measured cross-sections σ, and show how such formalism can incorporate the cavity enhancement effects through the spontaneous emission enhancement factor Fp and spontaneous emission coupling factor β.

The rate equations of the molecular populations in each level (N0 to N9), and the phenomenological rate equation describing the lasing photon number φs are

\[
\frac{dN_0}{dt} = -\frac{P_{src} \eta_s S_0 S_1}{\hbar f_p} (N_0 - N_3) - v_g \sigma_{abs} S_0 S_1 \Gamma_s \phi_s \frac{(N_0 - N_2)}{V} + \frac{N_1}{\tau_{10}} + F_p \left(1 - \frac{1}{\tau_{cav}}\right) N_2 \quad (6.8)
\]

\[
\frac{dN_1}{dt} = +\frac{F_p}{\tau_{spont}} \frac{\beta_{cav}}{\Gamma_s \phi_s} (N_2 - N_1) + N_2 \left(F_p \frac{\beta_{cav}}{\tau_{spont}} + \frac{1}{\tau_{21}}\right) + \frac{N_4}{\tau_{41}} - \frac{N_1}{\tau_{10}} \quad (6.9)
\]

\[
\frac{dN_2}{dt} = -\frac{F_p}{\tau_{spont}} \frac{\beta_{cav}}{\Gamma_s \phi_s} (N_2 - N_1) - v_g \sigma_{abs} S_2 S_1 \Gamma_s \phi_s \frac{(N_2 - N_8)}{V} - N_2 \left(F_p \frac{\beta_{cav}}{\tau_{spont}} + \frac{1}{\tau_{21}} + \frac{1}{\tau_{sc}}\right) - \frac{P_{src} \eta_s S_1 S_2 (N_2 - N_9)}{\hbar f_p} + v_g \sigma_{abs} S_1 S_2 \Gamma_s \phi_s \frac{(N_0 - N_2)}{V} + \frac{N_3}{\tau_{32}} \quad (6.10)
\]

\[
\frac{dN_3}{dt} = +\frac{P_{src} \eta_s S_0 S_1}{\hbar f_p} (N_0 - N_3) + \frac{N_3}{\tau_{32}} - \frac{N_3}{\tau_{32}} \quad (6.11)
\]

\[
\frac{dN_4}{dt} = -\frac{P_{src} \eta_s T_1 T_2 (N_4 - N_7)}{\hbar f_p} - v_g \sigma_{abs} T_1 T_2 \Gamma_s \phi_s \frac{(N_4 - N_6)}{V} + \frac{N_2}{\tau_{sc}} + \frac{N_5}{\tau_{54}} - \frac{N_4}{\tau_{bs}} \quad (6.12)
\]

\[
\frac{dN_5}{dt} = +\frac{N_6}{\tau_{65}} - \frac{N_5}{\tau_{65}} \quad (6.13)
\]

\[
\frac{dN_6}{dt} = +v_g \sigma_{abs} T_1 T_2 \Gamma_s \phi_s \frac{(N_4 - N_6)}{V} + \frac{N_7}{\tau_{76}} - \frac{N_6}{\tau_{65}} \quad (6.14)
\]

\[
\frac{dN_7}{dt} = +\frac{P_{src} \eta_s T_1 T_2 (N_4 - N_7)}{\hbar f_p} - \frac{N_7}{\tau_{76}} \quad (6.15)
\]
\[
\frac{dN_8}{dt} = + v_g \sigma_{\text{abs}}^S_{S_1} \phi_s \frac{(N_2 - N_8)}{V} + \frac{N_9}{\tau_{81}} - \frac{N_8}{\tau_{83}} \\
\frac{dN_9}{dt} = + \frac{P_{\text{src}}}{h \nu_p} \eta_{S_1 S_2} \left( \frac{(N_2 - N_9)}{N_{\text{tot}}} \right) - \frac{N_9}{\tau_{98}} \\
\frac{d\phi_s}{dt} = - \frac{v_g \Gamma_s \phi_s}{V} \left[ \sigma_{\text{abs}}^{S_0 S_1} (N_0 - N_2) + \sigma_{\text{abs}}^{S_1 S_2} (N_2 - N_8) + \sigma_{\text{abs}}^{T_1 T_2} (N_4 - N_6) \right] - \frac{\phi_s}{\tau_{\text{loss}}} \\
+ \frac{F_p \beta_{\text{cav}}}{\tau_{\text{spon}}} \Gamma_s \phi_s (N_2 - N_1) + \frac{F_p \beta_{\text{cav}}}{\tau_{\text{spon}}} \Gamma_s N_2.
\]

It is implicitly implied in the above that \( P_{\text{src}}, N_i (i = 0 \text{ to } 9), \) and \( \phi_s \) are time-dependent quantities. \( \Gamma_s \) and \( v_g \) are the confinement factor and group velocity of the lasing mode in the organic region while \( N_{\text{tot}} = \sum_{i=0}^{9} N_i = N_{\text{den}} \times V \) is the total molecular population obtained by taking the product of the molecular density and volume of the organic medium. \( \tau_{ij} \) is the non-radiative decay lifetime of the molecules from level \( i \) to \( j \). \( \tau_{\text{loss}} \) is the photon lifetime in the passive cavity and is related to the total quality factor of the cavity by \( Q_{\text{tot}} = \frac{2\pi f_l}{\tau_{\text{loss}}} \) (\( f_l \) is the lasing frequency). It accounts for the photon losses due to radiation into the far-field (with photon decay lifetime \( \tau_{\text{cav}}^{\text{rad}} \)), scattering from surface roughness and material absorption in actual devices. The laser output is then computed as \( P_{\text{out}} = h \nu_s \phi_s / \tau_{\text{cav}}^{\text{rad}} \).

In the excitation terms of the rate equations, \( P_{\text{src}} \) is the source power, \( f_p \) is the excitation frequency, and \( \eta^{S_0 S_1} \) is the fraction of the pump emission absorbed in the \( S_0 S_1 \) states of the organic medium. These pump terms excite molecules from the \( S_0, S_1 \) and \( T_1 \) states to their respective higher energy levels separated by the photon energy \( h \nu_p \). In a single-pass absorption of the pump light over a distance \( l \), \( \eta^{S_0 S_1} \) is typically approximated by \( \sigma_{\text{pump}}^{S_0 S_1} \Gamma_p l N_{\text{den}}^{\text{tot}} \) where \( \sigma_{\text{pump}}^{S_0 S_1} \) is the experimentally measured absorption cross section of the bulk medium at the excitation wavelength and \( \Gamma_p \) is the confinement factor of the pumping mode in the organic medium. However, in cases where the pump light is resonantly coupled into the organic medium and the absorption of light occurs over multiple passes, the relationship between \( \eta \) and \( \sigma_{\text{pump}} \) is less trivial. For instance, in Sec. 5.2 (and Appendix 5.F) of Chapter 5 where we compared the experimentally measured threshold and slope efficiency values to numerically predicted ones in a system consisting of R6G placed on top of a photonic crystal slab cavity, \( \eta^{S_0 S_1} \) is estimated by considering the total losses of the system (both absorption in the dye as well as radiation and scattering losses of the cavity) and applying the \( Q \)-matching arguments [104]. We note that although our numerical simulations of the
full model considered pumping transitions in $S_0S_1$, $S_1S_2$ and $T_1T_2$, only the excitation of
the molecules from $S_0$ to $S_1$ is significant because its absorption cross section is an order
of magnitude larger than those in $S_1S_2$ and $T_1T_2$. Moreover, the population of the ground
singlet state is also larger than those in $S_1$ and $T_1$, which further diminishes the importance
of the pumping transitions in both $S_1S_2$ and $T_1T_2$ when operating at power levels near
the lasing threshold. Thus, in deriving the analytic solutions in Sec. 6.2 of the main text,
only the pumping transition from $S_0$ to $S_1$ is considered. Apart from the pump-induced
excitations of the molecules, the emitted lasing light is also re-absorbed by the dye to excite
molecules from the $S_0$, $S_1$ and $T_1$ states to their respective higher energy levels separated
by the photon energy $h\nu$. These self-absorption transitions are represented in the rate
equations using the cross sections $\sigma_{abs}$.

Next, we explain how the cavity enhancement effects are introduced into the stimu-
lated and spontaneous emission terms of the rate equations. We begin by relating the
experimentally measured stimulated emission cross sections $\sigma_{se}$ to the spontaneous emis-
sion coupling factor $\beta$. From Ref. [129], $\sigma_{se} = \lambda_4^2 g(\lambda_i) / 8\pi n_{eff}^2 \tau_{spont}$ in bulk organic media
and from Ref. [73,132,133], $\beta = \lambda_4^2 g(\lambda_i) / 8\pi n_{eff}^3 V$ in bulk systems (i.e. large resonators).
In both relationships, $g(\lambda_i)$ is the photoluminescence spectrum normalized to the fluorescence
quantum yield $q_{dye}$ [129], $n_{eff}$ is the effective index of the lasing mode, and $c$ is the speed of
light. $\tau_{spont}$ is the spontaneous emission lifetime in $N_2$ and is related to the non-radiative
lifetime $\tau_{21}$ by $q_{dye}$, where $\tau_{spont} = \tau_{21}(1 - q_{dye}) / q_{dye}$. The energy separation between $N_2$
and $N_1$ is $h\nu$. From the expressions of $\sigma_{se}$ and $\beta$ for bulk systems above, we find that
$\alpha \sigma_{se} / n_{eff} V = \beta / \tau_{spont}$. This relationship is used to replace the typical stimulated emission
terms involving $\sigma_{se}$ with $\beta \Gamma_s \phi_s (N_2 - N_1) / \tau_{spont}$ in our rate equations. Such formulation
of the stimulated emission term is consistent with those presented in Ref. [134]. As in all
optically induced transitions including the absorption of the pump and emitted light, the
stimulated emission term is directly proportional to the field intensity (through $\phi_s$ or $P_{sec}$)
and the population difference between the two levels connecting these transitions. The cav-
ity enhancement effects are then included in our model by replacing $\beta / \tau_{spont}$ above with the
enhanced spontaneous emission rate defined in Eq. (6.1). We note that even though $F_p$ and
$\beta$ appear as two independent quantities in Eq. (6.1), the value of $\beta$ will become dependent
on $F_p$ when its magnitude becomes high [131].

In the rate equations, $\tau_{lsc}$ is the intersystem-crossing lifetime of the molecules from $S_1$
to $T_1$, while $\tau_1$ is the triplet decay lifetime (both radiative and non-radiative) of molecules from $T_1$ to $S_0$. Phosphorescence arising from the decay of triplet molecules in $T_1$ is usually overshadowed by fluorescence in $S_0S_1$ (i.e. $\tau_{\text{spont}} \ll \tau_1$) and is not considered in our model. The transitions of molecules between the singlet and triplet states are energetically allowed but spin-forbidden, and so, they occur at a slower rate ($> 10^{-7}\,\text{s}$) than the singlet-singlet transitions ($< 10^{-8}\,\text{s}$). This allows the triplet influences in an organic laser system to be minimized by pumping with a pulse width shorter than $\tau_{\text{isc}}$, or by circulating the organic dye solution through the cavity at a rate faster than $1/\tau_{\text{isc}}$. It is critical to avoid a build up of the triplet population which tend to degrade the lasing performances by increasing the threshold and bleaching rate, while simultaneously decreasing the slope efficiency (see Sec. 6.2 of the main text).

Photobleaching of the organic medium is modeled through the irreversible loss of molecules in the triplet state, which is typically regarded as unstable due to its high chemical reactivity in an oxygen environment [124, 125]. $\tau_{\text{bleach}}$ is the photobleaching lifetime in $N_4$. It can be markedly different depending if the medium is exposed to an oxygen-rich or oxygen-deficient surroundings, or when different solvents are used with the dyes. It is sometimes also modeled to occur in the excited singlet state $S_1$ [126]. In this work, we consider the conventional approach of modeling the loss of molecules in $T_1$ but note that the model can be extended to also include the photobleaching process in $S_1$. 

104
Table 6.1 provides the parameters used in the steady-state calculations of Sec. 6.3 based on solution of R6G gain medium [129, 135, 136] (laser wavelength $\lambda_l = 580$ nm, pump wavelength $\lambda_p = 532$ nm):

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Dye concentration</td>
<td>5 mM</td>
</tr>
<tr>
<td>Fluorescence quantum yield $q_{dye}$</td>
<td>0.9</td>
</tr>
<tr>
<td>Spontaneous emission lifetime $\tau_{spont}$</td>
<td>5 ns</td>
</tr>
<tr>
<td>Intersystem crossing lifetime $\tau_{isc}$</td>
<td>100 ns</td>
</tr>
<tr>
<td>Triplet to singlet relaxation lifetime (with triplet quencher) $\tau_t$</td>
<td>100 ns</td>
</tr>
<tr>
<td>Triplet photo-bleaching lifetime $\tau_{bleach}$</td>
<td>8 ms</td>
</tr>
<tr>
<td>Non-radiative relaxation lifetime from level $i$ to $j$ $\tau_{ij}$</td>
<td>1 ps</td>
</tr>
<tr>
<td>Self-absorption cross-section in $S_0S_1$ $\sigma_{abs}^{S_0S_1}(\lambda_l)$</td>
<td>$10^{-19}$ cm$^2$</td>
</tr>
<tr>
<td>Self-absorption cross-section in $S_1S_2$ $\sigma_{abs}^{S_1S_2}(\lambda_l)$</td>
<td>$10^{-17}$ cm$^2$</td>
</tr>
<tr>
<td>Self-absorption cross-section in $T_1T_2$ $\sigma_{abs}^{T_1T_2}(\lambda_l)$</td>
<td>$10^{-17}$ cm$^2$</td>
</tr>
<tr>
<td>Thickness of gain region $\Gamma_s$</td>
<td>0.2</td>
</tr>
<tr>
<td>Spontaneous emission enhancement factor $F_p$</td>
<td>1</td>
</tr>
<tr>
<td>Spontaneous emission coupling factor $\beta$</td>
<td>$1 \times 10^{-4}$</td>
</tr>
<tr>
<td>Total quality factor of passive cavity $Q_{tot}$</td>
<td>$5 \times 10^4$</td>
</tr>
<tr>
<td>Quality factor of cavity due to radiation-loss $Q_{rad}$</td>
<td>$1 \times 10^5$</td>
</tr>
</tbody>
</table>

Table 6.1: Parameters of organic laser system based on R6G solution assumed for steady-state calculations.
Chapter 7

Spatio-temporal theory of lasing action in optically-pumped rotationally excited molecular gases

In this chapter, we investigate laser emission from optically-pumped rotationally excited molecular gases confined in a metallic cavity. To this end, we have developed a theoretical framework able to accurately describe, both in the spatial and temporal domains, the molecular collisional and diffusion processes characterizing the operation of this class of lasers. The effect on the main lasing features of the spatial variation of the electric field intensity and the ohmic losses associated to each cavity mode are also included in our analysis. Our simulations show that, for the exemplary case of methyl fluoride gas confined in a cylindrical copper cavity, the region of maximum population inversion is located near the cavity walls. Based on this fact, our calculations show that the lowest lasing threshold intensity corresponds to the cavity mode that, while maximizing the spatial overlap between the corresponding population inversion and electric-field intensity distributions, simultaneously minimizes the absorption losses occurring at the cavity walls. The dependence of the lasing threshold intensity on both the gas pressure and the cavity radius is also analyzed and compared with experiment. We find that as the cavity size is varied, the interplay between the overall gain of the system and the corresponding ohmic losses allows for the existence of an
optimal cavity radius which minimizes the intensity threshold for a large range of gas pressures. The theoretical analysis presented in this work expands the current understanding of lasing action in optically-pumped far-infrared lasers and, thus, could contribute to the development of a new class of compact far-infrared and terahertz sources able to operate efficiently at room temperature.

### 7.1 Introduction

It has been forty years since the pioneering work of Chang and Bridges [137] demonstrated generation of submillimeter radiation via stimulated emission from optically-pumped methyl fluoride (CH₃F) gas. Since then, a rich spectrum of laser lines through the 50 μm – 2 mm wavelength region have been obtained from a variety of different molecular gases [47–55]. These developments have made this class of lasers—usually referred to as optically-pumped far infrared (OPFIR) lasers—a central resource for many scientific and technological applications. The physical mechanism underlying the operation of conventional OPFIR lasers has also been extensively studied in past decades [47–60]. In essence, the gain in these systems derives from an optically-driven population inversion between two rotational states in an excited vibrational level of a gas molecule; the laser emission frequency is determined by the frequency of the transition between these two rotational states. The feedback mechanism enabling laser action is often provided by designing the cavity, in which the molecular gas is confined, to act as a one-dimensional Fabry-Perot resonator for the emitted laser frequency.

However, although research on conventional OPFIR lasers is generally considered as a mature field, some central challenges remain for the ultimate application of this technology, such as low efficiency, poor tunability, or limited capability of integration into a compact platform. More than two decades ago it was shown [51] that some of the aforementioned drawbacks can be overcome by considering the active role that high-energy vibrational states play in the laser operation (previous approaches assumed that the populations of those vibrational states were negligible). These findings demonstrated the feasibility of developing OPFIR lasers able to operate at large pressures (larger than the pressure cut-off determined by the so-called vibrational bottleneck [51,52]) and featuring volumes up to four orders of magnitude smaller than those characterizing typical OPFIR lasers. The advent of
quantum cascade lasers to replace CO$_2$ pump lasers may combine to produce truly compact submillimeter-wave laser sources in the near future.

Surprisingly, despite their great promise, up to date there has been limited interest in further exploring the potential of these discoveries [51] for the development of a novel class of compact far-infrared and terahertz (THz) sources able to operate efficiently at room temperature. In order to do that, it is critical to develop a complete theoretical understanding of the physical mechanisms underlying lasing action in the considered systems. The purpose of this work is to present such a theoretical analysis. Specifically, we introduce a theoretical framework which generalizes previous works in this area to allow for a realistic description of the spatio-temporal dynamics characterizing the molecular collisional and diffusion processes occurring in the considered OPFIR lasers. Importantly, the effect on the main laser features of both the electric-field (E-field) intensity distribution inside the metallic cell and the ohmic losses associated with the penetration of the E-field inside the metallic walls are also included in our theoretical approach. As we show in this work, both effects are particularly important for a realistic description of the operation of compact OPFIR lasers based on a metallic cavity which waveguides the emitted laser frequency.

Using this theoretical framework, we perform a detailed numerical analysis of laser emission from optically pumped methyl fluoride (CH$_3$F) gas confined in a cylindrical copper cavity, and find that the model compares well against experimental results. Our calculations show that, for this exemplary system, the region of maximum population inversion concentrates near the cavity walls. The outcome of these calculations also implies that, as a consequence of this population inversion distribution, the lowest threshold intensity corresponds to the cavity mode whose E-field intensity profile presents an optimal spatial overlap with the population inversion distribution, while at the same time, the ohmic losses associated to that cavity mode are minimized. In the case of the considered cylindrical cavity both features are satisfied by the TE$_{01}$ mode. We also analyze how the threshold intensity depends on the gas pressure and the cavity radius. The results from these simulations are explained in terms of the corresponding variation of the overall gain of the system and the ohmic losses associated with the lowest threshold mode. Remarkably, we predict that as the cavity size is varied, the interplay between both factors yields an optimal value of the cavity radius for which the threshold intensity is minimized for a large range of gas pressures.
The chapter is organized as follows. Section 7.2 discusses the theoretical model used throughout this chapter. Section 7.3 presents a detailed analysis of lasing action in the considered molecular gas lasers, including comparisons to experimentally measured results. Finally, in Sec. 7.4 we provide a set of conclusions of this work.

7.2 Theoretical framework

We start by introducing the theoretical model used to describe the molecular dynamics of the considered optically-pumped polyatomic molecular gases. Fig. 7-1 illustrates the energy level diagram and the different energy transfer mechanisms that form the basic structure of our model. In the following, we describe each of the processes sketched in Fig. 7-1 and show how to quantify them using experimentally measured parameters. Before starting this analysis, we point out that the theoretical approach presented here is an extension of previous studies on methyl fluoride CH₃F [51,52] in order to account for the spatial variations and diffusion of the collisional processes. We also point out that although for demonstration the model is applied to the case of methyl-halide gases, our theoretical analysis can be easily generalized to most OPFIR systems, both pulsed and CW, and operating over a wide range of experimental conditions utilizing an appropriate gas that satisfied the desired pumping and lasing transition frequencies [60].

7.2.1 Vibrational states and thermal pools

The experimentally-motivated thermal pool concept has been previously introduced in OPFIR laser models [48,52,53,60]. In the case of molecules, a thermal pool is observed to be a collection of rotational states that are in thermal equilibrium, and thus, are populated according to a Boltzmann distribution at the operating temperature. Such equilibrium may be attained, for instance, due to the fast relaxation rates among the levels. However, the total population of these pools varies through the flow of energy governed by the collisional and diffusive processes shown in Fig. 7-1. This fortuitous consolidation of numerous rotational levels into a small number of pools makes numerical simulation tractable, and more importantly, makes rotational energy transfer studies possible in the first place. In the particular case of methyl halides, there exist two thermal pools for each vibrational state, corresponding to the two symmetry types (A– and E–type), but without loss of generality
Figure 7-1: Schematic diagram of the general model used to describe the dynamics of an OPFIR molecular gas. Details on each process labeled are described in the text.

they will be combined here to describe CW OPFIR laser operation at low pressure [53]. In addition, all other higher vibrational states that are not connected by the pump are also modeled as pools. This inclusion is important at high pressure and pump intensity since it provides an additional channel through which molecules from the excited vibrational state may be transferred out, and hence, reduces the absorption on the corresponding FIR lasing transition (represented as the pink arrow in the excited state of Fig. 7-1).

At the relatively low pressure regime considered in this work, molecules are returned to the ground thermal pool from the pools of the excited and higher excited states, shown as green arrows in Fig. 7-1, primarily by colliding with the walls of the cavity in which the molecular gas is confined. Unlike molecule-molecule de-excitation processes, these involve diffusion through the gas towards the wall where their rates of occurrences decrease with pressure, and can be defined as \( k_{\text{wall}} = \nu \lambda_{\text{MFP}} / R^2 \) [138]. \( \lambda_{\text{MFP}} \) is the pressure-dependent mean free path and \( \nu \) is the average relative velocity between molecules. At very low pressure (or equivalently, within \( \lambda_{\text{MFP}} \) from the cavity walls), the transport mechanism changes from diffusion to ballistic and the rate becomes pressure-independent so that \( k_{\text{direct}} = \nu_{\text{abs}} / R \), with \( \nu_{\text{abs}} \) defined to be the average absolute velocity of a molecule. It is clear from these expressions that the wall rates are very different across the cavity, and we explicitly account...
for this spatial variation in our numerical simulations. On the other hand, the vibrational relaxation between the excited and higher excited pools can be dominated by molecule-molecule collisions, in which case, the transition rates between them are $k_{exc} = N_{tot} \sigma_{exc}$.

Here, $N_{tot}$ is the total density of molecules and $\sigma_{exc}$ is the collision cross section measured from experiments.

7.2.2 Non-thermal rotational levels

Non-equilibrium among rotational and vibrational states in the model is solely introduced by the pump (i.e. the blue arrow in Fig. 7-1), and the resulting nonthermal distribution of population in some of the rotational levels can no longer be considered part of the thermal pool but must be modeled individually. These nonthermal states, illustrated in Fig. 7-1 as level $J_1$, $J_2$...etc. in each vibrational state, only exist in states that are of the same symmetry type and vibrational state as those connected by the pump. The total population of any state $J_i$ (i.e. $N_{tot,J_i}$) is simply the sum of its nonthermal and thermal parts, $N_{tot,J_i} = N_{J_i} + f_{J_i}N_{pool}$, where $f_{J_i}$ is the fraction of the total pool population $N_{pool}$ in state $J_i$, and $N_{J_i}$ is the amount the state population differs from rotational thermal equilibrium because of the pump. Note that $N_{J_i}$ can be positive (excited vibrational state) or negative (ground vibrational state).

In general, the pump will create significant non-equilibrium not only in the states that it directly connects, but also in those adjacent to it. To account for this, our numerical model allows one to include as many rotational states as are necessary to model such a pump-induced non-equilibrium accurately. Generally, the number of $J$ states to include depends on the ratio of the dipole-dipole rate to the K-swap rate (see end of paragraph for the description of these two processes) — the slower the K-swap rate, the more $J$ levels one needs to consider. As already pointed out, the total population of any nonthermal state in this model is the combined population from both the nonthermal and thermal contributions, but since the thermal pools are in rotational equilibrium, the rotational relaxations, shown as black arrows in Fig. 7-1, are only modeled for the nonthermal population. In methyl fluoride, the rotational nonthermal processes are primarily dipole-dipole collisions whose rates are computed from the dipole-dipole collision cross section $\sigma_{DD}$ measured from experiments [54]. Note that their allowed transitions are restricted to $\Delta J = \pm 1$ at the same $K = K_i$ as the pumping transition. Lastly, the K-swap process [53] (i.e. the red arrows in Fig. 7-1) allows
population to be transferred out of nonthermal states into the rest of the rotational levels within the same thermal pool, and its rate constant can again be measured experimentally. The K-swap collisional processes (which encompass the $\Delta K=3n$ process and the V-swap process [53, 60] when the $A$- and $E$-symmetry thermal pools are combined as they are here) convert the nonthermal molecules to thermal molecules but not vice versa, and thus, allowing the nonthermal states to return to zero when the pump is turned off.

All of the aforementioned energy transfer mechanisms except K-swap include reverse processes which account for the appropriate degeneracy of the upper and lower levels so that microscopic reversibility is satisfied. Moreover, spatial diffusion of the molecules within the cavity, though a relatively slow process, is also included in order to account for the molecular gas dynamics within the cavity.

### 7.2.3 Pump transition rate

The pump transition rate, assuming a cylindrical waveguide cavity, is determined from

$$R_p = \alpha P_{\text{pump}}/\pi R^2 N_{\text{tot}} f_{\text{flow}} h \nu_{\text{IR}},$$

where $P_{\text{pump}}$ is the pump power, $f_{\text{flow}} = f_{J_1}$ is the fraction of molecules in the lower ro-vibrational state $J_1$ (as depicted in Fig. 7-1), $\nu_{\text{IR}}$ is the pump infrared frequency, and $\alpha$, which takes into account both Doppler broadening and mismatch between pump and absorption lines, measures the gas' ability to absorb the infrared radiation. Hence, pump transition rate may be increased by reducing $R$, increasing $P_{\text{pump}}$, or decreasing $\nu_{\text{IR}}$. The last of these, however, is constrained by the need to match the frequency of the laser line with that of a ro-vibrational transition in the molecules, and in general, cannot be varied at will. Using such an implementation, our model accommodates both CW and pulsed pumping with arbitrary spatial profiles.

### 7.2.4 Semiclassical rate equations

We describe the various processes in the model via rate equations. The most general rate description of a rotational level would include rotational relaxation terms due to dipole-dipole collisions among the non-thermal $J$ levels, terms representing the pumping mechanism, K-swapping terms that allow flow of molecules out of the non-thermal levels (with equilibrating effects on non-thermal levels), and the stimulated emission terms at the FIR wavelength. Since we are interested in the steady state distribution of molecules across the cavity, spatial diffusion terms are also added to the rate equations describing each levels. Stimulated
and spontaneous transitions at the lasing wavelength can, however, be neglected in the rate
equations when operated near the threshold if the spontaneous emission lifetime, $\tau_{\text{spont}}$, is relatively longer than all other transition lifetimes in the considered system. In methyl
fluoride, for instance, $\tau_{\text{spont}}$ is on the order of $10^5$ or more. In contrast, the next longest
lifetime in the model occurs for the vibrational relaxation terms and yet, has lifetime on
the order of $1 \times 10^{-3}$ or less. The relative magnitude of these radiative terms involving
$\tau_{\text{spont}}$ in the rate equations are further diminished by the fact that only a fraction of these
transitions contributes to the desired laser mode, especially when the cavity is large. With
that, the rate equation describing the molecular density of $J_2$ level in the excited vibrational
state (i.e. $N_{J_2}^{\text{exc}}$) of Fig. 7-1 is given by

$$
\frac{\partial N_{J_2}^{\text{exc}}(r, t)}{\partial t} = - R_p(r, t)\Delta N_\text{pump}(r, t) - N_{J_2}^{\text{exc}}(r, t) \sum_{i=1}^{3} \frac{1}{\tau_{J_2, i}} - \sum_{j=1}^{2} \frac{N_j^{\text{exc}}(r, t)}{\tau_{j, J_2}}
+ D\nabla^2 N_{J_2}^{\text{exc}}(r, t)
$$

(7.1)

where $\Delta N_\text{pump}(r, t) = N_{\text{tot}, J_2}^{\text{exc}}(r, t) - g_{J_2}^{\text{exc}} N_{\text{tot}, J_1}^{\text{grd}}(r, t) / g_{J_1}^{\text{grd}}$ with $N_{\text{tot}, J_1}^{\text{grd}}$ being the total molecular density of the level $J_1$ in the ground vibrational state after accounting for the nonther-
mal density and the contribution from the corresponding fraction of the thermal pool, and
$g_j^{\text{grd}} = 2J + 1$ is the degeneracy of the ground state $J^th$ level. Similar definitions hold for
$N_{\text{tot}, J_2}^{\text{exc}}$ and $g_{J_2}^{\text{exc}}$. $\tau_{J_2, i}$ for $i = 1, 2, 3$ is the decay lifetime out of $N_{J_2}^{\text{exc}}$ to the two adjacent
rotational levels and also the thermal pool. $\tau_{j, J_2}$ for $j = 1, 2$ represents the decay into $N_{J_2}^{\text{exc}}$
from its two adjacent levels $N_{J_1}^{\text{exc}}$ and $N_{J_2}^{\text{exc}}$. Finally, $D$ is the diffusion constant given by
the product of the molecule velocity and mean free path. In the same way, the thermal
pool equation has terms from the K-swap and spatial diffusion processes, as well as vibra-
tional relaxations due to molecule-molecule collisions and diffusion to wall. The equation
describing the excited vibrational pool in Fig. 7-1 is then given by

$$
\frac{\partial N_{\text{pool}}(r, t)}{\partial t} = - N_{\text{pool}}^{\text{exc}}(r, t) \sum_{k=1}^{2} \frac{1}{\tau_{\text{exc}, k}(r)} + \sum_{n=1}^{2} \frac{N_n^{\text{pool}}(r, t)}{\tau_{n, \text{exc}}(r)} + \sum_{m=1}^{m_n} \frac{N_{J_2}^{\text{exc}}(r, t)}{\tau_{J_2 m}^{\text{swap}}}
+ D\nabla^2 N_{\text{pool}}^{\text{exc}}(r, t)
$$

(7.2)

The first two terms relate to the vibrational relaxations among the thermal pools, where
$\tau_{\text{exc}, k}(r)$ for $k = 1, 2$ is the decay lifetime from the excited thermal pool to the ground and

114
higher excited pools, while \( \tau_{n,\text{exc}}(r) \) is the reverse. As noted earlier, the values of these diffusive lifetimes depend on their distances from the cell wall. \( \tau_{\text{swap}}^m \) for \( m = 1, 2, \ldots, m_0 \) in the third term is the lifetime of the transition from each of the \( m_0 \) rotational levels to the excited pool due to the K-swap molecule-molecule collision process. As in Eq. (7.1), the last term represents spatial diffusion. Similar equations to Eq. (7.1) and Eq. (7.2) are used to represent every nonthermal level or thermal pool that contributes to the lasing action, which in turn depends on their degree of non-equilibrium caused by the pump (see Appendix 7.A). This way, the complete set of rate equations describing our laser system obeys the diffusion equation \( \partial N_{\text{tot}}(r,t)/\partial t = D \nabla^2 N_{\text{tot}}(r,t) \), and so, conserves the total molecular density.

In our simulations, we consider only the immediate nonthermal levels adjacent to the pumped levels and initialize the density of molecules in the thermal pools according to a Boltzmann-distributed partition based on their respective vibrational energies. The non-thermal \( J_i \) levels are set equal to zero before any pumping occurs. The set of equations is then evolved in time until steady state is reached, allowing the temporal development of the lasing action to be tracked.

Laser action in this class of OPFIR molecular gas lasers is obtained as follows: Pumping is achieved with a line-tunable laser (e.g. CO\(_2\) laser) which excites molecules from a specific rotational level in the ground vibrational state into a specific rotational level in an excited vibrational state. Because the excited vibrational level is comparatively empty, these photo-excited molecules create a population inversion between the pumped rotational state and the one immediately below it. This leads directly to stimulated emission between rotational states in the excited vibrational state. These photo-excited non-equilibrium molecules are subsequently rotationally and vibrationally relaxed to the ground state through collisions with other molecules and with the chamber walls, respectively.

### 7.2.5 Vibrational bottleneck

The population inversion responsible for lasing exists as long as the pump-induced difference in nonthermal population between the upper and lower state \( N_{J_i} - N_{J_i-1} \) is greater than the difference in the corresponding pool-contributed thermal populations \( (f_{J_i} - f_{J_i-1})N_{\text{pool}} \) for those states. As the pressure grows, the pool molecules have an increasingly difficult time reaching the walls and de-exciting; thus, the thermal contribution from the pool increasingly
quenches the non-thermal population inversion. Pool quenching creates the vibrational bottleneck that was once believed to limit OPFIR laser operation to low pressures regardless of pump intensity. However, it is now understood that as the excited vibrational state thermal pool fills, near resonant collisions among its constituents can excite molecules to even higher-lying vibrational levels, thereby providing an alternate relaxation pathway [51]. Therefore, the pressure at which vibrational bottleneck occurs for a given laser geometry depends on the pump intensity. By concentrating on low pressure operation in this manuscript, these additional pathways may be ignored to simplify the analysis without affecting the generality of the results.

7.3 OPFIR laser system: cylindrical waveguide resonator

Figure 7-2 (a) illustrates the setup of the OPFIR laser system considered in this section. A cylindrical waveguide resonator of length $L_{\text{cell}}$ and radius $R$ is filled with a suitable gas that lases at the desired THz frequency, while pump power at a much higher IR frequency enters the system from the front window. Depending on the reflectivities of the front and back windows at THz, lasing output power can then escape from the cavity via both channels. When molecules are photo-excited from the ground vibrational state to an excited state, two types of population inversion can be induced, and lasing arises from each. The first is the primary lasing transition already described between the pumped rotational state in the excited vibrational level and the rotational state below it. The second is the refilling transition between the IR pump depleted rotational state in the ground vibrational level and the rotational state above it. When the pump is eventually turned off, the K-swap process allows equilibrium conditions in the molecules to be restored. In the following illustration of this theoretical approach, without loss of generality, we present results only for the primary lasing transition.

In order to compare our numerical calculations with past experimental results, the cavity modeled in the following simulation has length $L_{\text{cell}} = 12 \text{ cm}$ and front window reflectivity $\Gamma_{\text{front}} = 0.96$. The back window is opaque (i.e. $\Gamma_{\text{back}} = 1$). Furthermore, $R$ is varied across a range of commercially available values from the smallest possible before modal cutoff at $0.08 \text{ cm}$ to $1 \text{ cm}$. The cavity is then filled with an isotopic isomer of methyl fluoride gas, $^{13}\text{CH}_3\text{F}$, where a total of six rotational levels, $J_{\nu_0} = 3, 4, 5$ in the ground vibrational state and
IR power in Front window transparent to IR pump

Figure 7-2: (a) The OPFIR laser system considered in our numerical modeling: A cylindrical waveguide resonator of length $L_{cell}$ and radius $R$ is filled with a suitable gas that lases at the desired THz frequency, while pump power at a much higher IR frequency enters the system from the front window. In our example, $^{13}$CH$_3$F is pumped with CO$_2$ laser at 31 THz to produce lasing at 0.245 THz. (b) Frequency dependence of the ohmic losses for the five lowest order modes (i.e. TE$_{11}$, TM$_{01}$, TE$_{21}$, TE$_{01}$, TM$_{11}$) and three higher order ones with low losses (i.e. TE$_{02}$, TE$_{12}$, TE$_{22}$). Plot is for a copper cavity with $R = 0.26$ cm (and assuming $L_{cell} \gg R$) so that several modes (not all shown in plot) have cut-offs below the THz lasing frequency. Despite this, only the lowest loss mode TE$_{01}$ exist in the cavity near the lasing threshold. Vertical green line shows the band of frequency at which our system operates. (c) Intensity profile of the three lowest loss modes supported by the $R = 0.26$ cm waveguide cavity.
\( J_{\nu_3} = 4, 5, 6 \) in the excited state, are included (see detailed rate equations in Appendix 7.A). Here, the K-swap processes occur at roughly 1-2 times the rates of the rotational transitions, and through this process, the lower and higher adjacent J levels can be modeled as part of their respective pool. For low pressure operation, we include the doubly degenerate \( \nu_6 \) state as our only higher excited vibrational state. As we shall see next, these relatively small number of rotational-vibrational levels are already sufficient to attain reasonable match between numerical predictions and experimental data. The system is uniformly pumped at room temperature \((T = 300 \, \text{K})\) with CW CO\(_2\) laser at 31 THz in which molecules are excited between \( J_{\nu_0} = 4 \) and \( J_{\nu_3} = 5 \) to produce primary lasing transition at 0.245 THz in \( J_{\nu_3} = 4 \rightarrow 5 \) of the \( \nu_3 \) vibrational state, with \( \tau_{\text{spont}} \) roughly chosen to be 15s [139]. It is also noted that the occupancy of \( \nu_3 \) is comparatively low \((f = e^{-E_v/kT} \approx 1/150)\), making the condition favorable for the creation of population inversion between \( J_{\nu_3} = 5 \) and the level immediately below. Lastly, \( K_1 \) in Fig. 7-1 equals 3 for \(^{13}\text{CH}_3\text{F}\) and we have approximated the K-swap process as the faster \( \Delta K = 3n \) process where the V-swap rate is slow and primarily equilibrates \( A \) and \( E \) states in the pool. Other molecules with a slower or absent \( \Delta K = 3n \) process will need to approximate the K-swap process by the V-swap process. The rate constants and other salient molecular parameters used are provided in Appendix 7.A.

Figure 7-2 (b) shows a plot of the ohmic losses in a \( R = 0.26 \, \text{cm} \) copper waveguide cavity as a function of frequency. This loss prediction is made assuming \( L_{\text{cell}} \gg R \) so that only the circular plane of the cavity matters and that the copper conductivity is high enough that the wall currents flow uniformly within a skin depth of the surface [140]. At the operating frequency, shown as a green vertical band in figure, the cavity supports multiple modes but only the five lowest order modes and three higher order ones that have low ohmic losses are included in the plot. The cut-off frequency of each of these modes can also be deduced from the figure. The lowest loss mode, \( \text{TE}_{01} \), will be the first to lase upon threshold and remains the only mode present for pump power near the threshold, despite the cavity’s ability to support multiple modes at THz. Alternatively, one can ensure single-mode operation even for large pump power by selecting the smallest possible cavity size with radius \( \sim \lambda_{\text{THz}}/2 \), where \( \lambda_{\text{THz}} \) is the wavelength of the THz lasing output. For our purpose, it suffices to simply operate near the threshold for single-mode operation since this also facilitates independent design optimizations of our system based on molecular gas physics and photonics considerations. Fig. 7-2 (c) depicts the intensity profiles of the three
Figure 7-3: Time dependent properties of laser system with CW pumping at $P_{\text{pump}} = 10$ W. (a) Time evolution of the inversion in $\nu_3$ state at 250 mTorr across the radial axis of the cavity until steady state behavior is observed. (b) Time evolution of the $\nu_3$ thermal pool population at 250 mTorr across the radial axis of the cavity until steady state behavior is observed. (c) Same as in (a) except operated at 350 mTorr (using the same magnitude range for the color bar). (d) Same as in (b) except operated at 350 mTorr (using the same magnitude range for the color bar).

lowest loss modes of the cavity. It may be noted that the magnitude of ohmic loss scales roughly with the fraction of modal intensity residing near the cell wall, which in turn relates to the amount of penetration, to within a skin depth, into the wall. At low enough pump rate, only losses of TE$_{01}$ mode (both ohmic losses and leakage through the end mirrors) can be compensated by the gain produced in $^{13}$CH$_3$F while the other modes remain suppressed. Thus, we shall consider TE$_{01}$ to be the lowest-threshold lasing mode.
7.3.1 Spatio-temporal analysis

Figure 7-3 plots typical spatio-temporal results of the numerical simulation. In particular, the evolution of the laser transition $\Delta N(\lambda_{\text{THz}})$ and the $\nu_3$ thermal pool to steady state are presented and compared at 250 and 350 mTorr, assuming $P_{\text{pump}}$ to be 10 watts in CW operation. A few initial observations are in order: (i) the radial spatial profile of $\Delta N(r)$ is the inverse of that for $\nu_3$ pool. This is the pool quenching that leads to the vibrational bottleneck mentioned earlier. Note the rapid removal of molecules from $\nu_3$ near the cell wall is critical to maintaining the inversion. (ii) The cell center takes a long time to attain steady state given that the wall rate is slow far away from the wall (i.e. molecules have to travel a long distance to reach the cell boundary). (iii) Similarly, a longer time is required to reach steady state at a higher pressure due to the decreased $k_{\text{wall}}$ near the cell center. In Fig. 7-3, steady state is attained after 100 $\mu$s at 250 mTorr while it takes 300 $\mu$s to do so at 350 mTorr. (iv) The magnitude of the variation across the radial direction of the cavity is greater at higher pressure, as will be explained shortly. While we mostly deal with CW systems in this chapter, the ability to track the full temporal development of the laser action allows pulsed systems to be studied as well.

7.3.2 Comparison to experiments

We next verify our numerical model against experimental results for a $R = 0.26$ cm copper cavity $^{13}$CH$_3$F laser. The experimental configuration was identical to that described in [51], with a 1 mm diameter output coupler hole in the front reflector through which the IR pump and THz laser power were transmitted [Fig. 7-2 (a)]. The slope efficiency was measured as the ratio of the THz laser power emitted to the corresponding IR pump intensity, and the threshold pump intensity was the extrapolation of this slope to zero THz power. Numerical predictions and experimental measurements of both the pump power and slope efficiency at threshold are compared at two pressures: 100 mTorr and 300 mTorr. Here, we adopt a semi-analytic approach for these comparisons, where knowledge of the losses in the OPFIR system and population inversions calculated with our model are used simultaneously. In the following, we first discuss the match for the threshold power intensity before addressing that of the slope efficiency.
Lasing thresholds

The blue lines in Fig. 7-4 (a) and (b) depict the unsaturated effective gain values of the system, as predicted by our model, for a range of pump intensity at 100 and 300 mTorr respectively. The gain for the \( J = 4 - 5 \) lasing transition in \( \nu_3 \) state of \( ^{13}\text{CH}_3\text{F} \) may be written as [73,78]

\[
\langle \gamma(\lambda_{\text{THz}}) \rangle = \frac{\lambda_{\text{THz}}^2}{4\pi^2 n^2 \tau_{\text{spont}} \Delta \nu(\lambda_{\text{THz}})}
\]

(7.3)

where \( \langle \Delta N \rangle \) is the pressure-dependent steady-state effective inversion defined as

\[
\langle \Delta N(t_\infty) \rangle = \frac{\int_{V_{\text{ACT}}} dr \left| E_0(r) \right|^2 \Delta N(r, t_\infty)}{\int_{V_{\text{ACT}}} dr \left| E_0(r) \right|^2}.
\]

(7.4)

\( E_0(r) \) is the normalized mode profile of the passive cavity (\( \int dr \varepsilon_0 \left| E_0(r) \right|^2 = 1 \)) and \( V_{\text{ACT}} \) being the volume of active region of the considered structure. In our case, \( V_{\text{ACT}} \) is the entire cavity (note that this implies that the gas's behavior is less relevant in regions where the modal intensity is small but plays an important role in regions of high field intensity). In the considered case, \( \Delta N(r) = N_{\text{tot}, J=5}(r) - g_{J=4}^J N_{\text{tot}, J=4}(r) / g_{J=5}^J \) with \( N_{\text{tot}, J}^{\nu_3} \) being the total molecular density of the upper \( (J = 5) \) and lower \( (J = 4) \) lasing transition level in \( \nu_3 \) state after accounting for the fractional contribution from the excited thermal pool, and \( g_{J}^{\nu} = 2J + 1 \) is the degeneracy of the excited state's \( J^{\text{th}} \) level. \( E_0(r) \) is the normalized field of the TE\(_{01}\) mode and \( n \approx 1 \) for the gas. The width of the transition line, \( \Delta \nu \), is broadened due to a combination of Doppler effects and molecular collisions and has a strong dependence on both pressure and transition frequency. The former is associated with the Gaussian lineshape while the latter, which leads to pressure broadening, is Lorentzian. Hence, \( \Delta \nu = (\Delta \nu_D^2 + \Delta \nu_P^2)^{1/2} \) where the two terms are the Doppler and pressure broadening widths respectively, and the overall lineshape function used is the one that corresponds to the larger broadening parameter of the two. It is noted that for the same half width, pressure-broadened lines are stronger far away from the line-center than the Doppler-broadened ones. The gain defined in Eq. (7.3) accounts for the number of modes only in the frequency interval \( \nu_{\text{THz}} \pm \Delta \nu/2 \) by assuming a large optical resonator whose dimension is several wavelengths so that its exact shape becomes less significant. We find that this applies in our case since the cylindrical copper waveguide is almost 100 \( \lambda_{\text{THz}} \) long and a few \( \lambda_{\text{THz}} \)s in diameter. Moreover, the inversion density, and hence gain, is not
saturated by stimulated emission for the entire range of input intensity plotted. As we are only interested in the threshold value in this part of the work, such approach remains valid.

The threshold for lasing may be approximated (neglecting spontaneous emission) to occur when the gain balances the total losses of the OPFIR system. Losses in our setup are assumed to be due to either ohmic losses from the $E$-field penetration into the copper wall or to energy leakage from the front mirror. For $L_{cell} \gg R$, we can treat the system as one-dimensional along the cylinder axis for loss analysis and quantify its coefficient as $2\alpha_{\text{ohmic}} - \ln(\Gamma_{\text{front}}/\Gamma_{\text{back}})/L_{cell}$, where $\alpha_{\text{ohmic}}$ is the amplitude loss coefficient shown in Fig. 7-2 (b). The magnitudes of these loss coefficients are indicated as horizontal red lines in Fig. 7-4 (a) and (b). As such, the input threshold intensity predicted from our model is determined at the intersection between the red and blue lines, which indicates the point at which losses are compensated by the gain at $\lambda_{\text{THz}}$ so that any further increase in pump intensity saturates the gain and is thereby channeled into coherent output power that escapes from the front window. From Fig. 7-4 (a) and (b), the numerical thresholds attained are 0.78 and 2.42 W/cm$^2$ for the laser operated at 100 mTorr and 300 mTorr, respectively.

To make realistic comparisons with experiment, the decreasing absorption of the IR pump beam as it propagates along the cavity has to be considered. The pressure-dependent IR absorption coefficient, $\alpha'$, for $^{13}$CH$_3$F is 71 m$^{-1}$Torr$^{-1}$ [59] so that the ability of the pump to excite the gas decreases with increasing pressure. The effect is to reduce the amount of IR power effectively pumping the gas and can be captured by defining an incident pump power $P_{\text{pump}}^{\text{inc}} = P_{\text{pump}}/\kappa$, where $\kappa$ is a pressure-dependent factor that accounts for the net fraction of incident IR power available for excitation in the $L_{cell}$ long cavity. This factor is simply given by $(1/L_{cell}) \times \int_0^{L_{cell}} e^{-\alpha' x} dx$. Hence, $\kappa = 0.67$ and 0.36 for our 100 mTorr and 300 mTorr setups resulting in actual pump thresholds ($P_{\text{pump}}^{\text{inc}}/\pi R^2$) of 1.2 and 6.7 W/cm$^2$, respectively. These results compare well with experimentally measured thresholds of 1.1 ± 0.4 and 5.5 ± 1.5 W/cm$^2$. Possible sources of discrepancy in the comparison include instabilities that make measuring the threshold difficult, deviation from spatially uniform pumping in the experiment, the one-dimensional treatment in balancing gain-loss, and the possibility that the emission was from the refilling transition in the $\nu_0$ state. Even so, the current treatment provides adequately accurate predictions of the lasing thresholds. More importantly, this comparison acts to verify our claim that the aforementioned model of the OPFIR is physically robust and rigorous, yet numerically tractable.
Figure 7-4: (a) Method of threshold predictions via gain-loss balancing used in plotting (c) for every pressure point. Here, we operate at 100 mTorr with $R = 0.26 \text{ cm}$. The blue line is the unsaturated effective gain value for a range of pump intensity predicted by the aforementioned model while the red line is the magnitude of the loss coefficient due to absorption by the metallic cavity and energy leakage from the front window. Intersection between the two lines corresponds to the lasing threshold. (b) Same as in (a) except that the pressure is at 300 mTorr. (c) Left axis depicts the relationship between the threshold intensity and pressure for a range of $R$ values from 0.08 cm to 1 cm as predicted from numerical model. The corresponding linewidth, $\Delta \nu$, of the lasing transition (over which it may be tuned) is also illustrated in the top axis. A general trend exists such that the lasing threshold increases with pressure. The right axis shows the mean free path, $\lambda_{\text{MFP}}$, within the gaseous system which at a fixed temperature, is inversely proportional to pressure. Inset is the ohmic loss [see Fig. 7-2 (b)] of the TE$_{01}$ mode at 0.245 THz, with the radii of interest marked as square markers.

Slope efficiencies

Next, we present a similar comparison for the slope efficiency at threshold using a semi-analytic approach. The set of rate equations in Eq. (7.1), with the inclusion of the induced lasing transition rate, are solved analytically at steady-state to obtain the following approximate relationship between output power $P_{\text{out}}$ and input power $P_{\text{in}}$ above threshold:

$$P_{\text{out}} = f_{\text{out}} P_{\text{sat}} \left( \frac{P_{\text{in}}}{P_{\text{th}}^{\text{in}}} - 1 \right)$$  \hspace{1cm} (7.5)
where \( f_{out} \) accounts for the fraction of emitted power that escapes via the front window, and \( P_{in}^{th} \) is the IR power at threshold. \( P_{sat} \) is the saturation power at THz in which the gain in the system is half its unsaturated value. In this particular system, \( P_{sat} = \hbar \omega_{THz} \int_{V_{ACT}} d\mathbf{r} \Delta N_{th}(\mathbf{r})/\tau_{eff} \) where \( \Delta N_{th} \) is the inversion at threshold and \( \tau_{eff} \) is an effective lifetime that accounts for the various rates between the rotational levels in the lasing \( \nu_3 \) state, and is found to be

\[
\tau_{eff} = \frac{1/\tau_{J_1}}{1/\tau_{J_1}^{\text{swap}} - 1/\tau'} - \frac{1/\tau_{J_2}}{1/\tau_{J_2}^{\text{swap}} - 1/\tau'} - \frac{1}{\tau_{J_3}}\left( \frac{1 - \tau_{J_3}^{\text{swap}}}{\tau_{J_3}^{\text{swap}} - 1/\tau_{J_3}} \right)
\]

The lifetimes in Eq. (7.6) describe the transition rates in the excited \( \nu_3 \) state where lasing occurs and are defined in accordance to their definitions in Fig. 7-1 and Eq. (7.1). With that, the slope \( dP_{out}/dP_{in} \) is \( f_{out}P_{sat}/P_{in}^{th} \). Using numerical results of \( \Delta N_{th} \) and \( P_{in}^{th} \) found from the full model and accounting for the pressure-dependent IR absorption discussed in the previous paragraph, we obtain a threshold slope efficiency of \( 24.5 \times 10^{-6} \) and \( 39.2 \times 10^{-6} \) at 100 mTorr and 300 mTorr, respectively. Note that the best slope efficiency for the considered \( ^{13}\text{CH}_3\text{F} \) laser is \( 8 \times 10^{-3} \), which is the Manley-Rowe limit when the quantum efficiency is 1. These calculated values compare reasonably well with experimental slope values of \( (44 \pm 22) \times 10^{-6} \) and \( (48 \pm 24) \times 10^{-6} \), for which the greatest source of uncertainty is the calibration of the Golay cell used to detect the THz radiation. Given this, these results further validate our model.

### 7.3.3 General analysis of laser performances

We extend the threshold determination method of Fig. 7-4 (a) and (b) to study how the threshold intensity behaves as both cell radius and pressure are varied in Fig. 7-4 (c). A general trend exists such that the lasing threshold increases with pressure for every radius plotted. This can primarily be attributed to the fact that the linewidth, \( \Delta \nu(\lambda_{THz}) \), broadens with increasing pressure so that \( \langle \gamma(\lambda_{THz}) \rangle \) reduces [see Eq. (7.3)]. Note that the variation of the linewidth with pressure is captured in the top axis of the figure. Since the total loss
of the system is pressure-independent, such gain reduction increases the threshold linearly with pressure for each value of $R$. Second order effects that further raise the threshold at high pressure originate from the vibrational bottleneck; a higher pressure impedes the molecular diffusion to the cell wall and the subsequent decrease in wall collision rates reduces the flow of molecules out of the excited thermal pool, leading to enhanced absorption and hence, a weaker inversion $\langle \Delta N \rangle$ and gain. Equivalently, the same effects on $\langle \Delta N \rangle$ can be understood by recognizing that $\lambda_{\text{MFP}}$ progressively makes up a smaller region near the cell wall as the pressure increases such that a greater proportion of the molecules are in the non-ballistic regime, which also leads to slower $k_{\text{wall}}$. Red dashed line plotted against the right axis depicts the pressure dependence of $\lambda_{\text{MFP}}$. Over the range of low pressures considered, the vibrational bottleneck is only observed for large radius cells (i.e. $R \geq 0.26 \text{ cm}$ in the current setup) while for smaller cavities, there is no vibrational bottleneck and the threshold dependence on pressure remains approximately linear.

From Fig. 7-4 (c), it can also be seen that the threshold intensity increases with radius for $R$ values larger than 0.26 cm but decreases with radius for $R$ values smaller than 0.26 cm. To understand why $R = 0.26 \text{ cm}$ is nearly optimal, the reader is referred to the inset that illustrates the reduction in ohmic losses of the $\text{TE}_{01}$ mode at 0.245 THz when $R$ grows from the waveguide cut-off value, with the radii of interest examined in the main figure marked as square markers. For small radius, the ohmic loss is high and hence, possesses great influence on the direction of threshold shift. In this regime, when the radius increases, the corresponding fall in ohmic loss has the tendency to lower the threshold. On the other hand, at larger $R$ values where the ohmic losses are low, the vibrational bottleneck increasingly raises or prevents threshold with increasing $R$ and pressure. However, at pressures less than 100 mTorr in large cells (i.e. $R \geq 0.26 \text{ cm}$), the pressure-dependent threshold lines in Fig. 7-4 (c) converge to the linear behavior observed for cells with $R \leq 0.26 \text{ cm}$. As we shall see next, this follows directly from the fact that $\lambda_{\text{MFP}}$ grows large at low pressures, so for a constant intensity and pump rate $R_{\text{pump}}$, the inversion $\langle \Delta N \rangle$ and gain exhibit little radial dependence. As a result, variation of $R$ has a much smaller effect on the wall collision rate, and correspondingly on $\langle \gamma \rangle$, at lower pressures. So, for cell radii where the ohmic losses are also low, the threshold remains almost unchanged as $R$ is varied.

To understand the spatial effects on laser performance more clearly, the radial dependence of the inversion and gain in a $R = 0.5 \text{ cm}$ cavity pumped by CW CO$_2$ laser at
Figure 7-5: (a) Radial spatial variation of the excited inversion in $\nu_3$ state of $^{13}$CH$_3$F for a range of operating pressure from 50 to 450 mTorr. 0 cm marks the center of the cylindrical cavity while 0.5 cm marks the copper wall. All four panels in this figure assumed CW pumping at $P_{\text{pump}} = 100$ W. Near the cell wall, the diffusion is ballistic allowing significant reduction of molecules from the $\nu_3$ pool, and hence, results in a higher population inversion. (b) Same as in (a) except that the transition width at 0.245 THz is also factored in to study the gain. (c) Gain dependence on pressure for a fixed set of pump parameters. From a molecular gas physics standpoint, results clearly indicate that small $R$ cavities are favored in terms of gain magnitude and pressure cut-off. (d) Data extracted from (c) where the left axis and right axis respectively depict the optimum operating pressure and the corresponding gain, as a function of cell radius $R$. Again, results here favored small sized cavity for high pressure operation.
\( P_{\text{pump}} = 100 \text{ W} \) are studied in Fig. 7-5. From the intensity-weighted averaging scheme used in Eq. (7.4), we have already noted that it is most desirable to match the peak of the cavity’s eigenmode intensity to regions where \( \Delta N(r) \) or \( \gamma(r) \) is maximum. Such consideration plays an important role in designing the cavity itself. Fig. 7-5 (a) illustrate the distribution of \( \Delta N(r) \) within the cavity for values of pressure ranging from 50 mTorr to 450 mTorr. In general, strong inversions are favored near the cell wall where the wall collision rates are the highest, and so, allow rapid depopulation of the excited and higher excited thermal pools to reduce pool quenching. This directly implies an advantage in exciting a lasing mode whose intensity peaks near the cell wall. However, such approach will also have to be weighted against the higher ohmic losses that arise from the field’s enhanced interaction with the cell, in a similar trade-off effects to what was already discussed in Fig. 7-4 (c). The details and treatment of the cavity design will be left for future work. We further note that the magnitude of radial variation in the cell increases with pressure. For instance, at 50 mTorr, a good proportion of the cavity is ballistic [see \( \lambda_{\text{MFP}} \) variation in Fig. 7-4 (c)] so that \( \Delta N \) is approximately uniform, i.e. \( \Delta N(r_{\text{cen}}) = 0.96 \Delta N(r_{\text{wall}}) \) where \( r_{\text{cen}} \) and \( r_{\text{wall}} \) indicate the position at the cell center and wall, respectively. On the other hand, at 450 mTorr, most of the molecules are in the non-ballistic regime where \( k_{\text{wall}} \propto 1/r^2 \) as opposed to \( k_{\text{direct}} \propto 1/r \). This leads to \( \Delta N(r_{\text{cen}}) \) to be only 14% of \( \Delta N(r_{\text{wall}}) \). Indeed, it was found that the maximum inversion for the set of parameters used in this example increases with pressure because of the corresponding increase in \( N_{\text{tot}} \). Fig. 7-5 (b) depicts a similar plot for the gain. The same characteristics are observed among the two except that \( \gamma(r) \) is also influenced by the laser transition linewidth which broadens as pressure increases. This decrease of gain with pressure is again consistent with the discussion presented for Fig. 7-4 (c), explaining why most OPFIR lasers use large diameter cavities operating at low pressure.

However, this design misses a critical point. Since \( R_p \) increases when the same pump power is more tightly confined, the gain tends to grow as cavity radii shrink until it eventually saturates (because \( N_{\text{tot}} \) is a conserved quantity). Fig. 7-5 (c) and (d) track the optimum radius size for lasing based on the numerical model developed. An optimum operating pressure exists for each radius where the gain is maximized. Beyond that, vibrational bottleneck begins to play a dominant role, explaining the fall of the gain as pressure rises. At high enough pressure, the lasing action will be quenched. For example, in Fig. 7-5 (c)
the cut-off pressure occurs at 380 mTorr for $R = 1 \, \text{cm}$. Because of the role of higher lying vibrational levels, the cutoff pressure is pump intensity dependent such that the greater the pumping intensity, the higher the cut-off pressure. Thus, the optimum pressure and gain results summarized in Fig. 7-5 (d) based on the molecular gas physics greatly favor small-sized cavities, especially if the effects of waveguide ohmic losses can be overcome.

7.4 Conclusion

We have presented a comprehensive theoretical analysis of lasing action in optically-pumped rotationally excited molecular gases confined in a metallic cavity. In order to do that, we have developed a theoretical framework that provides a realistic spatio-temporal description of the molecular de-excitation and diffusion processes occurring inside the cavity. Our approach also accounts for the influence on the main laser features of the electric-field intensity distribution and the ohmic losses associated to the different cavity modes. To illustrate this theoretical framework, we have analyzed in detail the case of lasing action from optically-pumped methyl fluoride gas confined in a cylindrical copper cavity. Our simulations have shown that for this system, the region of maximum population inversion is located near the cavity walls. The outcome of our calculations has also shown that the lowest lasing threshold mode corresponds to the cavity mode whose electric-field intensity profile presents an optimal spatial overlap with the corresponding population inversion distribution inside the cavity, and at the same time, minimizes the ohmic losses at the lasing frequency. Furthermore, we have predicted the existence of an optimal radius, determined by the interplay between the overall gain in the system and the corresponding ohmic losses, which minimizes the lasing threshold intensity for a large interval of gas pressures. We believe the results reported in this work could contribute to the development of a novel class of room-temperature compact sources of far-infrared and THz radiation.

7.A Appendix: Rate equations and rate constants of $^{13}\text{CH}_3\text{F}$ gas lasers

In this appendix, we first present the set of rate equations used to model the $^{13}\text{CH}_3\text{F}$ gas lasers. Fig. 7-6 and Eq. (7.7) to (7.15) refer to a specific example of a methyl fluoride gas
discussed in Sec. 7.3, adopted from the general model presented in Fig. 7-1, Eq. (7.1) and Eq. (7.2) of Sec. 7.2.4. We have labeled the rotational levels from $N_1$ to $N_6$, and the thermal pools as $N_A$, $N_B$, and $N_C$. Hence, $\tau_{12}$, $\tau_{23}$, $\tau_{45}$, $\tau_{56}$, and their reverse processes represent rotational transitions due to dipole-dipole collisions, $\tau_{1A}$, $\tau_{4B}$, and similar processes represent K-swap transition lifetimes from the rotational levels (i.e. $N_1$ and $N_4$) into their respective thermal pools (i.e. $N_A$ and $N_B$), and $\tau_{AB}$, $\tau_{AC}$, and their reverse processes are vibrational transitions which arise due to collisions with the cell wall. Readers should note that all notations in the main body of the chapter are kept consistent with those used in Fig. 7-1, Eq. (7.1) and Eq. (7.2).

$$\frac{\partial N_1(r, t)}{\partial t} = -N_1(r, t) \left( \frac{1}{\tau_{12}} + \frac{1}{\tau_{1A}} \right) + \frac{N_2(r, t)}{\tau_{21}} + D\nabla^2 N_1(r, t)$$  \hspace{1cm} (7.7)

$$\frac{\partial N_2(r, t)}{\partial t} = -N_2(r, t) \left( \frac{1}{\tau_{21}} + \frac{1}{\tau_{23}} + \frac{1}{\tau_{2A}} \right) + \frac{N_1(r, t)}{\tau_{12}} + \frac{N_3(r, t)}{\tau_{32}} + R_p(r, t) \Delta N_{\text{pump}}(r, t) + D\nabla^2 N_2(r, t)$$  \hspace{1cm} (7.8)

$$\frac{\partial N_3(r, t)}{\partial t} = -N_3(r, t) \left( \frac{1}{\tau_{32}} + \frac{1}{\tau_{3A}} \right) + \frac{N_2(r, t)}{\tau_{23}} + D\nabla^2 N_3(r, t)$$  \hspace{1cm} (7.9)
\[
\frac{\partial N_4(r,t)}{\partial t} = - N_4(r,t) \left( \frac{1}{\tau_{45}} + \frac{1}{\tau_{4B}} \right) + \frac{N_5(r,t)}{\tau_{54}} + D\nabla^2 N_4(r,t) \\
\frac{\partial N_5(r,t)}{\partial t} = - N_5(r,t) \left( \frac{1}{\tau_{54}} + \frac{1}{\tau_{56}} + \frac{1}{\tau_{5B}} \right) + \frac{N_4(r,t)}{\tau_{45}} + \frac{N_6(r,t)}{\tau_{65}} \\
- R_p(r,t) \Delta N_{\text{pump}}(r,t) + D\nabla^2 N_5(r,t) \\
\frac{\partial N_6(r,t)}{\partial t} = - N_6(r,t) \left( \frac{1}{\tau_{65}} + \frac{1}{\tau_{6B}} \right) + \frac{N_5(r,t)}{\tau_{56}} + D\nabla^2 N_6(r,t) \\
\frac{\partial N_A(r,t)}{\partial t} = - N_A(r,t) \left( \frac{1}{\tau_{AB}(r)} + \frac{1}{\tau_{AC}(r)} \right) + \frac{N_B(r,t)}{\tau_{BA}(r)} + \frac{N_C(r,t)}{\tau_{CA}(r)} \\
+ \frac{N_1(r,t)}{\tau_{1A}} + \frac{N_2(r,t)}{\tau_{2A}} + \frac{N_3(r,t)}{\tau_{3A}} + D\nabla^2 N_A(r,t) \\
\frac{\partial N_B(r,t)}{\partial t} = - N_B(r,t) \left( \frac{1}{\tau_{BA}(r)} + \frac{1}{\tau_{BC}} \right) + \frac{N_A(r,t)}{\tau_{AB}(r)} + \frac{N_C(r,t)}{\tau_{CB}} \\
+ \frac{N_4(r,t)}{\tau_{4B}} + \frac{N_5(r,t)}{\tau_{5B}} + \frac{N_6(r,t)}{\tau_{6B}} + D\nabla^2 N_B(r,t) \\
\frac{\partial N_C(r,t)}{\partial t} = - N_C(r,t) \left( \frac{1}{\tau_{CA}(r)} + \frac{1}{\tau_{CB}} \right) + \frac{N_A(r,t)}{\tau_{AC}(r)} + \frac{N_B(r,t)}{\tau_{BC}} \\
+ D\nabla^2 N_C(r,t) 
\]

where \( \Delta N_{\text{pump}}(r,t) = N_5^{\text{tot}}(r,t) - g_5 N_2^{\text{tot}}(r,t)/g_2 \) with \( N_2^{\text{tot}}(r,t) \) being the total molecular density of \( J = 4 \) nonthermal level in \( \nu_0 \) state after accounting for the fractional contribution from thermal pool \( A \), and \( g_2 = 9 \) is its degeneracy. Similar definitions hold for \( N_5^{\text{tot}} \) and \( g_5 = 11 \).

Table 7.1 provides the rate constants and relevant molecular parameters used in our calculations \([53, 59, 60]\).

<table>
<thead>
<tr>
<th>Mass (AMU)</th>
<th>35</th>
</tr>
</thead>
<tbody>
<tr>
<td>Gas kinetic collision cross section</td>
<td>44</td>
</tr>
<tr>
<td>( \nu_3 \to \nu_6 ) cross section at 300 K, ( \sigma_{\text{exc}} )</td>
<td>2.65</td>
</tr>
<tr>
<td>Dipole-dipole cross section at 300 K, ( \sigma_{DD} )</td>
<td>320</td>
</tr>
<tr>
<td>K-swap cross section at 300 K, ( \sigma_{K\text{-swap}} )</td>
<td>137</td>
</tr>
</tbody>
</table>

Table 7.1: Transitional cross sections (in \( \text{A}^2 \)) of \(^{13}\text{CH}_3\text{F}\).
Chapter 8

Conclusions

In this thesis, we have investigated the interaction of light with a variety of material systems, which will in turn guide the designs of subwavelength-scale lasers: applied an exemplary four-level gain model to the photonic crystal slabs, experimentally realized lasing in such structures using diluted solution of R6G organic molecules as the gain medium, proposed a larger-area single-mode PCSEL, developed a comprehensive theoretical framework to describe the dynamics behavior of organic nanostructured lasers, and generalized previous works on OPFIR gas lasers to allow for a realistic description of the spatio-temporal dynamics characterizing their molecular collisional and diffusion processes. These are all very exciting topics with numerous potential applications; and, looking forward, there are still interesting questions to be explored.

Today, commercially available single-mode sources are the edge-emitting DFBs and ridge-waveguide lasers, as well as the surface-emitting VCSELs. While the former suffers from high divergence of the output beam, the power outputs of the VCSELs are limited by their small cavity sizes. The surface-emitting PCSELs can potentially overcome both disadvantages; broad-area and single-mode PCSELs with good beam qualities (having only 1° divergence of their far-fields) have already been demonstrated [12,81]. However, unlike the DFBs and VCSELs, the PCSEL utilizes a two-dimensional feedback structure instead of one-dimensional, and thus, its realization is considered to be relatively difficult. With the rapid advance of nano-fabrication techniques in the past decade, and the constant demand for single-mode on-chip lasers having good beam qualities, we expect the PCSELs to become increasingly popular for numerous applications in the near future. Importantly,
our proposal in Chapter 4 to achieve PCSELs of higher output powers over typical ones will further expand their range of applications. In particular, lasing at an accidental Dirac-point in the Brillouin zone center has never been demonstrated, and it will be interesting – especially from the technological applications point of view – to experimentally measure the band structure (frequency-locking region) and to realize single-mode lasing at such a point using similar procedures described in Chapter 5.

Next, we note that optically pumped organic lasers have been widely demonstrated [113,114]. However, lasing in these systems is only possible with high peak power excitation sources of short pulses; no continuous-wave operated organic lasers has been demonstrated without liquid dye circulation. In Chapter 6, we explored the feasibility of achieving low-threshold lasing in a continuous optically pumped monolithic organic laser and theoretically illustrate systems where lasing action can be sustained over a time scale longer than all decay lifetimes in the organic medium, before photobleaching occurs. It would be interesting to realize lasing and measure the thresholds in suitably designed systems for such a purpose. While dye degradation that leads to photobleaching is inevitable, the photobleaching lifetime can be markedly different depending on the surroundings (i.e. oxygen content) of the medium, or when different solvents are used with the dyes. Further lengthening of the lasing duration can be achieved by controlling the optical and chemical properties of the laser systems, as described in Eq. (6.5).

Lastly, having generalized previous works of the OPFIR gas lasers to allow for a realistic spatio-temporal description of the molecular collisional and diffusion processes in Chapter 7, we now have a better understanding on how to overcome the vibrational bottleneck that has been limiting the laser performances. One of the simplest approaches is to maximize the resonators surface areas available for molecular collisions in order to promote the de-excitation of the excited vibrational states. As an avenue of further research, we plan to consider novel optical designs of the feedback structures based on these findings to eventually develop a new class of compact powerful (> 100 mW) far-infrared and terahertz sources able to operate efficiently at room temperature.
Bibliography


