

Phase Transitions of Exciton-Polaritons in Grating-Based Microcavities

by

Jiaqi Hu

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Doctoral Committee:

Professor Hui Deng, Chair
Professor Steven Thomas Cundiff
Professor L. Jay Guo
Professor Mackillo Kira
Professor Alex Kuzmich

Jiaqi Hu

hujiayi@umich.edu

ORCID iD: 0000-0002-1113-8860

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TABLE OF CONTENTS

ACKNOWLEDGMENTS	ii
LIST OF FIGURES	v
LIST OF ACRONYMS	vii
ABSTRACT	ix

CHAPTER

1 Introduction	1
1.1 Exciton-Polaritons in Semiconductors	1
1.2 Polariton Lasing and Condensation	6
1.3 Modifying the Properties of Polaritons by Engineering the Cavity	11
1.4 Subwavelength Grating Reflectors	13
1.5 Overview of This Dissertation	19
2 Polariton Laser in the High Density Regime	22
2.1 Experimental Approaches to High Density Polariton Systems	22
2.2 Sample Structure and Properties	24
2.3 Measurement Setup	26
2.4 Photoluminescence Spectra	29
2.5 Reflection Spectra	33
2.6 Density Estimation	41
2.7 Comparison with Theoretical Model	44
2.8 Detuning Dependence	48
2.9 Comparison with Planar DBR Cavity	50
2.10 Summary and Future Work	60
3 Tuning Polariton Properties with Subwavelength Grating	63
3.1 Linewidth Tuning and Non-Hermitian Physics	63
3.2 Monolithic Grating and Fabrication	66
3.3 Tuning Mechanism	69
3.4 Experimental Demonstration of Linewidth Control	72
3.5 Coupled Devices and Future Work	77

4 Polariton Cavity with a Fano Interference-Based Reflector	81
4.1 Fano Interference-Based Reflector and Cavity	81
4.2 Application to the Exciton-Polariton System	85
4.3 Sample Characterization	88
4.4 Future Work	92
5 Conclusion	94
BIBLIOGRAPHY	97

LIST OF FIGURES

FIGURE

1.1	Structure and properties of a typical exciton-polariton microcavity.	4
1.2	Polariton dispersion model.	5
1.3	An example of phase diagram of the exciton-polariton system.	6
1.4	Schematics of different phases in the exciton-polariton system.	10
1.5	Structure and resonance modes of a high-contrast SWG.	15
1.6	Guided-mode resonances of an SWG.	17
1.7	Examples of an SWG-DBR hybrid cavity.	18
2.1	Simulated reflectance of the grating-based linear polarization-selective mirror.	25
2.2	Polarization-selective grating-based microcavity.	26
2.3	Experiment setup for PL and reflection spectra measurement.	28
2.4	Emission intensities in the unetched region of the sample with different polarization configurations.	30
2.5	Emission properties of a polariton laser and a photon laser.	32
2.6	Real-space TE emission spectra of the two types of lasers.	34
2.7	Fourier-space TM emission spectra.	35
2.8	Measured reflection spectra and optical gain of a polariton laser.	36
2.9	TE reflection and emission spectra of a polariton laser.	38
2.10	TM-polarized reflection spectra at low pump power.	40
2.11	Examples of exciton energy estimation from TM reflection spectra.	40
2.12	Spatial profile of TM-polarized emission.	42
2.13	Measured and calculated gain and energetic positions of a polariton laser.	46
2.14	Calculated carrier distributions and wave functions.	47
2.15	Emission features of devices with different parameters and estimated threshold density.	49
2.16	Reflectance and gain of devices with different parameters.	51
2.17	Structure and reflection spectrum of the DBR microcavity.	52
2.18	Estimating the coupling strength and detuning of the DBR sample.	54
2.19	PL spectra under CW pump with a 3 μm spot.	55
2.20	Image of the ring-shaped pump on the sample.	56
2.21	PL spectra under CW pump showing two types of lasing transition.	57
2.22	Summary of the detuning dependence of the PL spectra features.	59
2.23	The change in polariton effective mass with pump power.	61
3.1	Complex eigenvalues of the matrix in Eq. 3.1.	65
3.2	Mechanism of the high reflectivity of MHCG.	68

3.3	SEM images of MHCG.	70
3.4	Reflectivity and reflection phase of a grating-based mirror.	71
3.5	Fourier-space PL spectra of typical devices showing equal LP energy but different linewidths.	73
3.6	Measured and simulated lower polariton energy and linewidth at zero wave vector for two series of gratings.	74
3.7	A collection of measured PL energy and linewidth of LP modes at zero wave vector of gratings with various parameters on two different sample pieces.	76
3.8	Coupled grating-based polariton devices.	79
4.1	Schematic and reflection spectrum of a Fano interference-based reflector.	83
4.2	Microcavity using the Fano interference-based reflector.	87
4.3	Strong-coupling between exciton and photon with the Fano cavity.	88
4.4	Simulated and measured angle-resolved reflection spectra of the reflectors.	89
4.5	Open cavity experimental setup.	91
4.6	Differential reflection spectra of the Fano cavity.	92

LIST OF ACRONYMS

BCS	Bardeen–Cooper–Schrieffer
BEC	Bose–Einstein condensate
BKT	Berezinskii–Kosterlitz–Thouless
CW	continuous-wave
FP	Fabry–Pérot
FWHM	full width at half maximum
GMR	guided-mode resonance
HHX	heavy-hole exciton
LHX	light-hole exciton
LP	lower polariton
MHCG	monolithic high-contrast grating
MP	middle polariton
PL	photoluminescence
PT	parity-time
Q-factor	quality factor
QW	quantum well
RCWA	rigorous coupled-wave analysis
SEM	scanning electron microscope
SWG	subwavelength grating
TE	transverse-electric
TM	transverse-magnetic

TMDs transition metal dichalcogenides

UP upper polariton

VCSEL vertical-cavity surface-emitting laser

WGA waveguide array

ABSTRACT

Exciton-polaritons in semiconductor microcavities have been a platform to study macroscopic quantum phenomena. They exhibit phases like Bose–Einstein condensate (BEC) and conventional photon lasing. Due to their fermionic constituents, a Bardeen–Cooper–Schrieffer (BCS)-like phase has also been predicted at density levels in between that of a BEC and photon lasing. The pumping and decay of polaritons not only modify these phases but may lead to a range of non-equilibrium phenomena that are more accessible in the polariton system than in closed systems. Studies of the BCS-like phase and other phenomena in the polariton system require the capability to control the polariton properties. In this dissertation, experimental methods are developed to control the polariton system by engineering its photonic component using grating-based microcavities. The tuning of energy, decay rate, polarization, and mode profile of the photonic mode in compact devices is demonstrated and utilized to identify and characterize new types of phases of exciton-polaritons.

A high-contrast grating that has polarization-selective high reflectivity is used as a reflector for a GaAs/AlGaAs microcavity with GaAs quantum wells. In the linear polarization with high-reflectivity, spontaneous transitions of exciton-polaritons from the normal phase to phases with coherent light emission under non-resonant pumping are observed, while the response of the underlying excitons can be probed using the orthogonal linear polarization. The features of the polariton emission spectra resemble a polariton BEC or a photon laser as detuning changes, while the reflection spectra of the excitons reveal that the coherent emission is due to stimulated emission under population inversion in both cases, ruling out the possibility of a polariton BEC. The comparison with a theoretical model suggests the possibility of the polariton BCS phase that has been postulated in the past but still lacks direct experimental confirmation. These observations contribute to the understanding of the phases of exciton-polaritons at high densities.

By altering the grating geometry, it is possible to independently control the resonant frequency and the decay rate of the cavity mode and therefore of the resulting polariton mode. The tuning of polariton energy and linewidth with about 1 meV step size is demonstrated with fabricated devices. Combined with the capability of designing localized polariton modes, realizing these different parameters in a single device may facilitate the study of phase transitions arising from non-Hermitian physics.

A type of microcavity that utilizes the interference effect between the guided-mode resonance of the grating reflector and the background transmission continuum to achieve a high Q-factor is designed and tested. Reflection spectra measurements with a tunable cavity length setup show signatures of the cavity resonance. The Q-factor of the resonance has a strong frequency dependence that may allow leveraging the nonlinearity of polaritons for new devices.

CHAPTER 1

Introduction

1.1 Exciton-Polaritons in Semiconductors

Advances in the study of light-matter interactions have been one of the cornerstones of modern science and technology. Optical spectroscopy is widely used to learn the properties of new materials and to uncover new phenomena, while the optical properties of materials are utilized for the generation, detection, and manipulation of light. The past decades in particular have witnessed the burgeoning of many optical devices based on semiconductors. The relative ease of access and the development of manufacturing techniques of these materials have enabled practical devices from light emitting diodes and semiconductor lasers, to photovoltaic cells and image sensors. In the meantime, continued progress has been happening in emerging fields, such as low-dimensional materials, interaction with nonclassical light states, or nonlinear optical response due to the many-body interactions of charge carriers, an important characteristic of the semiconductor optoelectronic system.

At the heart of many properties of semiconductors are the electrons and holes, quasiparticles created by interband transition between the valence and conduction bands. Carrying opposite electric charge, they can form hydrogen-like bound states, called excitons, via their Coulomb interaction. In materials whose interband transition satisfies parity and spin selection rules, excitons can couple to light. The coupling strength $\hbar g$ in the dipole approximation is proportional to the interband transition dipole matrix element d_{cv} and the wave function of the electron-hole relative motion $\phi(r)$ at $r = 0$ [1],

$$\hbar g \propto d_{cv} \phi(r = 0). \quad (1.1)$$

When the thermal energy $k_B T$ is lower than the binding energy, the discrete exciton resonances often dominate the optical response near the band gap. In the low density limit $na_0^d \ll 1$, where n is the density, a_0 is the Bohr radius, and d is the dimensionality, excitons obey Bose–Einstein statistics. It has long been predicted that in quasi-thermal equilibrium, where the timescale of interband relaxation is much longer than intraband relaxation, excitons can form a Bose–Einstein condensate (BEC) [2, 3, 4], where a macroscopic population of the particles occupies the state with the lowest energy and the system exhibits off-diagonal long range order. Although first realized and subsequently extensively studied in atomic gas systems, BECs are expected to be achievable in less stringent conditions in semiconductor exciton systems, without the extensive experimental setup needed for trapping and extreme cooling. The critical temperature for an ideal non-interacting system [5]

$$T_c = \left(\frac{n}{\zeta(3/2)} \right)^{2/3} \frac{2\pi\hbar^2}{mk_B} \quad (1.2)$$

depends on m , the effective mass of the particles, and n , the density of the particles, and for excitons, is anticipated to be about 1 K. Experimental searches for exciton condensates have been attempted in different systems like cuprous oxide, coupled quantum wells (QWs), and transition metal dichalcogenides (TMDs) [6, 7, 8].

In more recent years, exciton-polaritons (hereinafter also abbreviated as polaritons) have attracted much attention as a promising system for BEC in semiconductors [9, 10, 11, 12]. When the coupling strength between the exciton and a photon mode exceeds the decay rates of the exciton and the photon mode, known as the strong-coupling regime, they will form new eigenmodes which are superpositions of exciton and photon, named exciton-polaritons [13, 14]. The lower and higher energy modes with eigenenergies

$$E_{\text{LP,UP}} = \frac{1}{2} \left(E_{\text{ex}} + E_{\text{ph}} \pm \sqrt{(E_{\text{ex}} - E_{\text{ph}})^2 + 4\hbar^2|g|^2} \right), \quad (1.3)$$

with $E_{\text{ex,ph}}$ being the exciton and photon energies and $\hbar g$ being the coupling strength, are often referred to as lower and upper polariton (LP and UP) respectively. A typical experimental structure

for exciton-polariton is a two-dimensional microcavity, formed by two parallel planar reflectors and embedded with QWs [Fig. 1.1]. The photon modes between the reflectors have an energy-wave vector dispersion

$$E_{\text{ph}} = \frac{\hbar c}{n} \sqrt{\left(\frac{2\pi}{L}\right)^2 + k_{\parallel}^2}, \quad (1.4)$$

where n and L are the refractive index and length of the medium between the reflectors and $k_{\parallel}$ is the in-plane wave vector parallel to the reflectors. Fig. 1.2 shows examples of the resulting polariton dispersion computed from Eq. 1.3 and Eq. 1.4 for three different cavity-exciton detunings. The effective masses of the polaritons are then a weighted mean of the two components,

$$m_{\text{LP,UP}} = \left(\frac{|X|^2}{m_{\text{ex,ph}}} + \frac{|C|^2}{m_{\text{ph,ex}}} \right)^{-1}, \quad (1.5)$$

where X and C are the coefficients of the superposition and $m_{\text{ex,ph}}$ are the effective masses of the exciton and the cavity photon. Because the effective mass of the cavity photon is usually orders of magnitude smaller than the exciton, the effective mass of the polaritons can also be much smaller. The critical temperature is raised even higher compared to excitons, ranging from cryogenic temperature to room temperature in a variety of materials like III-V and II-VI compounds [15, 16, 17, 18], organic semiconductors [19], perovskites [20], and TMDs [21], all of which are being actively studied nowadays. The spatial confinement of the exciton in the out-of-plane dimension also contributes to the realization of polariton BEC in the system. The Bohr radius is reduced compared to the three-dimensional system, resulting in enhanced coupling strength for reaching the strong-coupling regime. The increased binding energy raises the temperature at which the excitons will dissociate. Optical access is made easier due to the relaxing of the momentum matching condition in the dimension of confinement allowing all states within the light cone to be probed from the outside of the cavity. Utilizing this feature, polaritons can be measured when they decay via photons emitted from the cavity, since the photons inherit from polaritons properties like energy and phase, in-plane position and wave vector, and spin and orbital angular momentum. And finally, as the coupling strength is also proportional to the overlap between the photon mode and

the QW, the reduction of the photon mode volume increases the coupling strength too.

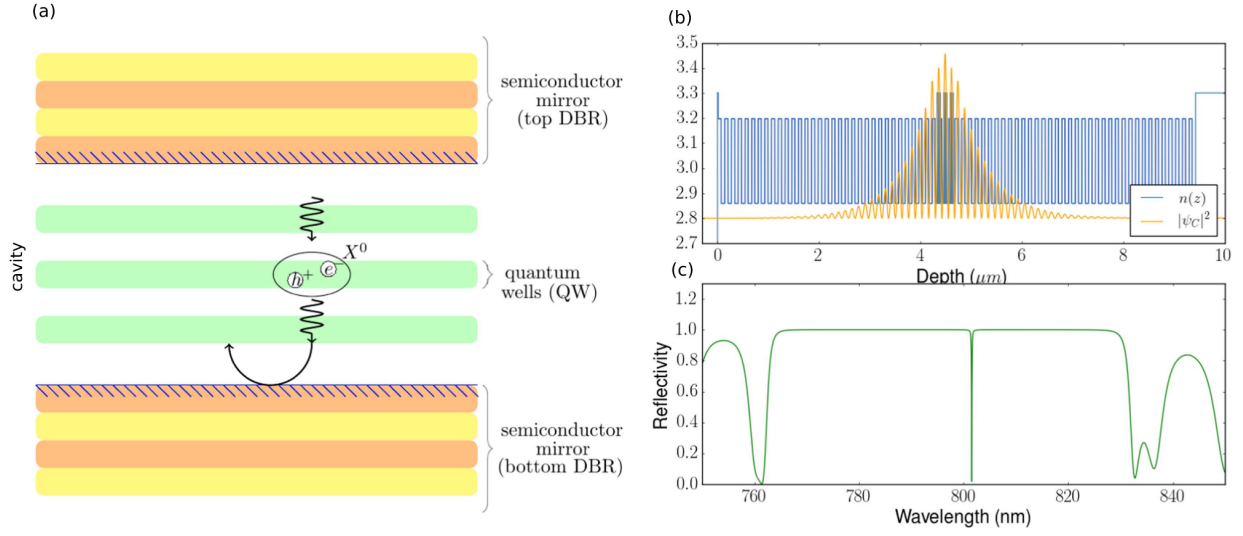


Figure 1.1: Structure and properties of a typical exciton-polariton microcavity. (a) Schematic of a microcavity with distributed Bragg reflectors (DBR) and embedded QW. (b) Refractive index $n(z)$ and cavity mode field distribution $|\psi_C|^2$ of a microcavity. (c) Computed reflection spectrum of the microcavity in (b). This figure is reprinted from Ref. [11].

The properties and applications of exciton-polaritons remain an ongoing topic of investigation. On the fundamental side, the nature of polariton condensates has yet to be fully understood [10]. Two important characteristics of polaritons play important roles here. First, the photon component of polariton reduces both the lifetime of the particles and their scattering rate compared to excitons. The lifetime of polaritons in GaAs QW microcavities is typically on the order of 10 ps, comparable to the timescale of their scattering dynamics. As a result, quasi-thermal equilibrium is not always reached in exciton-polariton systems and the observed condensation phenomena does not always satisfy the conventional definition of a BEC. Second, at high density, when the inter-particle distance is not much larger than the exciton Bohr radius, the excitons and polaritons cannot be simply treated as bosons anymore, and the fermionic statistics of the constituent particles of the excitons have to be considered. As the density increases, a transition towards a conventional photon laser can happen, specifically a vertical-cavity surface-emitting laser (VCSEL) for the microcavity structure. An intermediate state where the electrons and holes are no longer well-defined excitons but still pair up, analogous to the electron Cooper pairs in the Bardeen–Cooper–Schrieffer (BCS)

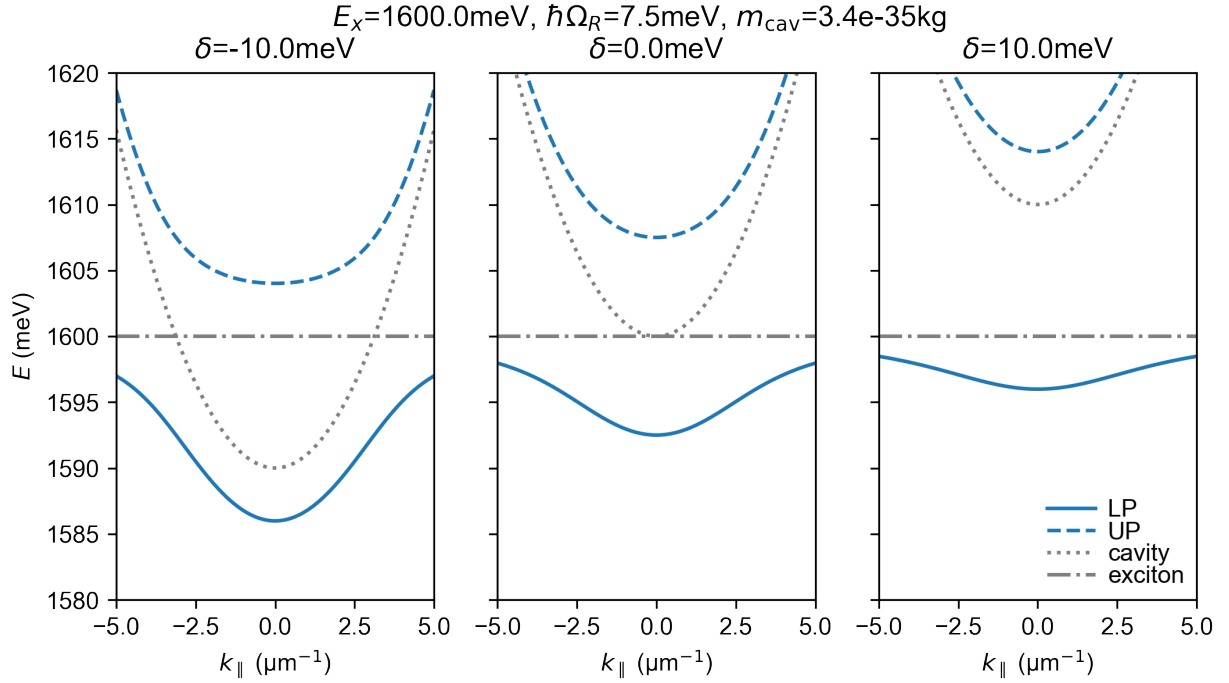


Figure 1.2: Polariton dispersion model. Examples of the polariton dispersion computed from Eq. 1.3 and Eq. 1.4 for three different cavity-exciton detunings δ . The gray dotted (dash-dotted) curves show the uncoupled cavity (exciton) dispersion. The blue solid (dashed) curves show the resulting LP (UP) dispersion. The parameters, exciton energy E_x , coupling strength $\hbar\Omega_R$, and cavity mode effective mass m_{cav} , are given in the titles.

theory, has been postulated. Theoretically, there have been attempts to map out a phase diagram for the polariton system [22, 23, 24, 25] [see Fig. 1.3 for an example], and experiments have been conducted to search for the signatures of these different phases [16, 26]. Despite the remaining unclear parts of the theory, on the application side, the exciton-polariton platform has already been shown to be capable of serving many roles like a low-threshold coherent light source [27] or a way of introducing nonlinearity into photonic devices [28]. Many types of devices have been implemented in the exciton-polariton system, including quantum simulators [29], topological insulators [30], ultra low energy switches [31], and neural networks [32].

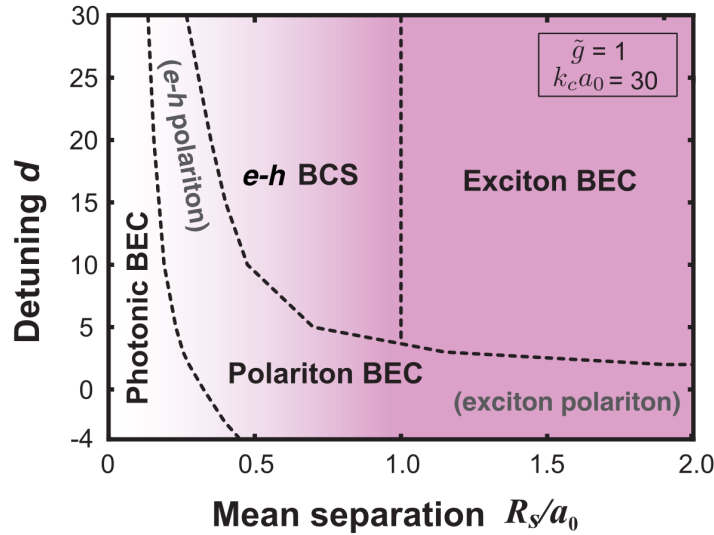


Figure 1.3: **An example of phase diagram of the exciton-polariton system.** Phase diagram at zero temperature in the parameter space of detuning d and mean separation R_s/a_0 , where a_0 is the exciton Bohr radius. $\tilde{g}$ is the coupling strength and k_c is the cutoff in momentum. The dashed lines mark crossovers between the phases, and the pink shade marks the region where Coulomb interaction dominates photon-mediated interaction. This figure is reprinted from Ref. [24].

1.2 Polariton Lasing and Condensation

Since the early observations of nonlinearity in polariton photoluminescence (PL) [33, 34, 35], one of the expected signatures of a polariton BEC, there have been different interpretations of the mechanism of such phenomena [36, 37]. There are multiple facets to this problem. According to the Hohenberg–Mermin–Wagner theorem, an infinite two-dimensional system does not undergo a BEC

transition at non-zero temperature [38]. In experiments, in-plane confinement is imposed on the polaritons, the simplest being just a finite-sized optical pump [16]. Here, the distance the polariton can propagate away from the pumping area is limited by their short lifetime. Other methods such as a stress trap or etched micropillars have been shown to facilitate polariton condensation [39, 40]. Instead of a BEC, a Berezinskii–Kosterlitz–Thouless (BKT) transition is expected for an infinite system with interaction [41, 42]. The signature of the transition would be the existence of bound vortex pairs and an algebraic decay of first-order phase correlation, also called quasi-long range order, below the critical temperature. With state-of-the-art wafer growth techniques, the lifetime of the cavity photons, and thus of the polaritons, has been prolonged by over ten-fold, allowing polaritons to propagate for a longer distance and form condensates with a much larger area [43, 44]. Algebraic decay of both spatial and temporal first-order coherence have been measured in these samples, providing evidence for the BKT phase. However, so far the spontaneous vortex pairs have not been discovered.

In addition, the polariton system is inherently non-equilibrium [45]. Typically, following excitations of the carriers, the relaxation towards the lowest energy state is driven by polariton-phonon and polariton-polariton scattering. The scattering is accelerated by the boson stimulation effect [46, 47, 48, 49], where the scattering rate towards a state of occupation number N is proportional to $(N + 1)$ [36], and exhibits a threshold-like behavior in the input-output relation. If the scattering rate is not much faster than the decay rate, quasi-thermal equilibrium is not always reached in exciton-polariton systems. It has been shown that the non-equilibrium nature, coming from the decay and pump, can significantly modify the properties of the condensate like the dispersion of the excitation spectrum [50, 51]. In many experiments, macroscopic occupation does not occur in the lowest energy state but a higher energy state that satisfies the balance between gain and loss [52], and the states can often be selected by adjusting the detuning or pumping condition [53]. Such situations are sometimes referred to as a polariton laser instead of a condensate, but at other times these terminologies are also used interchangeably. With recent progresses in long-lifetime polariton samples, evidences for quasi-equilibrium have been studied [54, 55].

Experimentally, to reach the critical condition for condensation in the polariton system, increasing the density, e.g. with increasing optical pump intensity, is often more convenient than decreasing the temperature. However, another complication arises with the change in density. In the low density limit $na_0^d \ll 1$, it is a good approximation to treat the excitons and polaritons as bosons. The shape of the interband polarization P_k is approximated by the exciton wave function

$$P_k = \sqrt{n}\phi_k, \quad (1.6)$$

and the distribution function of electrons and holes f_k , proportional to the density n ,

$$f_k = n|\phi_k|^2, \quad (1.7)$$

is far below 1. However, when the inter-particle distance becomes comparable with the Bohr radius, $na_0^d \gtrsim 1$, also referred to as reaching the Mott density, the fermionic statistics of constituent particles of the excitons has to be considered [56]. f_k starts to saturate and the shape of P_k deviates from the exciton wave function, which is referred to as the phase-space filling effect. At the same time, there is the screening of Coulomb interaction. Both reduce the electron-hole overlap and thus the coupling strength to light [57, 58]. For a laser-like transition observed as the density increases, the gain mechanism can depend on the density at the threshold. In the low density limit, the system can undergo a bosonic condensation, where the gain is due to bosonic stimulation of the polaritons. In the opposite limit, excitons are no longer bound but become electron-hole plasma, and the system is expected to be a conventional photon laser, with gain provided by stimulated emission of photons into the cavity mode when there is population inversion of the electronic band.

At intermediate densities, the electrons and holes are no longer well-defined excitons. It has been postulated that the carriers will form overlapping pairs similar to Cooper pairs in superconductor BCS theory [59], weakly bound by the screened Coulomb interaction as well as the effective attractive interaction induced by the coupling to photons. Following the original superconductor BCS theory, a BCS-like phase has been first predicted for excitons in semiconductors without a

cavity [3, 4, 60, 61]. A smooth crossover between the BEC and the BCS phases, akin to that in atomic gases [62], is expected since both are U(1) symmetry breaking, with no other symmetry breaking in between them. The two phases share the same form of mean field wave function at zero temperature

$$|\Phi\rangle = \exp(\lambda\psi_0^\dagger)|0\rangle = \prod_k \exp(\lambda\phi_k a_k^\dagger b_k)|0\rangle = \prod_k (1 + \lambda\phi_k a_k^\dagger b_k)|0\rangle \rightarrow \prod_k (u_k + v_k a_k^\dagger b_k)|0\rangle, \quad (1.8)$$

where $\psi_0^\dagger$, $a_k^\dagger$, and b_k are the creation operators of exciton, electron, and hole, $|0\rangle$ is the ground state, $|\lambda|^2 = N$ is the mean exciton density, and the last step is normalization. External light-driven electron-hole BCS phases have also been studied [63, 64]. Combining the two situations, BCS-like phases have been theoretically proposed for microcavity polariton systems, with a model of localized two-level systems coupled to cavity [23, 65]. More recent theories considered realistic band structures, with Coulomb interaction in Hartree-Fock approximation at zero temperature and a wave function in the form of

$$|\Phi\rangle = \exp(\alpha a^\dagger - \alpha^2/2) \prod_k (u_k + v_k a_k^\dagger b_k)|0\rangle \quad (1.9)$$

where $a^\dagger$ is the creation operator of the cavity photon mode and $|\alpha|^2 = n_{\text{ph}}$ is the photon density [66, 24], or with a contact potential in non-equilibrium open-dissipative systems, yielding predictions of gain spectra and phase diagrams including non-equilibrium phases [67, 68, 25]. It has been found that the coupling term between the electron-hole pair and the cavity photon $\hbar g \alpha$ shows up in parallel with the gap function

$$\Delta_k = \sum_{k'} V_{k-k'} u_{k'} v_{k'} \quad (1.10)$$

which is attributed to the Coulomb interaction V , both of which provide the binding of the electron-hole pair. The polarization $P_k = u_k v_k$ can be maximal at non-zero wave vector, while the distribution $f_k = v_k^2$ can be broadly spread out. Fig. 1.4 shows a schematic of the charge carriers in the BEC, BCS, and photon laser phases.

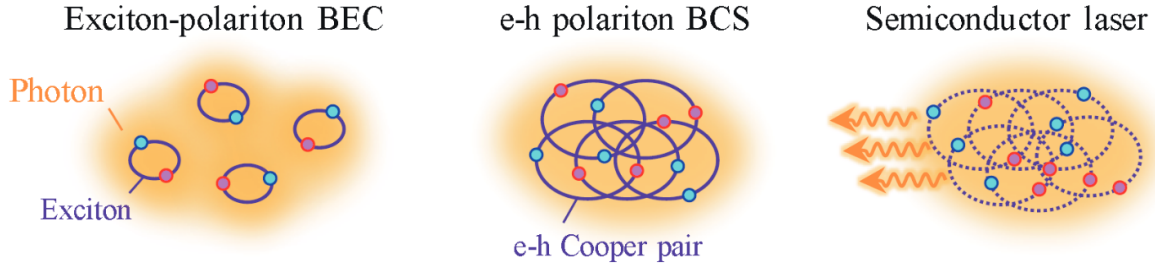


Figure 1.4: **Schematics of different phases in the exciton-polariton system.** This figure is reprinted from Ref. [69].

The identification of all these phases in polariton systems is still a challenge of both theoretical and experimental interest. While all three lead to coherent emission, they have distinct microscopic characteristics such as their gain mechanism, quasiparticle type, and electron-hole distribution functions.

The first demonstrations of laser-like threshold behavior of polaritons have been shown to be in the weak-coupling regime [33, 34]. Only with improved sample design and fabrication techniques has convincing evidence of polariton condensation been observed [15, 16]. Here, a main factor is the increase of the number of QWs in the cavity, which increases the coupling strength $\hbar g \propto \sqrt{N_{QW}}$ and reduces the density of excitons per QW at the same polariton density. In many experiments, the criteria for distinguishing between a polariton BEC and a conventional photon laser include the energy of the emission. While the polariton energy is lower than both exciton and cavity due to the Rabi splitting, at high density the system enters weak-coupling [70], and the laser emission is expected to be at the cavity energy. But this is not always reliable as other factors can lead to a shift of the lasing energy [71]. Another important criterion is the energy-wave vector dispersion because the polariton and cavity modes have different effective masses. But this is not always easily observable in all types of cavities, and may also be subject to exciton shifting. A second threshold has been observed under various pumping conditions, and is attributed to the transition between a polariton BEC to a conventional photon laser [35, 72, 40]. However, the mechanism of this transition has not yet been subject to systematic experimental study [73].

Experimentally, a BCS-like phase remains elusive. Despite the different lasing mechanisms, these phases all have coherent emission, making it difficult to distinguish between them. Only a few experiments have reported non-standard PL features that could be possibly linked to BCS physics [26, 74]. The transition between BEC-BCS-photon laser also remains to be explored.

1.3 Modifying the Properties of Polaritons by Engineering the Cavity

Exciton-polaritons have become a versatile platform for a wide range of research including many-body physics, quantum optics, non-equilibrium dynamics, and their intersections. Many of these studies involve engineering of the polariton modes. For example, by reducing the spatial size of the polariton mode, coherence properties and interaction strength of a polariton laser can be modified [40, 27, 75], and the enhanced interaction is desirable for quantum photonic studies [28, 76]. By constructing a periodic potential, energy band structures can be realized in polaritons, leading to novel phenomena, such as polariton topological insulator [30], and enabling devices like Hamiltonian simulators [29].

As superpositions of an exciton and a photon mode, the polaritons are affected by both their matter and light components. The energy and the interaction strength of the polaritons can be affected by those of the underlying excitons. One can distort the lattice with strain or mechanical waves to change their energy [39, 77]. By modulating the local density of excitons, for example, with a structured optical pump, an effective potential for the excitons that originates from the repulsive interaction between them can be created [78]. More recently, Moiré patterns in stacked two-dimensional materials are found to modulate the excitons [79, 80]. The excitons also respond to external electric or magnetic fields [81]. The photon mode is typically determined by and confined within a cavity, which can have a great variety of designs and can be constructed to high precision at large scale using modern fabrication techniques. Properties of the polariton mode such as spatial distribution, energy-wave vector dispersion, and decay rate crucially depend on the photonic mode and therefore can be tailored to a large extent. The most widely used cavities for polaritons have been based on Fabry–Pérot (FP) cavities formed by distributed Bragg reflectors

(DBR). Unconventional structures, such as photonic crystals, have emerged, and they allow more versatility in mode engineering and control.

The FP microcavity made of two DBR mirrors has been an archetypal structure for exciton-polaritons [13]. A DBR mirror is built up with quarter-wavelength layers of two alternating materials of different refractive indices. Due to the interference between the waves reflected at the interfaces between the layers, the mirror has a stop band in its reflection spectrum with high reflectivity. The width of the stop band increases with the contrast between the two refractive indices, and the peak reflectivity increases with the number of the layers. Formed by two parallel DBR mirrors, the photonic resonance mode has a quasi-two-dimensional geometry, featuring a parabolic in-plane dispersion at small wave vectors [Eq. 1.4], with tight confinement in the other direction [see Fig. 1.1(b) for the intensity distribution]. Two properties of such planar cavities can be readily tuned: the quality factor (Q-factor) and the resonance energy of the cavity mode. For III-V semiconductors, monolithic DBR-DBR cavities with the QWs embedded inside can be grown epitaxially with minimal defects or impurities. By increasing the number of layers in the DBR, Q-factors higher than 3×10^5 has been achieved [55]. The cavity layer thickness typically decreases from the center of the wafer toward the edge, leading to a gradual change of the cavity-mode resonance. The polarization dependence of the reflection coefficient of the mirrors can give rise to a TE-TM energy splitting of the cavity mode [82] which contributes to the optical spin Hall effect of polaritons [83, 84].

There are various methods to modify the DBR cavity locally, especially the resonance energy and the spatial extent of the modes. A non-destructive method is depositing patterned metal films on the top surface of the sample. These metal films will locally shorten the effective cavity length and increase the resonance energy due to suppression of the electromagnetic field inside them. The modulation strength is limited by the thickness of the upper DBR and is typically on the order of 0.1 meV. So this method is best suited for creating a weak periodic modulation for polaritons. It was used to demonstrate the first one-dimensional periodic potential for microcavity potentials, leading to the observation of phase-locked condensate arrays [85]. Later, many types of two-dimensional

lattices have also been created, with dynamic condensation in high-orbital states [52, 86, 87, 88, 89]. To create a stronger modulation on the cavity mode, after the growth of the bottom DBR and cavity layer with QWs, the cladding of the cavity can be patterned through lithography and etching and the growth of the top DBR continues afterwards [90]. The pattern embedded at the bottom of the top DBR modulates the cavity length and the cavity resonance up to a few meV. Polariton PL and condensation have been studied in single potential traps [91, 92, 93] as well as lattices [94, 95, 96]. Alternatively, etching patterns from the top of the DBR to the cavity layer or to the substrate can impose strong lateral confinement that shapes local modes. Polariton lasing in such lower-dimensional structures was first demonstrated in quasi-zero-dimensional micropillar cavities down to about $3\text{ }\mu\text{m}$ in diameter [40]. Similarly, quasi-one-dimensional devices have been created, which can be used to guide polariton flow [97, 98, 99, 100, 101]. When the micropillars overlap, localized modes at neighboring sites can couple and form molecule-like devices [102]. Extended to a larger array, a lattice of polaritons can be created. In one-dimensional modulated wire and one-dimensional Lieb lattice, effects related to interaction and disorder such as gap soliton [103, 104] and localization [105] have been observed. Two-dimensional honeycomb [106, 107] and Lieb [108] lattices have been studied. With an external magnetic field that breaks the time reversal symmetry, topological edge modes have been found in a honeycomb lattice [30, 107]. Finally, concave DBR mirrors, fabricated as a standalone mirror or integrated into the end of a fiber, give rise to a focusing photon mode, which also leads to laterally confined polaritons [109, 110, 111]. Polariton lasing has been observed in such a system [110, 112], and the TE-TM anisotropy can be strong enough to induce energy splittings between different vortex states [113]. The lateral confinement leads to enhanced interactions and photon antibunching has been measured [28, 76].

1.4 Subwavelength Grating Reflectors

The DBR can be replaced by slab photonic crystals, which enable not only potential landscape engineering, but also modification of other fundamental properties of the cavity mode, such as its polarization state and energy-wave vector dispersion. While the design flexibility of slab photonic

crystals has been intensely researched since the late 1990s with many remarkable demonstrations, a challenge in using them to construct a vertical cavity is the requirement of high reflectivity. One solution is to use gratings with a period smaller than the wavelength to eliminate diffraction other than the zeroth order, and then utilize destructive interference of the internal resonances at the interface to achieve high reflectivity in a broad spectral range. Fig. 1.5(b) shows an example of a subwavelength grating (SWG) [114]. Here, the structure is periodic in x -direction with two alternating materials of different refractive index. The period is denoted by Λ and is smaller than the working wavelength in the surrounding low refractive index material, and the duty cycle $\eta = w/\Lambda$ is the ratio between the width of the high index material and the period. It has a finite thickness t_g in z -direction, but is infinite in y -direction. Light is incident from above the x - y plane, with polar and azimuthal angles (θ, ϕ) with respect to the z -axis. The resonances of the grating can be viewed as approximately FP resonances of waveguide array modes propagating along z -direction. Assuming that t_g approaches infinity, the structure would become an array of slab waveguides [Fig. 1.5(a)], and the dispersion relations of the waveguide array (WGA) modes at normal incidence is shown in Fig. 1.5(c). For finite t_g , they are reflected at the boundaries, and there will be mixing between these modes. Nevertheless, the resulting eigenmodes will be superpositions of these modes, and will form FP resonances when the round-trip phase condition is satisfied. A plane wave at normal incidence can excite the resonances of even modes (with respect to the y - z plane of symmetry of the structure), with different amplitude and phase depending on the wavelength and t_g . The t_g -dependence of the resonance frequencies of the lowest two even modes is shown in Fig. 1.5(d). A checkerboard pattern shows up in the reflection map [Fig. 1.5(e)] and the boundaries agrees well with the curves of the FP resonances. The high reflectivity regions correspond to when the zeroth-order components of two or more resonances cancel out at the interface between the grating and the medium on the transmission side. Because of the subwavelength period, higher order components do not couple to the surrounding low index medium, and all the energy flow has to return as reflection.

Alternatively, these resonances can be viewed as superpositions of counter-propagating modes

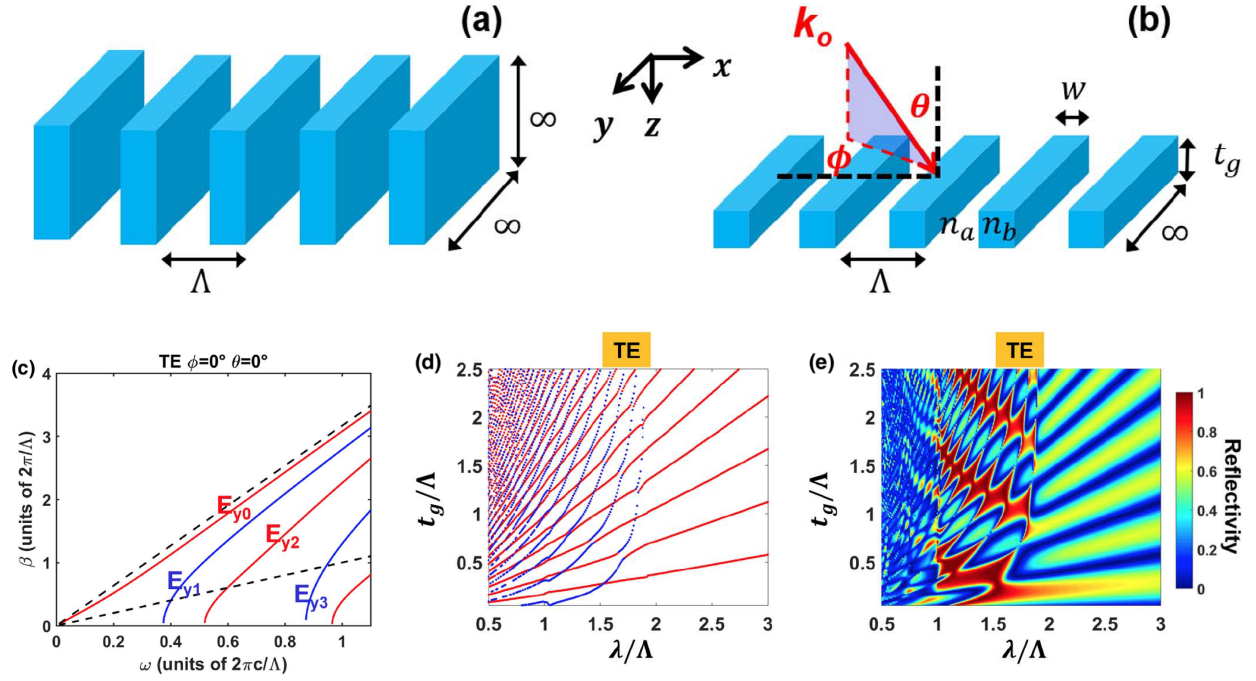


Figure 1.5: **Structure and resonance modes of a high-contrast SWG.** (a) Schematic of a one-dimensional WGA. (b) Schematic of a one-dimensional high-contrast SWG with period Λ , thickness t_g , bar width w . The refractive indices of the two materials are n_a and n_b . The incident light of wave vector k_o and angles (θ, ϕ) is also marked on the subfigure. (c) Dispersion of TE modes of the WGA at normal incidence. The red and blue colors represent even and odd modes. The dashed lines are the light lines in the two materials. (d) TE-polarized resonances of the SWG corresponding to the lowest two even modes of the WGA. The resonances occur when the FP condition is satisfied. (e) Reflectivity of the SWG. This figure is reprinted from Ref. [114].

in a single slab waveguide modulated by the periodic refractive index [Fig. 1.6(a)] [115, 116]. If the duty cycle $\eta = 1$, the structure is a slab waveguide extending in the x - y plane and the waveguide modes are below the light line and do not couple to the outside. However, the periodic structure provides the additional momentum for phase-matching of the coupling between the diffraction orders, and the dispersion curves of the waveguide modes are folded and exposed into the light cone [Fig. 1.6(b)]. These modes are also referred to as guided-mode resonances (GMR). Two counter-propagating modes along the direction of the periodicity can be coupled via second order or higher diffraction and form an even and an odd eigenmode at $k_x = 0$, both being standing wave patterns, with a gap in frequency between them. The even mode can couple to the $k_x = 0$ plane wave outside and induces a reflection peak via interference effect with the transmission. Multiple such modes can be arranged to form a wide reflection band. The odd mode, forbidden by symmetry, does not couple to the outside, and is often referred to as a bound state in the continuum (BIC). The WGA and GMR interpretations of the grating have the following connection: the transverse order of the WGA modes corresponds to the order of folding of a single GMR mode, while the order of FP resonances of a single WGA mode corresponds to the transverse order of the GMR mode.

Using such an SWG reflector to replace the DBR has been demonstrated and actively studied in semiconductor VCSELs [118, 119, 114]. Their application to the polariton lasers was first demonstrated in Ref. [120] [see Fig. 1.7]. The gratings are usually suspended to suppress non-zeroth order diffraction. However, monolithic variants have been demonstrated for polariton lasers, allowing higher mechanical strength and stability [121].

The SWG reflectors provide many degrees of freedom to control the polariton modes. Strong lateral confinement of the polaritons can be achieved by controlling the sizes and patterns of the grating, without the need to etch through the QWs or to embed structures inside the cavity structure [120]. For suspended gratings, stress-induced bending of the grating can also be utilized to modulate the cavity length and potentials can be created with additional etched patterns around the gratings [122, 123]. These quasi-zero-dimensional devices exhibit discrete states and shot-noise-limited intensity stability owing to suppressed mode competition and depletion [27].

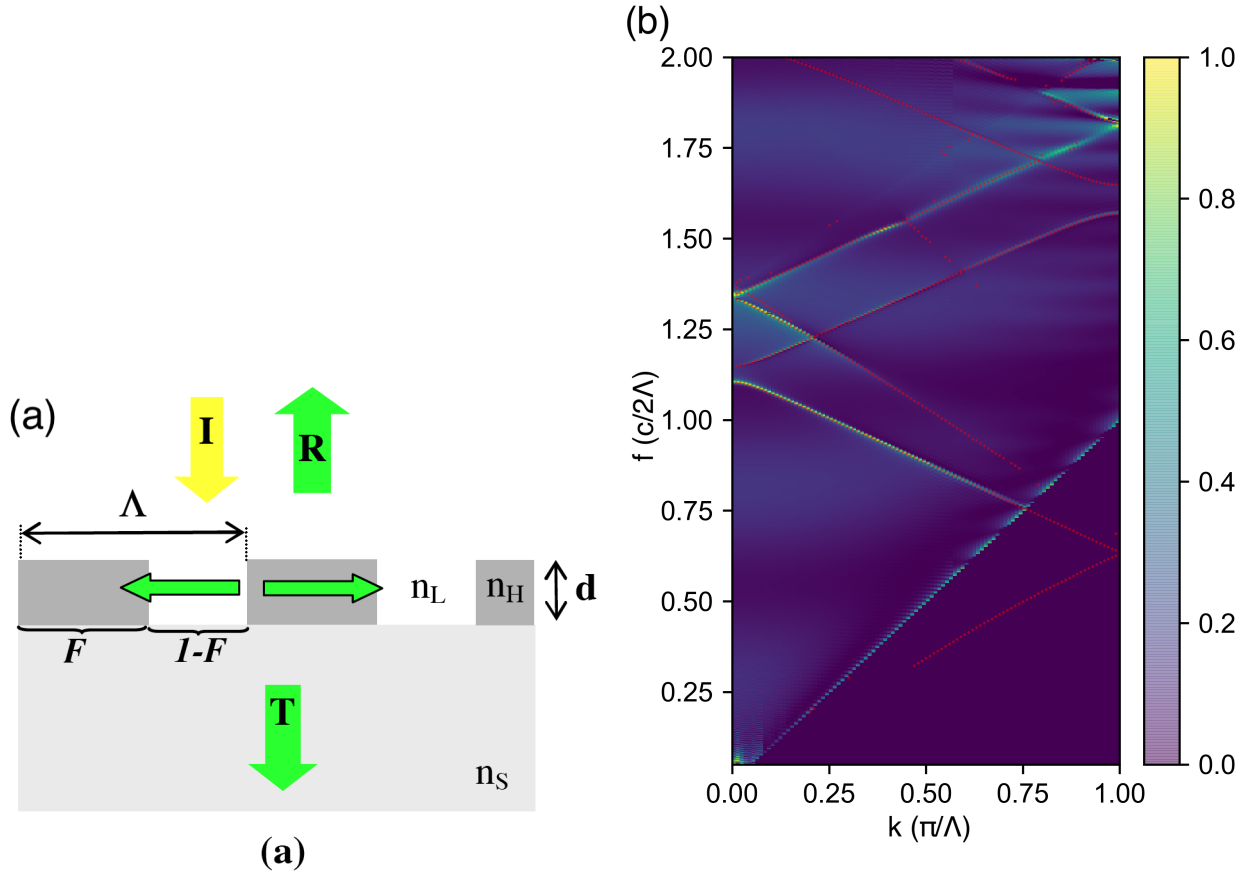


Figure 1.6: **Guided-mode resonances of an SWG.** (a) Schematic of an SWG with materials of refractive indices n_L , n_H , and n_S as the low-index, high-index parts of the grating, and the substrate respectively. The grating has period Λ , thickness d , and duty cycle F . The arrows labeled I , R , and T denote the incidence, reflectance, and transmittance and the unlabeled arrows denote the guided modes. This subfigure is reprinted from Ref. [116]. (b) Simulated reflection spectra of an SWG with refractive indices $n_L = 1$, $n_H = 2.01$, and $n_S = 1.45$. The units are normalized to a grating period of $\Lambda = 0.5$. The thickness of the grating is $d = 0.5$ and the duty cycle is $F = 0.9$. The red dots mark resonances with $Q > 50$ found numerically. The simulation is performed with a finite-difference time-domain (FDTD) program [117].

The grating also has distinct features unavailable with DBRs, such as strong polarization dependence and controllable reflectivity and reflection phase. By designing the reflection phase of grating, energy-momentum dispersion of the polaritons can be engineered, such as to produce a lighter or heavier effective mass, flat-band polaritons, or double-well-shaped dispersion [124]. The grating can generally provide high reflectivity in one linear polarization but low reflectivity in the orthogonal one, due to the differences between the two sets of internal resonance modes belonging to the two polarizations. This gives the cavity high and low Q-factors for the two polarizations. Hence, strong-coupling and weak-coupling coexist at the same spatial location when the grating mirrors are used [120].

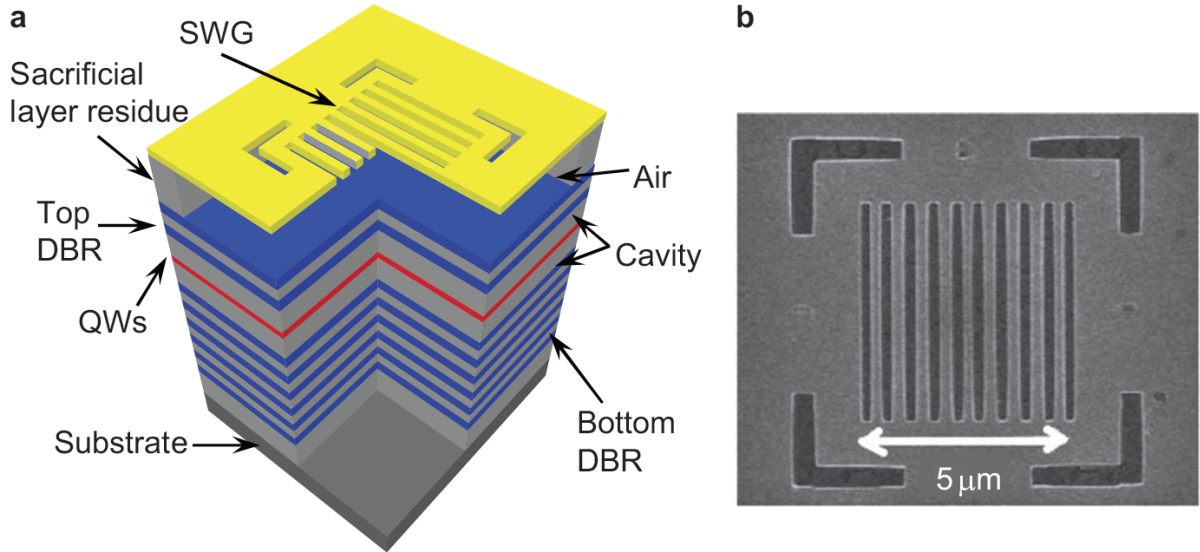


Figure 1.7: **Examples of an SWG-DBR hybrid cavity.** (a) A schematic of a zero-dimensional hybrid cavity with an SWG mirror. (b) Top-view SEM image of a fabricated zero-dimensional cavity with a $5\,\mu\text{m} \times 5\,\mu\text{m}$ SWG. This figure is reprinted from Ref. [120].

A related idea is that, instead of using a slab photonic crystal as a mirror, the photonic crystal can function as the photonic resonator by itself. The photonic band structure can be tailored directly via the lattice structure instead of relying on modifying an FP resonance. For example, a defect in the photonic crystal can form a resonance in the photonic band gap that has high Q-factor and diffraction limited mode volume [125]. However, the integration of an exciton medium with a

photonic crystal is challenging for conventional semiconductors. The QW can be embedded in and patterned together with the photonic crystal [126], or be placed away from the photonic crystal but couple to the evanescent field of the photonic crystal mode, with a reduced coupling strength [127]. Atomically thin van der Waals semiconductors crystals can be placed directly on top of a photonic crystal slab, where the field enhancement remains large [128]. However, the Q-factor of these schemes is still much lower than the best FP cavities.

1.5 Overview of This Dissertation

Experimental research on the photonic control of the exciton-polariton system, mainly utilizing microcavities with an SWG reflector, will be presented in this dissertation. The engineered properties of the system provide possibilities to probe polariton-related physics, such as the identification the lasing mechanisms and therefore the type of phase transitions in the high density regime. They also bring forth potential in realizing novel devices, such as devices implementing a non-Hermitian model.

First, with a polarization-selective hybrid SWG-DBR polariton microcavity, direct access to the electronic component of the polariton in the presence of lasing becomes possible. This is utilized to explore the lasing mechanism of the polariton system in the high density regime, where different phases may exist, including a theoretically predicted BCS-like phase whose experiment observations are still elusive to date. With a technique combining an off-resonant continuous wave (CW) pump and a resonant pulsed probe in time-resolved spectroscopy, both the absorption and PL emission spectra of both the polariton lasing mode and the electronic reservoir are measured. Optical gain is identified in the polariton laser above the Mott density, which distinguishes it from a polariton BEC. At the same time, the emission spectra manifest signatures of bound electronic states in contrast to a conventional photon laser. To study the nature of the observed polariton laser, the experimental results are compared to a microscopic many-particle theory for the Coulomb correlated electron, hole and photon system, which reveals distribution functions and interband polarizations resembling those of an ideal ($T = 0$ K) polariton BCS state [66, 24], but modified

by cavity dissipation and thermal dephasing. The experimentally observed emission energies and absorption spectra agree well with the theoretical predictions. The dependence on cavity-exciton detuning of the system is studied and a transition from a polariton BCS-like phase to a phase consistent with conventional photon lasers as the detuning increases is observed. To verify the generality of the phenomena, PL measurements are performed on a conventional DBR-DBR microcavity with fewer QWs, which reaches Mott density at lower pump powers, and similar results are observed.

With a change of the grating reflector from a suspended structure to a monolithic one, finer control of the polariton mode properties becomes possible. This is particularly relevant to the study of non-Hermitian physics, a framework for describing open-dissipative systems, including but not limited to optics as well as exciton-polaritons, and currently heavily influencing photonics research and applications. It often requires precise tuning of the system parameters, including the real and imaginary part of energies of the individual modes and the coupling strength between them. A method to independently control both the energy and linewidth of the photonic cavity mode, and consequently the polariton mode, is demonstrated. The reflectivity and reflection phase of the grating, which affect the cavity decay rate and resonance energy respectively, can be tuned via the geometric parameters of the grating. The complex eigenenergy of the polariton is determined in a completely passive way during the fabrication of the device. The gratings also define the in-plane spatial extent of the polariton mode, therefore allowing the creation of multiple localized modes with controllable coupling strengths that could be potentially extended to large scale devices. Devices are fabricated and characterized, showing a tuning range on the order of a few meV for the eigenenergy and from sub-meV to a few meV for the linewidth, appropriate for models that require the energy, the linewidth, and the coupling strength to be comparable. Initial results on the controlled coupling between neighboring localized modes are presented.

Finally, an open cavity structure consisting of a base sample with DBR and QW and an SWG reflector on a separate sample is proposed. With this type of cavity, the material choice of the grating is no longer limited to the same category of epitaxial materials as in the previous examples.

The cavity should be applicable to both GaAs and TMDs QWs and can potentially realize features like Q-switching with specialized design. A grating reflector design harnessing a Fano resonance is used, and numerical simulations show that the resulting hybrid cavity supports polariton modes and provides high Q-factor. Preliminary measurements of an empty cavity without QWs are shown.

CHAPTER 2

Polariton Laser in the High Density Regime

2.1 Experimental Approaches to High Density Polariton Systems

To date, most experiments on the spontaneous formation of an exciton-polariton condensate under non-resonant optical pumping have been conducted in the low density regime, where polaritons are formed by cavity photons and bound state excitons and can be treated as bosons. These include the measurements of lasing characteristics [15, 16, 39], correlation functions [129, 130, 27, 131, 75, 132], thermalization [54, 55], interaction strength [133], excitation spectra [134, 135], and vortices [136, 137, 138]. Some commonly observed features are a nonlinear threshold-like behavior in the light intensity input-output relation, and across this threshold the narrowing of emission linewidth, the increase of degree of polarization, and the increase of spatial and temporal coherence. As a result of polariton-polariton interaction, a blueshift of emission energy with increasing density is observed, as well as linear excitation spectra at small wave vector. While these features are consistent with the polariton BEC picture, experimentally it is difficult to estimate the carrier density in the system to verify that the system is indeed below Mott density. The density can be measured from pump or emission intensity [72, 40, 139, 140], but may have large uncertainties due to the nonradiative states and nonradiative decay channels.

In some experiments two types of thresholds in the emission intensity are observed. The two thresholds can sometimes be observed in the same sample with the same experiment conditions except for the pump power and therefore the carrier density [35, 40, 141, 142, 143, 126, 144, 73]. In other cases the type of the threshold observed is affected by other conditions such as the cavity-exciton detuning [72, 145, 126], the temperature [144], the existence of a potential trap

[141, 142, 73], the time after pulse excitation [146, 147, 134], or the strength of an applied external electric field [148]. Often the threshold at a lower pump power is attributed to polariton BEC condensation while the one at higher pumping power is attributed to photon lasing with unbound electron-hole pair and population inversion, based on features of the emission such as whether the energy is below or at the bare cavity energy, whether the dispersion curve has the effective mass of the polariton or the cavity, or whether the second order temporal correlation function $g^{(2)}(0)$ decreases to 1 or not. But these criteria are not always reliable and consistent among different experiments. For example, although the carrier density when population inversion occurs is comparable to or above the Mott density, and the photon lasing energy is expected at the cavity resonance due to the loss of strong-coupling, it has been shown that the emission energy after the high-pump-power threshold can be either below or above the cavity and can either be pinned at one energy or shift with changes of density [71, 141, 73]. The effective mass measured from dispersion can be affected by the change of detuning due to exciton blueshift, and is also difficult to determine for finite-sized devices with discrete states. The correlation function $g^{(2)}(0)$ can also be strongly modified by the size of the device [75].

Contrary to the expectation that at high density the system becomes a photon laser, possibly after a second threshold, a few experiments focusing on the high density regime have provided evidence calling for new interpretations. Ref. [26] reported high-energy side peak emission towards the Mott density, partly consistent with a non-equilibrium polariton BCS theory. In Ref. [74] the negative branch of the excitation spectrum indicative of the presence of strong-coupling is observed even at high densities where a transition to weak-coupling is typically expected. In Ref. [133] the dependence of emission blueshift on density departs from the linear relationship at low density as the system approaches Mott density. These results also hint at possible BCS-like states but verifications are still needed.

An alternative category of methods to determine the mechanism of the coherent emission from polariton system is to probe the underlying electron-hole medium. Compared to analyzing the PL spectra, they often provide evidences that are less model-dependent. With terahertz (THz)

spectroscopy, bound and unbound electron-hole pairs show different characteristic lineshapes [149]. Zeeman splitting [150] and diamagnetic shift [151] have also been used as indicators of the presence of excitons.

In the following sections, a method of optically probing the matter component of a polariton laser will be presented. This is based on the polarization selectivity of a microcavity with an SWG reflector. Strong-coupling and weak-coupling coexist in this cavity, but in orthogonal linear polarizations. From the PL spectra of the polariton laser in the strong-coupling polarization, the observed features are consistent with what would typically be regarded as a polariton BEC. There is a single threshold where the emission intensity of the lowest state grows super-linearly and becomes the dominant component in the spectra. The emission energy has a smooth blueshift before and after the threshold, without features expected for a photon laser transition, such as any sudden jumps in the energy [152, 35, 153, 141, 146, 144] or large broadening before or even after the threshold similar to previous reports [40, 141, 126, 146, 145]. In the weak-coupling polarization, however, reflection spectra show a transition from a discrete exciton resonance to band-edge-like absorption before the onset of lasing emission, and optical gain accompanying the lasing. This puts the sample in the high density regime, but the discrepancies need explanation.

2.2 Sample Structure and Properties

The microcavity sample used here has the same structure as in Ref. [120] and is shown schematically in Fig. 1.7 and Fig. 2.2(a) and the gratings are fabricated by Seonghoon Kim. It has three sets of four 12 nm wide GaAs QWs with 4 nm AlAs barriers embedded at the three central anti-nodes of a $\lambda/2$ AlAs cavity (with two of the three sets inside the mirror layers next to the cavity). The bottom mirror is a DBR with 30 pairs of $\text{Al}_{0.15}\text{Ga}_{0.85}\text{As}/\text{AlAs}$ layers. The top mirror consists of 2.5 pairs of DBR and an $\text{Al}_{0.15}\text{Ga}_{0.85}\text{As}$ high-contrast SWG suspending over the DBR. The grating is about 80 nm thick, with a duty cycle of 0.4 and a grating period of 520 nm. The size of the grating is $7.5\text{ }\mu\text{m} \times 7.5\text{ }\mu\text{m}$. The grating has very high reflectivity for transverse-electric-polarized (TE, electric field parallel to the grating bars) light, but low for transverse-magnetic-polarized (TM,

electric field perpendicular to the grating bars) light. Fig. 2.1 shows simulated¹ reflection spectra of the grating for light near 800 nm at normal incidence with TE and TM polarization. In addition to the grating, the 2.5 pairs of DBR layers on top of the cavity increase the reflectance for both polarizations. But the reflectance for TM-polarized light is still much lower than TE.

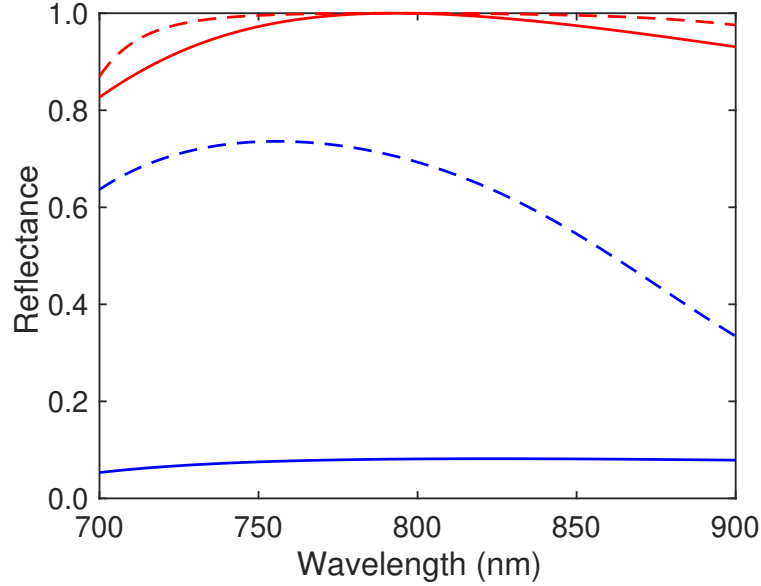


Figure 2.1: **Simulated reflectance of the grating-based linear polarization-selective mirror.** Numerical simulation of the reflectance of the SWG (solid lines) and the top mirror (dashed lines) consisting of the grating and 2.5 pairs of DBR, at normal incidence, for TE-polarized light (red lines) and TM-polarized light (blue lines). The top mirror has very high reflectance for TE and low reflectance for TM.

As a result, TE-polarized polaritons are formed while TM-polarized excitons remain in the weak-coupling regime [Fig. 2.2]. The different reflectance under the grating and outside the grating leads to laterally confined, fully discrete TE-cavity and TE-polariton modes.

The energies of lower polariton (LP) and upper polariton (UP) resonances of each device are measured by reflection and low-power photoluminescence spectroscopy [see Section. 2.3]. The exciton resonances are measured from the unetched part next to the device. Using the measured LP, UP and exciton resonances, the cavity resonance energy, detuning, and normal-mode splitting are estimated for each grating device.

¹The simulations are performed with a rigorous coupled-wave analysis (RCWA) [154] program [155].

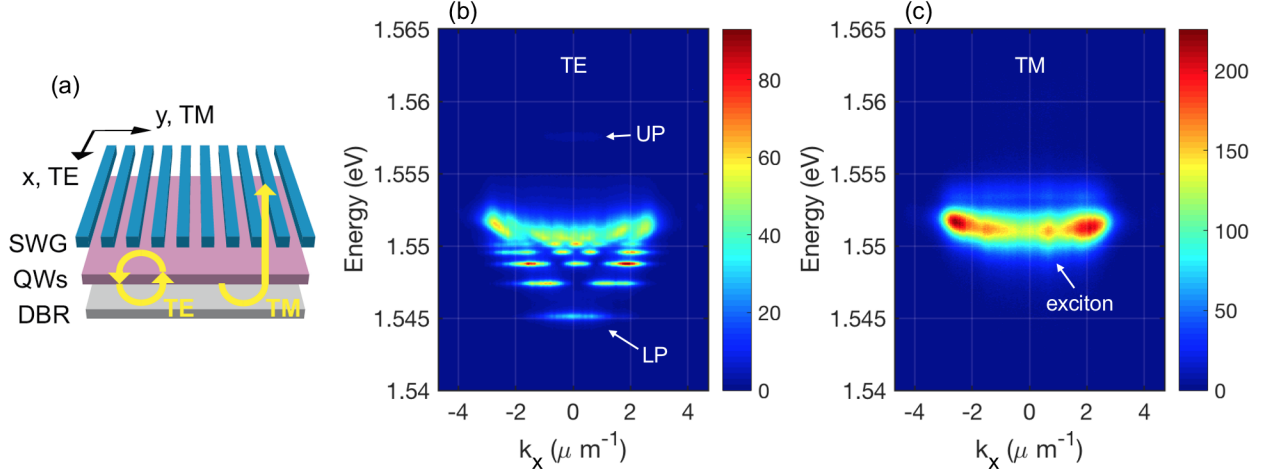


Figure 2.2: **Polarization-selective grating-based microcavity.** (a) Schematic of the microcavity showing SWG as top mirror and DBR as bottom mirror. The reflectance of the SWG is high for TE-polarized (also labeled x) light and low for TM-polarized (also labeled y) light. (b) PL spectrum from the microcavity in TE-polarization. There are LP and UP modes due to strong-coupling. (c) PL spectrum from the microcavity in TM-polarization. The exciton is only weakly-coupled to light.

2.3 Measurement Setup

The main measurements performed on the samples are PL and reflection spectra under varying pump power, for both TE- and TM-polarization. Fig. 2.3 shows a schematic of the measurement setup. The sample is kept at 10 K in a Janis ST-500 or Montana CR-509 cryostat for all the measurements. A continuous-wave (CW) pump is used for excitation, at 784 nm, about 30 meV above the exciton resonance, focused to a Gaussian spot of about $3\text{ }\mu\text{m}$ in diameter at the center of the square grating. PL is measured with a spectrometer through a $4f$ confocal relay, which provides switchable real-space and Fourier-space imaging capabilities. The reflection spectra, with the presence of the PL, is measured with a streak camera after a monochromator. Since the transmission through the bottom mirror of the sample is expected to be negligible, the absorption spectra can be obtained from the reflection spectra. A resonant pulsed laser of about 150 fs pulse width is used as a probe, overlapping with the pump spatially, with a similar spot size. The PL and the reflected probe can then be separated, since the reflected probe appears as a resolution-limited sharp peak in time in the streak image whereas the PL, produced under CW pumping, has uniform

intensity in time. Hence, the PL is readily subtracted from the reflection. The total intensity of reflected pulse is obtained by integrating over the whole pulse, and is then divided by the reflection spectra from a reference gold mirror to produce the reflection spectra of the sample. At high pump powers, optical chopping of the pump laser is required to limit sample heating, and the microchannel plate gate of the streak camera is used to block out the probe signal while the pump is off. This requires synchronization of timing between the chopper and the streak camera, which also has to be synchronized to the pulsed laser. For this purpose, a photodiode picks up the laser pulses which have a repetition rate of 80 MHz, and the signal is split into two paths. One goes to the streak camera via a delay unit for the synchronous sweeping electric field, and the other goes to a function generator after being converted to 10 kHz, and the function generator provides a 10 kHz 1 % duty cycle square wave for both the electro-optic modulator (EOM) in the CW pump laser path and the streak camera microchannel plate gate.

The gain mechanism in the TE mode lasing regime is therefore to be identified by monitoring the TM mode reflection spectra. However, this method can only be justified if the TE and TM modes share a common electron and hole reservoir [156]. This is the case under the non-resonant pumping, in which carriers are efficiently scattered between different polarizations as they relax to the energies of long-lifetime exciton states. To confirm this, the TE- and TM-polarized emission spectra in the unetched region of the sample (without the grating mirror) are measured under TE- and TM-polarized pumping. Fig. 2.4(a) and (b) show the emission spectra of the four polarization configurations at pump powers of about 1 μW and $10^4 \mu\text{W}$ respectively. If there is no imbalance due to pump polarization, one would expect the emission spectra of the four configurations to be identical. The spectra show that, firstly, indeed all four configurations yield spectra of similar lineshapes. Secondly, the TE (or TM) emission, indicative of TE (or TM) reservoir population, also has nearly the same intensities, regardless of the pump polarization (comparing lines of different color). The small difference in the intensity is due to a slight difference in the TE and TM pump power. Thirdly, a higher intensity is measured for the TM emission than for the TE emission by about the same ratio, again regardless of the pump polarization and pump power (comparing solid

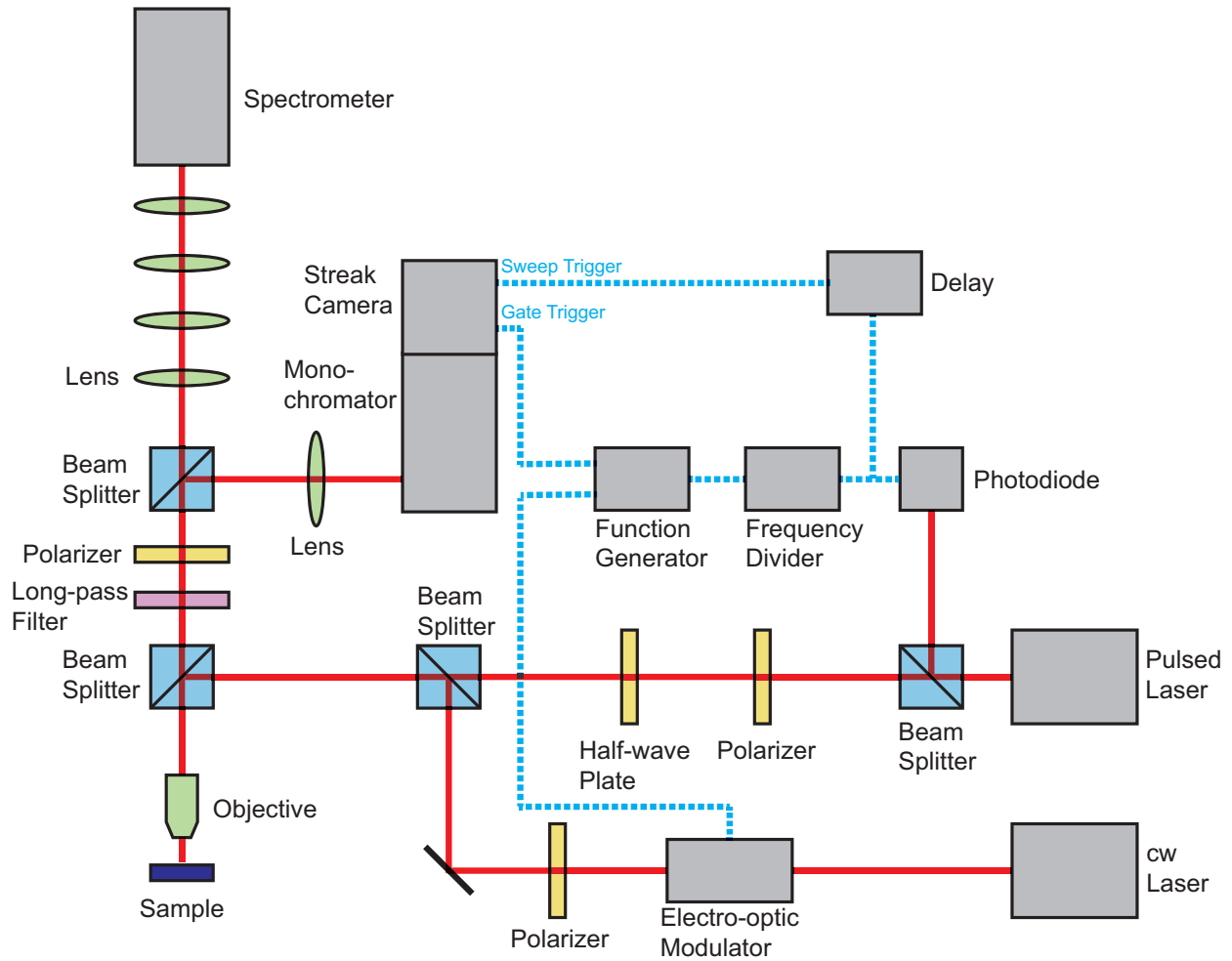


Figure 2.3: **Experiment setup for PL and reflection spectra measurement.** The CW laser is used as the non-resonant excitation and the pulsed laser is used as a resonant probe. The signal from the sample is collected with the spectrometer and the streak camera. Optical chopping is applied to the CW laser with the electro-optic modulator and to the streak camera with its built-in microchannel plate gate. Red solid lines denote optical paths and blue dashed lines denote electric timing signals.

lines with dotted lines). This is due to different collection efficiencies for TE and TM emission in the experimental setup. The ratio between the TE and TM intensities, integrated in photon energy as a function of TE emission intensity is plotted in Fig. 2.4(c). In this way, the preference of a certain emission polarization by the same pump polarization will be manifested as a difference of the ratio under the two pump polarizations at equal TE emission intensity. In the data, for pairs of TE and TM pump powers that produce comparable TE emission intensities, the ratio is very close for each pair, with a difference smaller than 7.3%. There is an overall decreasing trend of this ratio as TE emission intensity increases, possibly because the collection efficiencies also change with photon energy distribution. In summary, The observation of equal TE and TM emission outside the grating independent of the pump polarization confirms efficient scattering between different polarizations.

Moreover, within the grating area, the weakly and strongly coupled modes differ only over a small range of in-plane wave vector $q_{\parallel}$ near $q_{\parallel} = 0$ compared to the large range of $q_{\parallel}$ occupied by all excitons or polaritons. This ensures that the large- $q_{\parallel}$ modes, unmodified by strong coupling, do not have a significant difference of population between TE and TM polarizations, despite very different dispersions and decay rates of the TE and TM modes near $q_{\parallel} = 0$. The large- $q_{\parallel}$ modes account for most of the total carrier population, as the lowest TE state polariton density is estimated to be 3.6×10^{-5} of the total carrier density below threshold and 1.2×10^{-3} even at the highest power used. Hence, the TE and TM modes share the same electron and hole reservoir, and via polarization-resolved spectroscopy, one can access simultaneously the optical properties of the polariton system and its underlying electronic media.

2.4 Photoluminescence Spectra

Two distinct types of lasing transition are measured for the TE-polarized modes in devices with different cavity-exciton detunings. In devices with negative to small positive detunings, polariton lasing is measured with typical features that have been so far identified with those of a polariton BEC. In more blue-detuned devices, clear features of a photon laser are observed. An example of

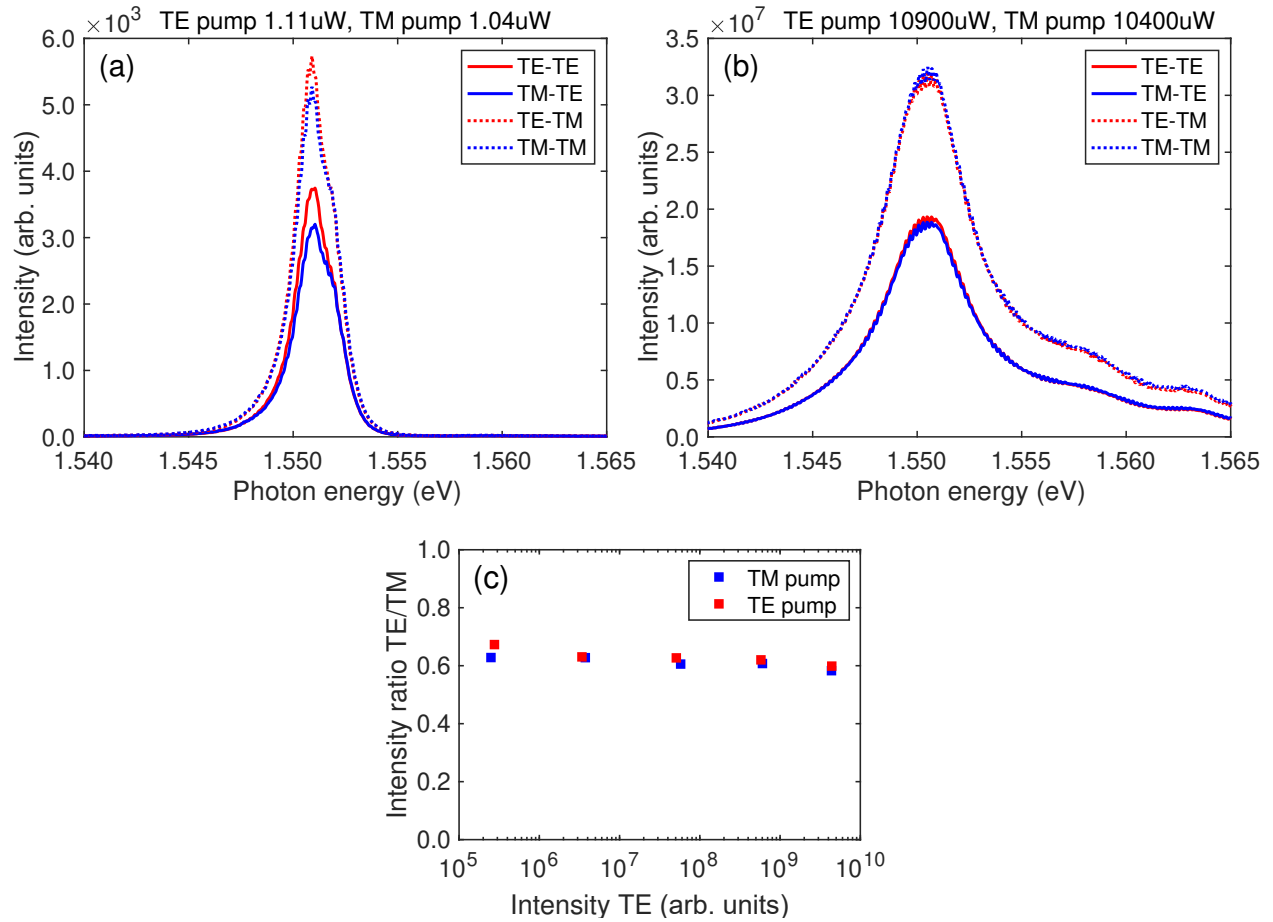


Figure 2.4: **Emission intensities in the unetched region of the sample with different polarization configurations.** (a) and (b) show the emission spectra at different pump powers. The four configurations are: TE pump and TE collection (red solid line), TM pump and TE collection (blue solid line), TE pump and TM collection (red dotted line), TM pump and TM collection (blue dotted line). (c) The ratio between integrated intensities of TE and TM emission as a function of integrated intensity of TE emission. The ratio under TM pump is marked in blue squares while the ratio under TE pump is marked in red squares. (a) and (b) correspond to the pairs of data points in (c) near 3×10^5 and 4×10^9 on horizontal axis respectively.

each type is shown in the top row and bottom row of Fig. 2.5, respectively.

In a polariton laser [top row in Fig. 2.5], the lowest state remains distinct throughout the measured densities, with only slight line broadening below threshold and resolution-limited linewidth above threshold. Its energy blueshifts continuously with power and remains well below the cavity and exciton resonances [Fig. 2.5(a) and (c), top]. These features suggest the lowest state remains a coupled state between electron-hole pair and photon and are the same features as widely reported for polariton BEC in the literature [15, 16, 39].

In stark contrast, in a photon laser [bottom row in Fig. 2.5], the LP modes can no longer be discerned as the pumping density increases [Fig. 2.5(a), bottom middle panel]. A mode with a rather broad linewidth emerges near the cavity resonance energy near the threshold and becomes pinned at the cavity energy above threshold [Fig. 2.5(c), bottom]. These features are fully consistent with the dissociation of bound states before the transition to photon lasing, both of tightly bound excitons and of electron-hole pairs at the many-body level. The result suggests that an electron-hole plasma is formed before a phase transition threshold is reached. This may be because of both a heavier effective mass of the LP that leads to a higher threshold density and also a larger exciton fraction that leads to stronger scattering-induced dephasing.

Similar photon lasing transitions in polariton cavities have also been studied in earlier works [71, 40, 141]. Like in the current observations, the photon lasing transitions are accompanied by significant linewidth broadening, many times more than that of a polariton lasing transition [40, 141], as consistent with the disappearance of a bound state. In some of these works, the photon lasing energy is reported to be below the original cavity resonance when the detuning is within about ± 10 meV, which is attributed to a redshift of the cavity resonance at high carrier densities [71, 141]. However, no quantitative models are given to explain such a shift, and the carrier densities used in the cited studies are likely much higher. The detuning of the polariton and photon lasing devices in this work are similar in magnitude, -2.4 meV and 3.5 meV respectively for the examples shown in Fig. 2.5. The photon lasing observed takes place close to the original cavity resonance despite a higher threshold density than that of polariton lasing. This confirms that there

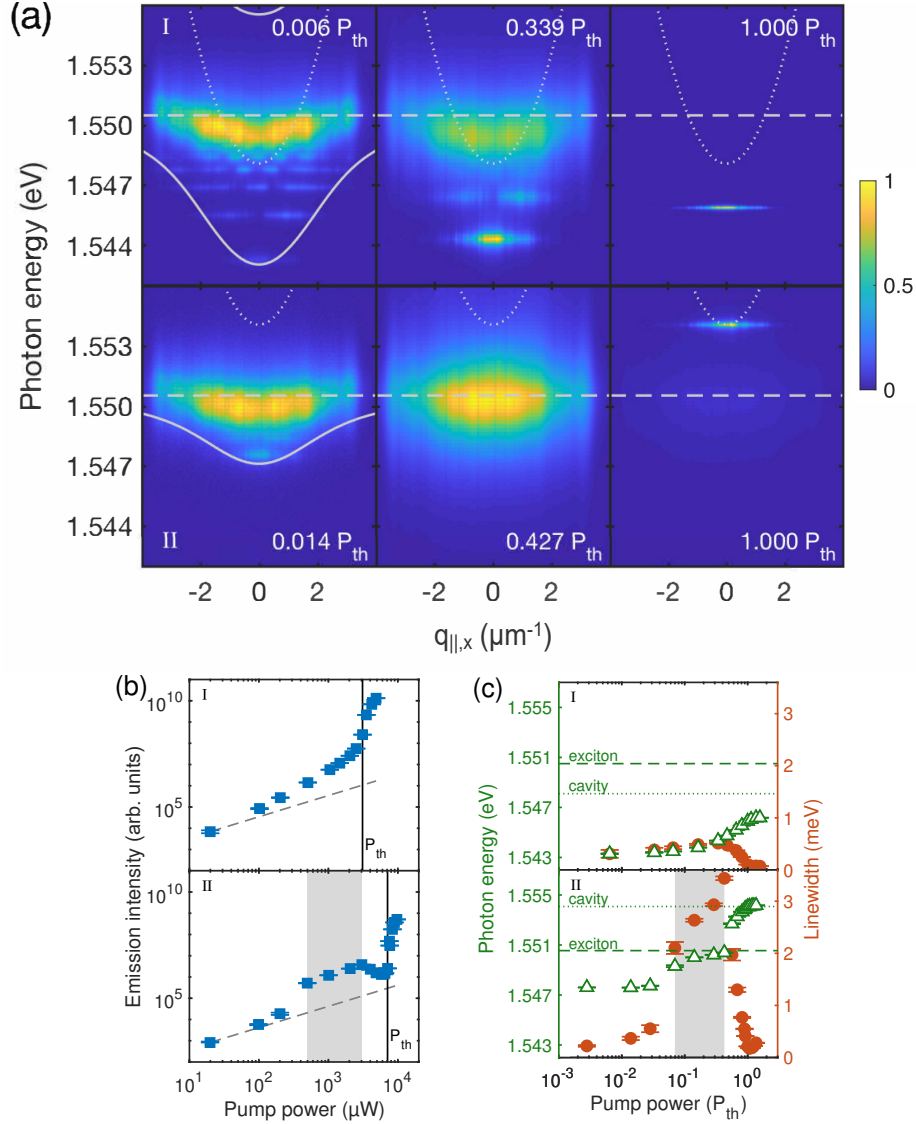


Figure 2.5: Emission properties of a polariton laser and a photon laser. In all subfigures the top row shows the polariton laser with detuning -2.4 meV and the bottom row shows the photon laser with detuning 3.5 meV. (a) Fourier-space spectral images of TE-polarized emission from below to at the lasing threshold at $q_{||,y} \sim 0$ at pump powers as labeled. The top middle panel is at the pump power where the $q_{||} = 0$ mode has the largest linewidth. The bottom middle panel is at the pump power where the $q_{||} = 0$ mode is much weaker than the peak at TM exciton energy and cannot be discerned. The dashed lines mark the exciton resonances measured outside the grating region by reflection without a pump. The dotted (solid) curves are the calculated empty cavity (polariton) dispersions assuming no in-plane confinement; they are based on the measured exciton energy and the LP and UP energies measured from TE reflection without pump [Fig. 2.9]. The intensities are normalized to the maximum value in each spectrum. (b) The emission intensity of the TE $q_{||} = 0$ mode versus the pump power. The dashed line is a reference of linear dependence. (c) Pump power dependence of the emission photon energy (open green squares) and linewidth (filled orange circles) of the TE $q_{||} = 0$ modes. The empty cavity at $q_{||} = 0$ (dotted lines) and exciton (dashed lines) resonances are also shown. In (b) and (c), the gray region corresponds to where the lowest TE state is much weaker than the peak at TM exciton energy and cannot be discerned; in (b), the vertical solid lines mark the lasing threshold.

is negligible shift of the cavity resonance in these devices at the carrier densities used.

Real-space TE-polarized emission spectra of the two example devices in Fig. 2.5 are shown in the top and bottom rows of Fig. 2.6. The $7.5\ \mu\text{m} \times 7.5\ \mu\text{m}$ grating regions are centered at the origin and the spectra are taken at $y \sim 0$. At low pump powers, discrete LP states are visible in both devices inside the grating. With increasing pump power, in the polariton laser shown in the top row of Fig. 2.6, the lowest polariton state retains its spatial profile and a narrow linewidth of a fraction of an meV, clearly separate from the excited states, and remains well below the cavity resonance even above the lasing threshold, suggesting the bound state is retained in the system and evolves continuously. In contrast, in the photon laser shown in the bottom row of Fig. 2.6, the emission from inside the grating region broadens and blueshifts drastically even before reaching the lasing threshold [Fig. 2.6, bottom middle panel], spreading out over 3 meV and therefore indistinguishable from the excited states, characteristic of emission from the electron-hole continuum. Above the threshold power, lasing starts near the cavity resonance. These observations are consistent with the Fourier-space spectra.

Fourier-space TM-polarized emission spectra of the example devices shown in Fig. 2.5 are given in Fig. 2.7. The pump laser powers here are chosen to be close to those in the TE-polarized spectra. For both devices, at low pump laser power, the TM-polarized spectra show only discrete exciton emission. As pump laser power increases, the emission linewidth broadens. However, near TE lasing threshold, the spectra of both devices have an electron-hole continuum-like pattern, with a significant portion of emission at photon energies higher than the exciton energy at low pump power.

2.5 Reflection Spectra

While the observed polariton laser shows spectral features distinct from a photon laser and similar to a polariton BEC, the absorption spectra of the electronic media reveal a many-body phase different from a BEC. As described in Section. 2.2 and Section. 2.3, with the linear polarization selectivity of the cavity, it is possible to probe the electronic gain of the strongly coupled TE mode via the

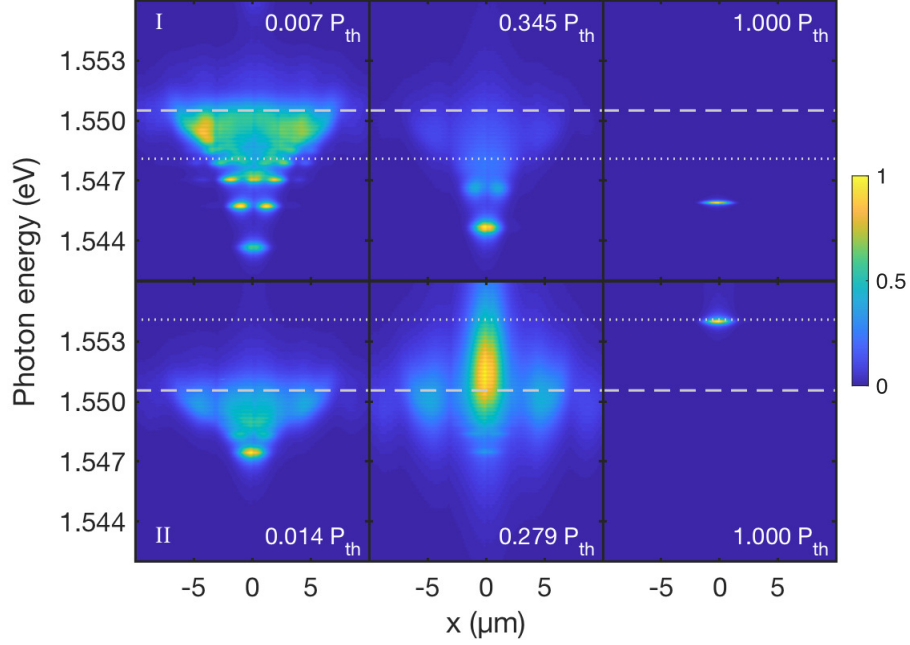


Figure 2.6: **Real-space TE emission spectra of the two types of lasers.** Real-space spectral images of TE-polarized emission of the polariton laser (upper row) and photon laser (lower row) shown in Fig. 2.5. The grating of size $7.5\,\mu\text{m} \times 7.5\,\mu\text{m}$ is centered at the origin and the spectra are taken at $y \sim 0$. The dashed lines mark the exciton resonances measured outside the grating region by reflection without a pump. The dotted lines mark the calculated empty cavity resonance at $q_{\parallel} = 0$. The intensities are normalized to the maximum value in each spectrum. The lowest polariton state goes through a continuous blueshift towards lasing in a polariton laser but is significantly broadened before threshold in a photon laser.

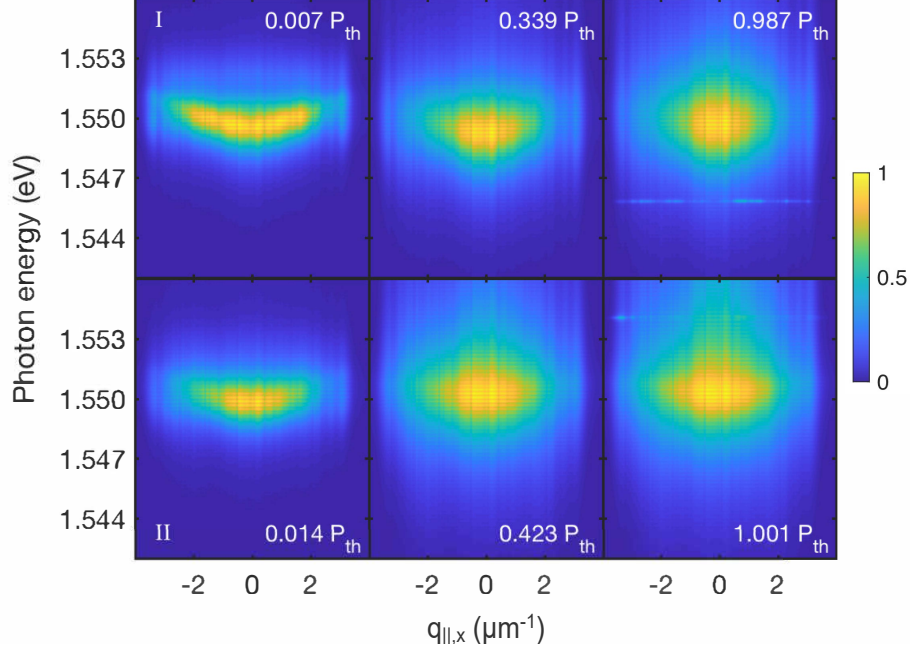


Figure 2.7: **Fourier-space TM emission spectra.** Fourier-space spectral images of TM-polarized emission of the polariton laser (upper row) and photon laser (lower row) shown Fig. 2.5, at $q_{\parallel,y} \sim 0$ and at pump powers close to those shown in Fig. 2.5. The intensities are normalized to the maximum value in each spectrum.

weakly coupled TM mode using time-resolved reflectance of a pulsed probe laser. Examples of the TM reflection spectra of a polariton laser are shown in Fig. 2.8(a) (note that this is not the same one as in Fig. 2.4, which will be discussed in Section. 2.8 and has the same characteristics). Without the pump, absorption is observed at the exciton resonance below the band continuum and above the TE polariton energy. With increasing pump power the exciton absorption becomes stronger. At even higher pump power but still below threshold, the discrete exciton resonance is no longer resolvable, and the spectra have the shape of an absorption edge of the band continuum. This implies that there is strong screening and band gap renormalization to lower energies, and that the carrier density is already near the Mott density. Close to threshold, a peak above unity emerges; its height and width increase with further increase of the pump power [Fig. 2.8(a), inset]. This shows optical gain due to population inversion.

The TE-polarized reflection and emission spectra of the same device, at different pump laser powers and integrated over q , can be found in Fig. 2.9. LP and UP resonances are clearly seen in the

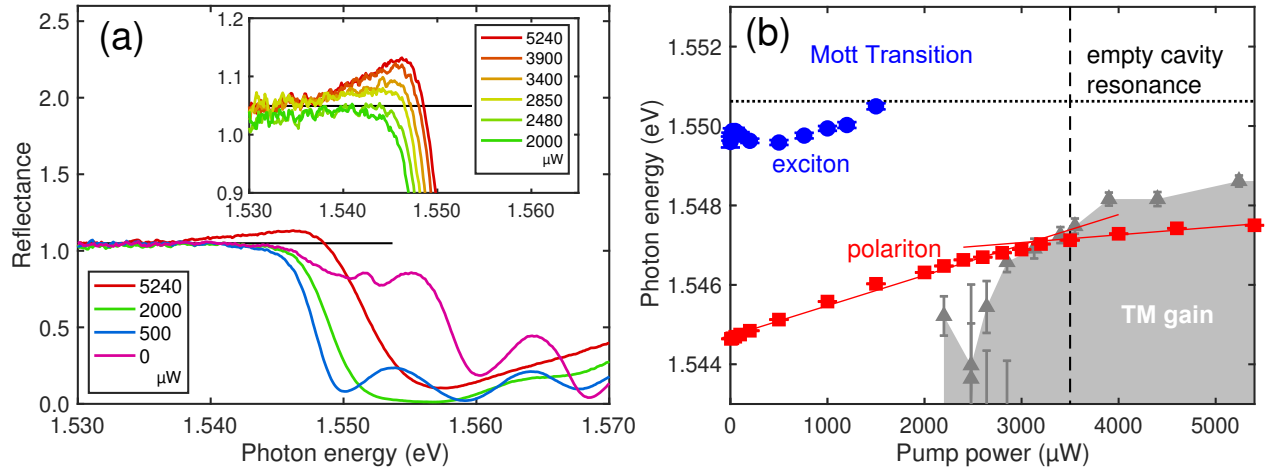


Figure 2.8: **Measured reflection spectra and optical gain of a polariton laser.** (a) TM reflection spectra for a polariton laser at different pump powers. The horizontal black lines mark reflectivity of 1. Inset: enlargement near the gain. (b) Measured pump power dependence of the energetic positions of the TE $q_{\parallel} = 0$ mode (red squares), TM exciton (blue circles), and the spectral bounds of gain (gray triangles). Error bars on the bounds are obtained by dividing the standard deviation of the reference of unity reflectivity by the local slope at the boundary. The empty cavity resonance is marked by the black dotted line. The polariton lasing threshold, marked by the black dashed line, coincides with the onset of the fermionic gain. Red lines are linear fitting to TE polariton energy below and above threshold. The exciton mode, extracted from TM reflection spectra, is no longer resolvable above 1500 μW , due to dissociation of the excitons above the Mott density.

reflection spectra at low pump powers. The multiple discrete LP modes due to the finite in-plane size of the grating are also clearly resolved at low pump powers. As the pump power increases, the emission intensity from the lowest LP state increases sharply, much faster than other modes, signaling lasing. UP modes are no longer visible. At the highest power, the oscillations near the lasing peak in the reflection spectra are artifacts due to the subtraction of the strong lasing emission.

To analyze the reflection data quantitatively and compare with the PL spectra, several relevant energy values are obtained from the spectra. To get the energies of LP and UP modes at zero pump, curve fitting is used for TE reflection spectra with the model,

$$R_{\text{TE}}(E) = b - \sum_i c_i L_i(E, E_{0i}, \Gamma_i), \quad (2.1)$$

$$L_i(E, E_{0i}, \Gamma_i) = \frac{(\Gamma_i/2)^2}{(E - E_{0i})^2 + (\Gamma_i/2)^2}, \quad (2.2)$$

where b , c_i , E_{0i} , and Γ_i are the fitting parameters. The Lorentzian function $L_i(E, E_{0i}, \Gamma_i)$ represents the resonance with energy E_{0i} and FWHM Γ_i . The number of resonances used in the model depends on the number of resonances visible in the spectra. The energy of the lowest resonance in the LP (UP) region is used as the lowest discrete LP (UP) state energy. In all the pump-power-dependent data, the emission energies of the lowest polariton or photon mode are obtained from TE emission spectra by curve fitting with the model

$$I_{\text{TE}}(E) = \sum_i c_i f_i(E, E_{0i}, \Gamma_i), \quad (2.3)$$

where c_i , E_{0i} , and Γ_i are the fitting parameters. Each of the peaks $f_i(E, E_{0i}, \Gamma_i)$ is either Lorentzian

$$f_i = \frac{(\Gamma_i/2)^2}{(E - E_{0i})^2 + (\Gamma_i/2)^2}, \quad (2.4)$$

typically at low pump powers, or Gaussian,

$$f_i = e^{-(E-E_{0i})^2 / \{2[\Gamma_i / (2\sqrt{2\ln 2})]^2\}}, \quad (2.5)$$

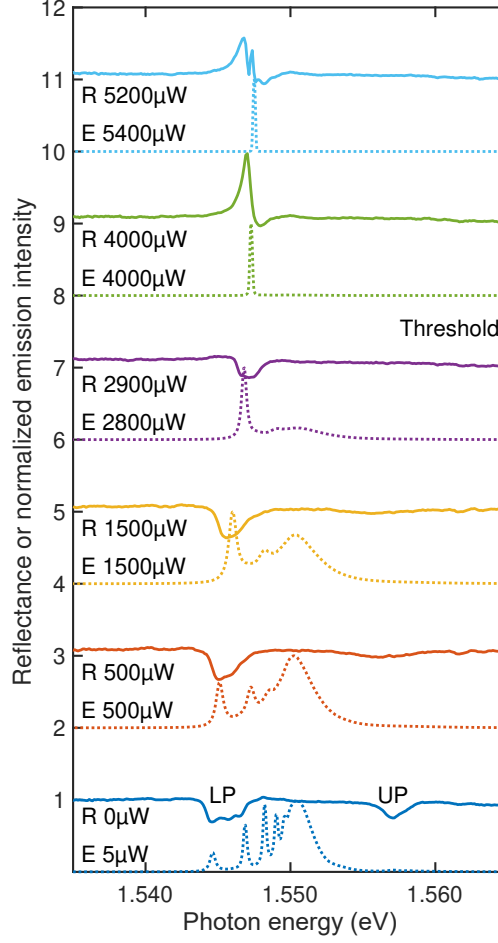


Figure 2.9: **TE reflection and emission spectra of a polariton laser.** The spectra at different pump laser powers are displaced vertically for clarity. Because of the finite in-plane size of the grating, the polariton modes are discrete, shown as multiple peaks or dips in the spectra. The reflection (solid lines) is integrated in both q_x and q_y , where all the modes are detectable; the emission intensity (dashed lines) is only integrated along q_x at $q_y \sim 0$; therefore, only the modes with nonzero intensity along $q_y = 0$ are detectable [120]. For example, at lowest pump laser power ($0 \mu\text{W}$ for reflection and $5 \mu\text{W}$ for emission), the lowest energy peak in both reflection and emission spectra corresponds to the lowest LP state. The second-lowest energy peak in emission spectrum is the second excited state that has a node at $q_x = 0$, and it also shows up in the reflection. In between these two peaks, there is another peak in the reflection spectrum corresponding to the first excited state that has a node at $q_y = 0$. In the reflection spectrum, only one UP mode can be resolved. The UP mode has low emission intensity and appears only as a very small peak in the emission spectrum. All the detected modes match closely in energy between the reflection and emission spectra.

typically at high pump powers, depending on which model returns a smaller mean squared error.

The TM reflection spectra are used to estimate the exciton energy at low pump powers, with the model

$$R_{\text{TM}}(E) = b - \sum_i c_i f_i(E, E_{0i}, \Gamma_i). \quad (2.6)$$

The four peaks $f_i(E, E_{0i}, \Gamma_i)$ used in the fitting are, from high energy to low energy, a broad TM cavity mode, the light-hole exciton, and two heavy-hole exciton peaks possibly due to the inhomogeneity among the QWs. The energy of the lowest peak is used as the exciton energy. The line shapes of each peak are either Lorentzian or Gaussian [Eqs. 2.4 and 2.5] depending on which combination gives the smallest mean squared error. The complete series of data at low pump power in Fig. 2.8(a) are shown in Fig. 2.10, and examples of curve fitting at two different pump powers in Fig. 2.11. At even higher pump powers a redshift of the band gap is expected, and it is observed that the discrete peaks are no longer resolvable [see Fig. 2.8(a)] and therefore this energy estimation is not applied to those pump powers.

To determine the gain bandwidth requires measurement of the absolute reflectivity, which is obtained with a two-step calibration process. First the reflectivity is calibrated by normalizing the reflected pulse laser intensity on the sample to that on a gold mirror mounted in the cryostat next to the sample. There is still error due to imperfect reflection from the gold mirror as well as laser fluctuation and slight changes in the optical path when moving between the sample and the mirror. The error is then estimated and corrected for using a spectral range well below any cavity resonances but still inside the DBR stop band, where the reflectivity should be unity at zero or low pump powers. As shown in Fig. 2.8(a), such a spectral range corresponds to 1.53–1.54 eV, where the reflectivity is close to 1 and varies within about 2.4 %. Hence, the mean of the reflectivity in this spectral range between 0 and 500 μW pump power is used as a reference of unity. In this way the deviation from unity in the reflection spectra is accurately determined.

Gain is identified where a local maximum of the reflectivity between 1.54 and 1.55 eV is above unity as determined from above. The spectral boundaries of the gain are defined as the energies where the reflectivity drops to unity from the local maximum, and the uncertainties of the boundaries

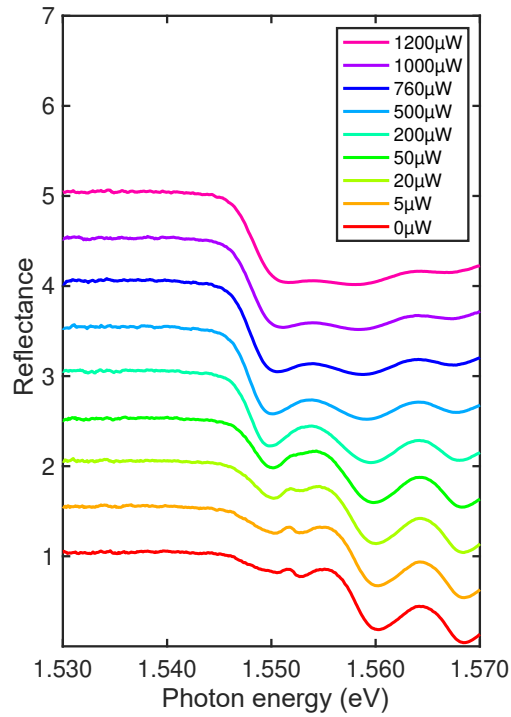


Figure 2.10: **TM-polarized reflection spectra at low pump power.** These spectra are used for the exciton energy estimation. The spectra at different pump laser powers are displaced vertically for clarity.

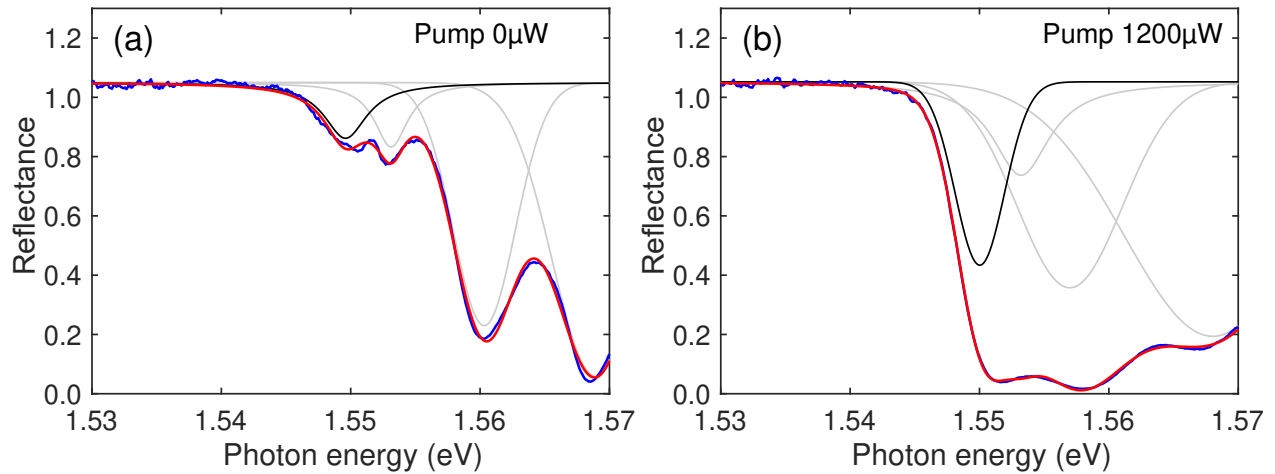


Figure 2.11: **Examples of exciton energy estimation from TM reflection spectra.** (a) pump powers $0 \mu\text{W}$ and (b) $2000 \mu\text{W}$. The blue curves are the experimental spectra and the red curves are the fitted spectra. The black curves are the lowest energy peak used in the fitting, whose centers are taken to be the exciton energies, and the gray curves are the other three peaks described in the text.

are estimated by dividing the standard deviation σ of the reference reflectivity by the local slope of the spectra at the boundaries.

A summary of the experimental data, including the evolution of the resonances and gain with the pump power, is shown in Fig. 2.8(b). The TM exciton resonance blueshifts with increasing pump power but quickly broadens and becomes nonresolvable, corresponding to the Mott transition at moderate pump powers well below threshold. In contrast, the discrete TE resonance continues to exist despite the Mott transition and the presence of gain in TM; it blueshifts continuously with increasing carrier density throughout the pump powers used. Lasing takes place as the energy of the lowest polariton state enters the gain region, and the energy stays within the gain region at higher pump power, showing clearly a lasing transition driven by fermionic population inversion.

2.6 Density Estimation

As a consistency check, the carrier density is estimated, though it should be noted that density estimates in polariton systems typically carry large uncertainties and are therefore not used here for the direct identification of the lasing mechanism. For these calculations, nonradiative processes are ignored.

An experimental pump density is defined based on the absorption of the pump laser and the PL decay time of the TM-polarized emission:

$$\Phi_{\text{pump}} = P \frac{\lambda}{hc} \eta \alpha, \quad (2.7)$$

$$n_{\text{pump}} = \Phi_{\text{pump}} \tau \frac{1}{A} \frac{1}{N_{\text{QW}}}. \quad (2.8)$$

Here Φ_{pump} is the population of carriers excited by the pump in unit time, P is the pump laser power measured before the objective lens, $\eta = 0.7$ is the transmission of the objective lens, α is the total absorption of the pump, hc/λ is the photon energy at the excitation wavelength, and τ is the average lifetime of carriers. A is the diffusion area of the carriers measured from real-space PL [example and pump power dependence given in Fig. 2.12], and N_{QW} is the number of QWs in

the cavity. To determine α , the reflectivity of a TM-polarized laser at the excitation wavelength of 784 nm is measured independently. At this wavelength, the top mirror has low reflectivity while the bottom mirror has very high reflectivity, with negligible transmission through the bottom mirror and thus negligible absorption in the substrate. The materials of the DBR and grating layers also have larger band gaps than the photon energy at this wavelength and their absorption should likewise be negligible. It is assumed that all of the pump laser not reflected from the sample is absorbed by the QWs, i.e. $\alpha = 1 - R$. α and A vary among different devices. For the device shown in Fig. 2.8, $\alpha \sim 0.3$ and $A \sim 32 \mu\text{m}^2$. τ is determined from the decay time of TM emission, measured by time-resolved PL of the TM emission with a pulsed excitation laser centered at the same wavelength as the CW pump laser used in the main measurement. It is found to be around 600 ps for excitation powers 1–300 μW .

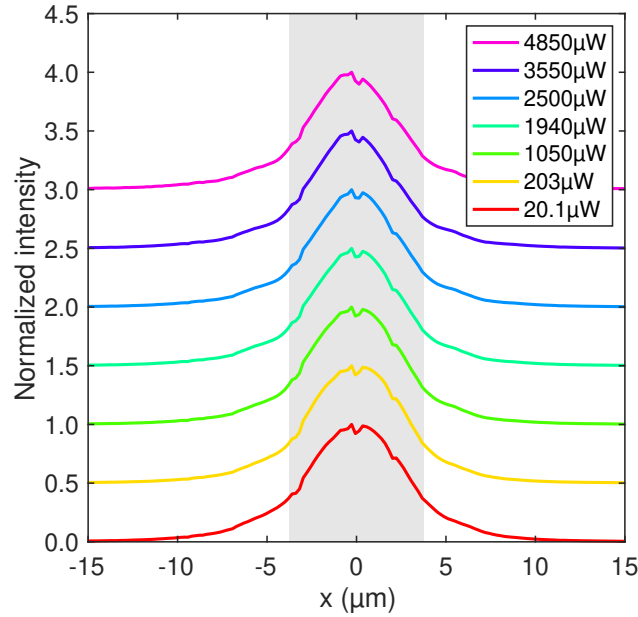


Figure 2.12: **Spatial profile of TM-polarized emission.** Spatial profile of the emission from a grating-based device along x -direction at $y = 0$ in TM polarization. The profiles at different pump laser powers are displaced vertically for clarity. The grating region is shaded in gray. The emission remains mostly inside the grating region within the pump powers used.

Below threshold, in the linear regime, the steady-state carrier density n_{carrier} is assumed equal to the pump density n_{pump} . Above threshold, it is necessary to account for the strong, superlinear emission from the lasing mode. It is assumed that all decay processes other than the laser emission

are still linear and proportional to the carrier density. Therefore, the emission rate of the laser Φ_{laser} is subtracted from the pump to get an estimate of the carrier density:

$$\Phi_{\text{laser}} = N_c \frac{1}{\eta_{\text{col}}} \frac{A_{q_{\text{mode}}}}{A_{q_{\text{col}}}}, \quad (2.9)$$

$$n_{\text{carrier}} = (\Phi_{\text{pump}} - \Phi_{\text{laser}}) \tau \frac{1}{A} \frac{1}{N_{\text{QW}}}. \quad (2.10)$$

Here N_c is the collected photon count from the emission of the lasing mode, $\eta_{\text{col}} = 2.85 \times 10^{-3}$ is the independently calibrated total collection efficiency, and $A_{q_{\text{mode}}}/A_{q_{\text{col}}} \sim 7.5$ is the ratio of the area of the lasing mode to the area of collection in Fourier-space, which also varies among devices.

Polariton density in the lowest energy state is estimated from

$$n_{\text{LP}} = \Phi_{\text{LP}} \tau_{\text{LP}} \frac{1}{A_{\text{LP}}}, \quad (2.11)$$

where Φ_{LP} is the emission rate of the lowest polariton state, calculated similarly to Φ_{laser} but using the emission photon count and the mode area of the lowest polariton state. Above threshold, the lasing state (polariton lasing or photon lasing) is used in this estimation, $\Phi_{\text{LP}} = \Phi_{\text{laser}}$. $\tau_{\text{LP}} = 1.5$ ps is the polariton lifetime and $A_{\text{LP}} \sim 5.3 \mu\text{m}^2$ is the real-space area of the polariton mode.

In this density estimation, quantities P , λ , N_{QW} , η , η_{col} , R , τ , and N_c are directly measured. α is estimated from R . A , $A_{q_{\text{mode}}}$, $A_{q_{\text{col}}}$, and A_{LP} are estimated based on measured one-dimensional spatial profiles of the emission along the x and y directions. τ_{LP} is estimated from the measured emission linewidth. This density estimation aims to provide an upper bound on the density close to the true value.

The total carrier density per QW at threshold is estimated to be $n_{\text{th}} \sim 4 \times 10^{11} \text{ cm}^{-2}$ which is greater than n_{Mott} , consistent with the onset of population inversion and fermionic gain.

2.7 Comparison with Theoretical Model

The experimental observations are summarized as follows. The spontaneous phase transition is evidenced in the characteristic superlinear increase of the emission intensity at zero in-plane wave vector, accompanied by sharp linewidth narrowing [Fig. 2.5(b-c)]. These are common for the formation of all three possible phases in the system. However, distinct from a photon laser, a well-defined single mode across the transition threshold is observed, with an emission linewidth that remains narrow and an emission energy well below the cavity or exciton resonance energies and no sudden energy shifts [Fig. 2.5(a) and Fig. 2.5(c)]. This shows the emission originates from a bound state of electron, hole and photon. Furthermore, the system is distinguished from a BEC-like phase by the measurement of fermionic gain in its electronic reservoir due to population inversion, at a carrier density above the Mott density [Fig. 2.8]. Therefore, these observations suggest the possibility of a polariton BCS-like phase.

The experimental results are compared to the predictions of a theoretical model developed by Prof. Rolf Binder and Prof. Nai-Hang Kwong [reported in Section. V of Ref. [157]]. A brief summary of the model is provided here. The model uses realistic electronic band structure and two-dimensional Coulomb interaction, but extended to include electronic correlations beyond the Hartree-Fock approximation due to screening, while also considering the two linear polarizations and the incoherent pump of the system. It can be expressed as a set of semiconductor Bloch equations for the interband polarization and electron distribution function of the system,

$$i\hbar \frac{\partial}{\partial t} P_\ell(\mathbf{k}) = [\varepsilon_{\text{tot}}(\mathbf{k}) - \hbar\omega_0 - i\gamma] P_\ell(\mathbf{k}) - [1 - 2f(\mathbf{k})] \Omega_{\text{eff}}^\ell(\mathbf{k}) + i\hbar \frac{\partial}{\partial t} P_\ell(\mathbf{k}) \Big|_{\text{corr}}, \quad (2.12)$$

$$\hbar \frac{\partial}{\partial t} f(\mathbf{k}) = \text{Im} [\Omega_{\text{eff}}^x(\mathbf{k})^* P_x(\mathbf{k}) + \Omega_{\text{eff}}^y(\mathbf{k})^* P_y(\mathbf{k})] + \hbar \frac{\partial}{\partial t} f(\mathbf{k}) \Big|_{\text{relax}} + \hbar \frac{\partial}{\partial t} f(\mathbf{k}) \Big|_{\text{pump}}, \quad (2.13)$$

and an equation for the optical field,

$$i\hbar \frac{\partial}{\partial t} E_x = (\hbar\omega_{\text{cav}} - \hbar\omega_0 - i\gamma_{\text{cav}}) E_x - \frac{N_{\text{QW}}}{A} \sum_{\mathbf{k}} a_{\mathbf{k}}^{c*} P_x(\mathbf{k}). \quad (2.14)$$

Here, $\ell = x, y$ labels the polarization, $\varepsilon_{\text{tot}}(\mathbf{k})$ is the interband transition energy including Hartree-Fock self-energy, ω_0 is the oscillation frequency of $P_\ell(\mathbf{k})$ to be solved for, γ is a dephasing constant, $\hbar\omega_{\text{cav}}$ and γ_{cav} are the cavity mode energy and decay rate, N_{QW} is the number of QWs, A is the cross-sectional area of the system, and $a_{\mathbf{k}}^c$ is the coupling coefficient between electron-hole and the cavity mode. And the effective Rabi frequency is

$$\Omega_{\text{eff}}^\ell(\mathbf{k}) = \left(a_{\mathbf{k}}^c E_\ell + \frac{1}{A} \sum_{\mathbf{k}'} V_{\mathbf{k}-\mathbf{k}'}^c P_\ell(\mathbf{k}') \right), \quad (2.15)$$

where $V_{\mathbf{k}-\mathbf{k}'}^c$ is the unscreened two-dimension Coulomb potential. Note that the screening effects in the plasmon-pole approximation [1] are included in the term $i\hbar \frac{\partial}{\partial t} P_\ell(\mathbf{k})|_{\text{corr}}$, while the intraband relaxation, the nonradiative decay, and a pump term are included as $\hbar \frac{\partial}{\partial t} f(\mathbf{k})|_{\text{relax}} + \hbar \frac{\partial}{\partial t} f(\mathbf{k})|_{\text{pump}}$. From the solution, the emission energy is taken to be the energy of maximum intensity of the TE field $|E_x(\omega)|^2$, and the spectral location of the gain is obtained by adding a probe pulse and computing the susceptibility.

The predictions of the various energies, including the TE emission and TM exciton at low pump power and the gain region at high pump power, are shown in Fig. 2.13 alongside the experiment results duplicated from Fig. 2.8(b). The model has a good agreement with the main features of the experimental observations: The TM excitons blueshift and then dissociate as the band gap renormalizes to lower energies. The TE emission has a continuous blueshift. At higher pump powers, the range of optical gain includes the TE emission energy. Note that the TE emission energy is well below what is theoretically expected from a photon laser, which can be obtained by omitting the correlation terms and keeping only the Hartree-Fock self-energy. It is instead more similar to the ideal polaritonic BCS solution obtained by leaving out the correlation terms and the pump and decay terms.

The theoretical quantities not measured experimentally, namely the distribution function, the interband polarization, and the pair-breaking energy gap, are given in Fig. 2.14. The interband polarization of the polariton laser has a broad distribution in wave vector indicating bound states,

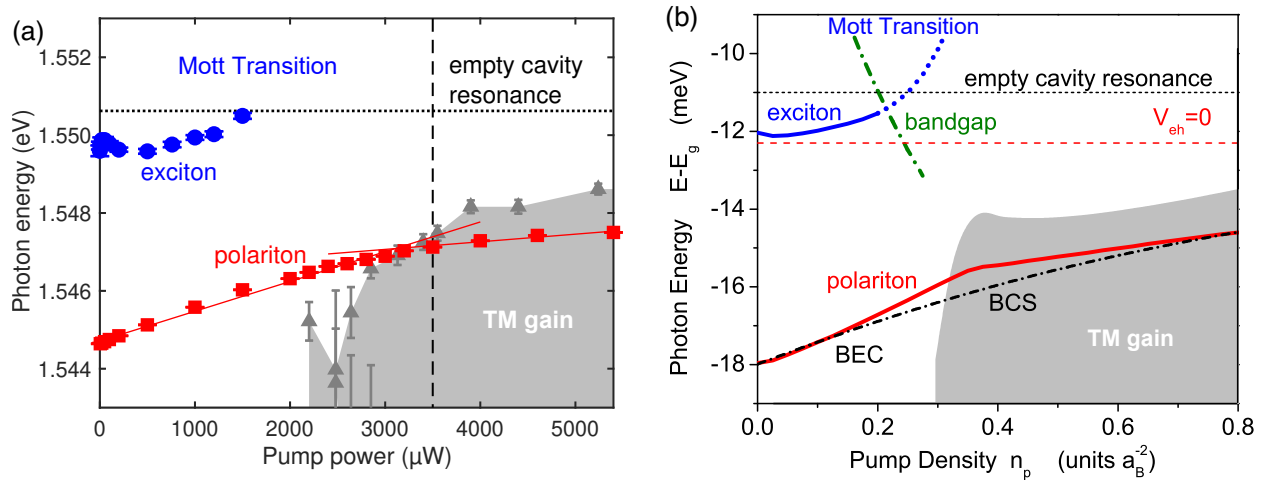


Figure 2.13: **Measured and calculated gain and energetic positions of a polariton laser.** (a) Same as Fig. 2.8(b), for comparison. (b) Calculated pump density (n_p) dependence of the energetic positions of the lowest TE state (red solid line), TM exciton (blue solid line), gain region (gray area), ideal polariton BEC/BCS quasichemical potential assuming density $n = n_p$ (black dash-dotted line), the TE emission without $e-h$ interaction corresponding to an ideal photon laser (red dashed line), renormalized band gap (green dash-dotted line), and empty cavity resonance (black dotted line). $a_B = 14$ nm and E_g are the exciton Bohr radius and QW band gap energy, respectively. Effective electron temperature $T[\text{K}] = 40 + 50n_p a_B^2$. This subfigure is reprinted from Ref. [157].

unlike the narrow peak shape of a photon laser. The upper bound of the interband polarization is $1/\sqrt{2}$ because there is strong field only in one of the linear polarizations. The distribution function is lower than the photon laser case and does not have kinetic hole burning. For both the distribution function and the interband polarization, the polariton laser resembles the ideal BCS solution and differs from the photon laser. However, the amplitude of the interband polarization and the energy gap size are expected to be reduced compared to the ideal BCS case due to cavity dissipation and dephasing. The good agreement in the observable quantities between the experiment results and the theoretical model predictions and the similarities in the microscopic details between the full solution of the model and the ideal BCS solution suggest that the experimentally observed polariton laser may have a BCS-like mechanism.

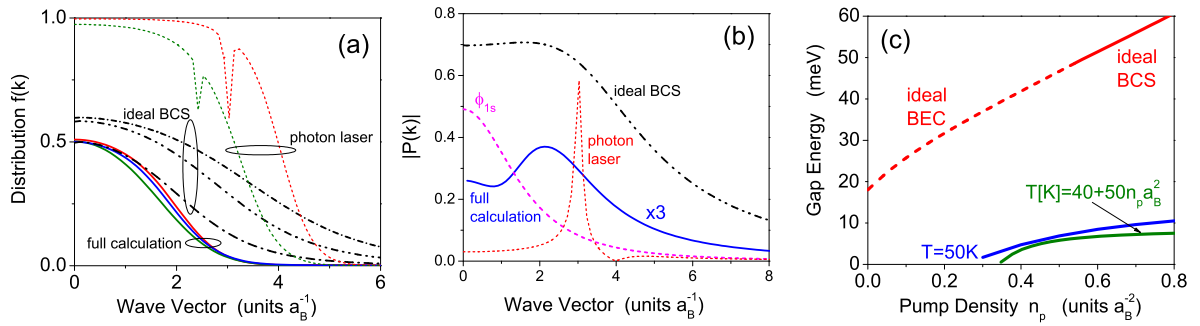


Figure 2.14: Calculated carrier distributions and wave functions. (a) Distribution functions at $T = 50$ K. Solid lines: full calculation at pump densities $n_p a_B^2 = 0.3, 0.45$ and 0.6 (threshold pump density $n_p^{\text{th}} \approx 0.3 a_B^{-2}$), $\gamma = 1.5$ meV. Short dashed lines: without e - h interaction at $n_p a_B^2 = 1.6$ (green) and 2.6 (red), $n_p^{\text{th}} \approx 1 a_B^{-2}$, $\gamma = 0.15$ meV. Black lines: ideal polariton BCS at densities $n a_B^2 = 0.6$ (long dash-dotted line), 1.2 (dash-dot-dot line) and 1.8 (short dash-dotted line), showing that at large densities $f_{\text{BCS}}(k = 0)$ saturates at about 0.6 . (b) Corresponding magnitudes of polarizations (effective wave functions); same line styles as in (a). The dashed magenta line shows the $1s$ exciton wave function normalized to a density of $4 \times 10^{10} \text{ cm}^{-2}$ (c) Pair gap energy of ideal system (red line) and phenomenological estimate of the pair gap energy of the experimental system (green and blue lines). This figure is reprinted from Ref. [157].

2.8 Detuning Dependence

Devices with different grating parameters and at different positions on the sample can have different cavity-exciton detuning and normal mode splitting. On the sample from which data are taken for Section. 2.4 there are a series of devices with different parameters. At negative or small positive detunings, the emission has features of the BCS-like polariton laser described in Section. 2.4, namely a continuous blueshift to a lasing energy that is significantly lower than the empty cavity resonance, with narrow linewidth throughout. At large positive detunings (and also smaller normal mode splitting), however, the lasing transition resembles a conventional photon laser, with a large blueshift towards an energy close to the empty cavity resonance, and large linewidth broadening before threshold.

Fig. 2.15(a) shows the photon energy difference between the empty cavity resonance and the laser emission at threshold pump power. At large positive detunings, the lasing energy at threshold is close to the empty cavity. The linewidth of the emission, both at low pump power, where the system is in the low density regime with bosonic exciton polaritons, and at the pump power before threshold, where the linewidth sees maximum broadening, are also compared to the amount of blueshift from low pump power to threshold pump power. For the BCS-like polariton laser, the linewidth remains close to that of low pump power, and only has small broadening compared to the amount of blueshift, whereas for the devices similar to a conventional photon laser, the broadening is over one order of magnitude, and is comparable to the amount of blueshift. The latter can be considered as a discontinuous jump of the energy from the polariton energy to the cavity energy. The lasers at larger positive detuning have higher estimated threshold density [Fig. 2.15(b)].

From reflectance measurements it can be confirmed that all these devices measured have optical gain throughout lasing and therefore the lasing is due to the population inversion of the fermionic particles [Fig. 2.16]. For this sample, because it is closer to a broad cavity resonance, there is a non-zero slope in the reflectance in the range of wavelengths used as a reference to determine the gain. In this case a linear fit applied to the slope at low pump power is used as a reference, rather

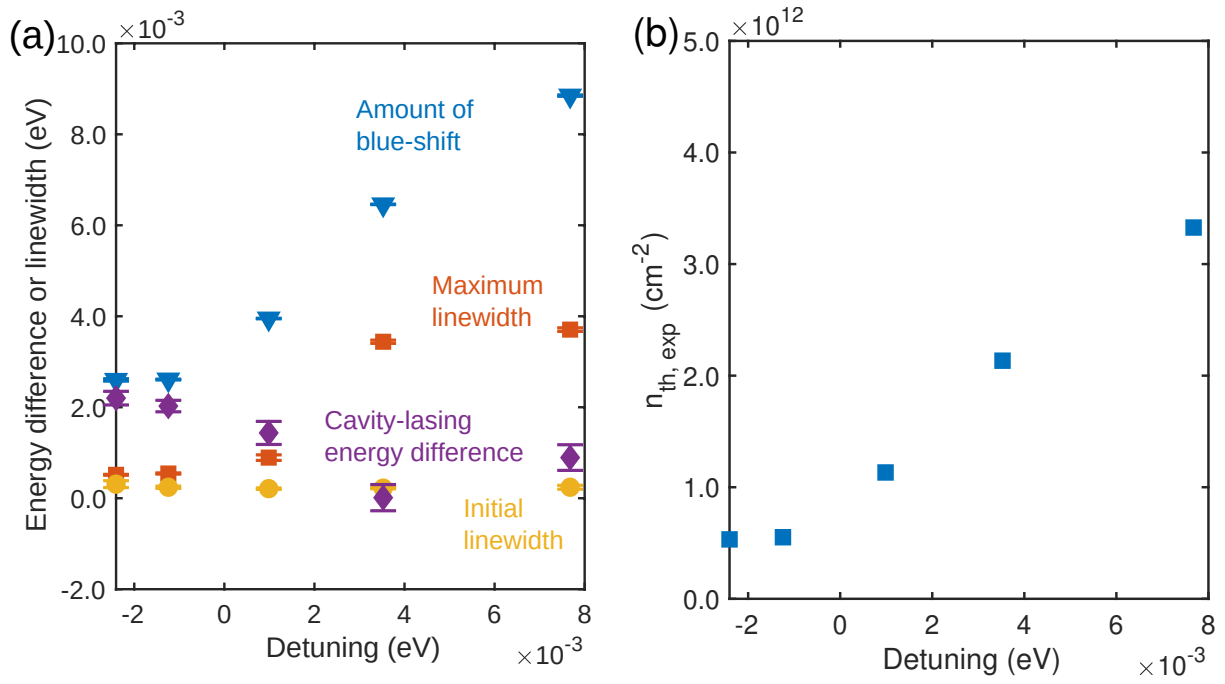


Figure 2.15: **Emission features of devices with different parameters and estimated threshold density.** (a) The photon energy difference between the empty cavity resonance and the laser emission at threshold pump power (purple diamonds), the linewidth of the laser emission at low pump power (yellow circles), and the maximum linewidth before threshold (orange squares), compared to the amount of blueshift from low pump power to threshold pump power (blue downward-pointing triangles). (b) Estimated threshold density.

than the mean, [Fig. 2.16(a)].

While possible BCS-like polariton lasing and photon lasing are observed in the devices, a transition to BEC is not observed, possibly due to limited polariton lifetime. The transition from the BCS regime to photon lasing is expected with increasing excitation. However, presently the pump power can only be increased to about twice the threshold before sample deformation takes place. Within twice the threshold, a large shift of the lasing mode is not observed, which suggests the system remains in the same phase.

2.9 Comparison with Planar DBR Cavity

Although the grating-based cavities make it possible to measure the reflection spectra of the excitons for a polariton laser, some of their features modify the polariton properties. For example, the in-plane confinement can change the density of states and can increase the temporal coherence of the laser [27, 75]. There are other features limiting the experimental reach of parameters. For example, due to the large variations in fabrication, the available detunings are often opportunistic. Furthermore, the limited size of each device and the spacing between the devices necessary to maintain suspension of the gratings also rule out continuous changes of the cavity-exciton detuning. Measurements under higher pump powers are also not possible due to the mechanical properties of the gratings. To find out if the BCS-like polariton lasing state measured in the grating-based cavities can be generalized to more common systems and to observe possible crossovers to other related phases, a planar microcavity with DBR as both reflectors is measured here.

The structure of the DBR cavity sample is shown in Fig. 2.17(a). It has a $3\lambda/2$ AlAs cavity with 9 GaAs QWs, each of 20 nm thickness. The QWs are arranged in three groups at the three anti-nodes of the electric field distribution of the cavity resonance. For each group the three QWs are separated by 4 nm AlAs layers. The DBRs are made of $\text{Al}_{0.2}\text{Ga}_{0.8}\text{As}$ /AlAs quarter wave layers, 33 pairs of layers between the GaAs substrate and the cavity and 28 pairs on top of the cavity. Two of the $\text{Al}_{0.2}\text{Ga}_{0.8}\text{As}$ layers have C and Si doping for future electric field tuning experiments. For the lower DBR, a 3 nm GaAs layer is added between the $\text{Al}_{0.2}\text{Ga}_{0.8}\text{As}$ and AlAs layers of each pair

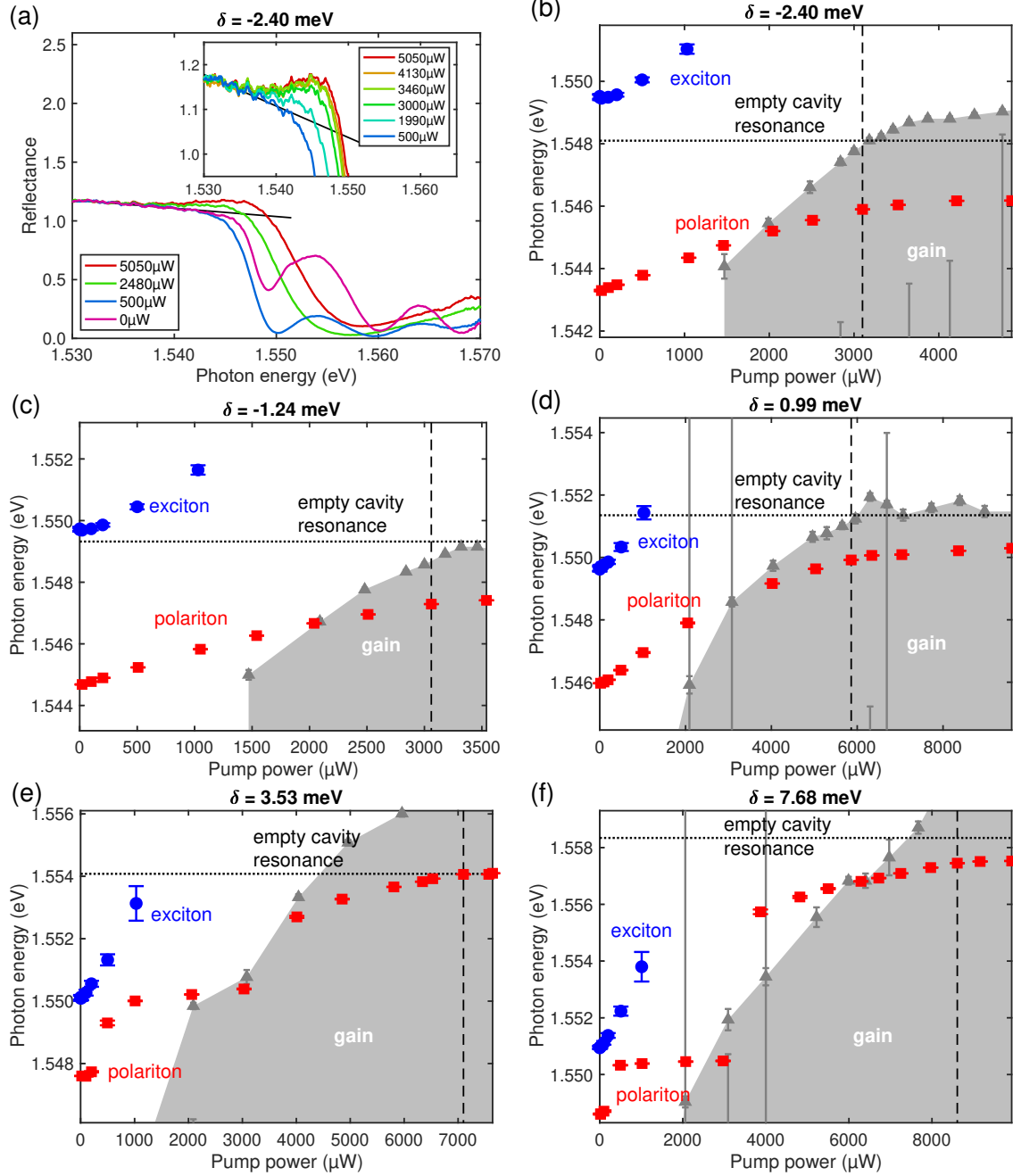


Figure 2.16: Reflectance and gain of devices with different parameters. (a) TM reflectance spectra of the BCS-like polariton laser presented in Section. 2.4, at different pump powers. The reference level of unity is marked with tilted black lines. Inset: near the peak of gain. (b-f) Pump power dependence of measured energetic positions of devices at different detunings, similar to Fig. 2.8(b): TE polariton emission energy (red squares) and TM exciton absorption energy (blue circles) as a function of pump power, as well as the empty cavity resonance (black dotted lines). The spectral bounds of gain are marked by the dark gray triangles. Error bars on the bounds are obtained by dividing the standard deviation of the reference of unity reflectance by the local slope at the boundary. Vertical dashed lines mark the threshold pump power. (b) and (e) are from the polariton and photon lasing devices shown in Section. 2.4, respectively.

to improve the quality of the epitaxial growth. The simulated reflection spectrum of the sample [Fig. 2.17(b)] shows a DBR stop band of about 85 nm and a cavity resonance with $Q > 10^5$. The epitaxial layers have a tapered thickness from the center of the wafer to the edge of it, thereby providing a continuous change of cavity-exciton detuning. The 20 nm QW thickness is larger than the 12 nm thickness in the grating cavity sample and the 7 nm thickness in other widely-used high-Q samples for polariton studies [139], allowing for a larger exciton Bohr radius. The fewer QWs also imply lower carrier density per QW at the same polariton density. Both factors should contribute to stronger phase space filling and screening effects at the same polariton density.

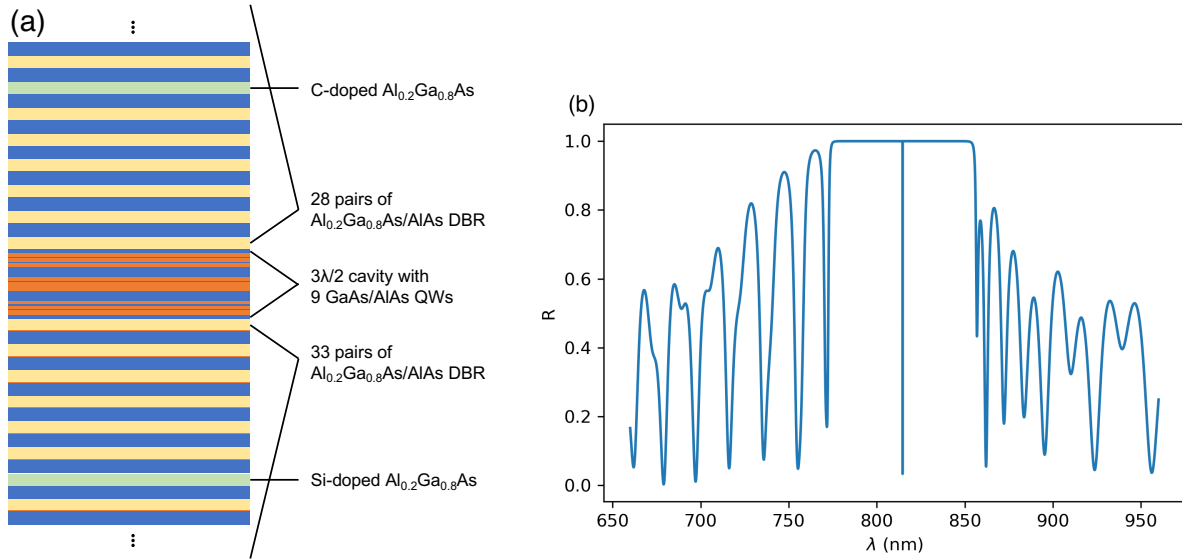


Figure 2.17: Structure and reflection spectrum of the DBR microcavity. (a) Schematic of the structure of the DBR microcavity. (b) Calculated reflection spectrum showing a cavity resonance near 810 nm.

In PL measurements with low excitation power, three polariton branches can be observed for a large range of detuning. With very large positive or negative detuning, the uncoupled exciton states are also visible, and the energy of the heavy-hole exciton (HHX) and the light-hole exciton (LHX) are near 1525 meV and 1529 meV. Fig. 2.18(a) shows an example of the PL spectra and Fig. 2.18(c) shows the estimated energies of the different modes of the system. To determine the exact coupling strength and detuning between the cavity and the excitons, the energy-wave vector dispersion of the LP is extracted from the PL spectra and then compared to the lowest eigenvalue

E_{LP} of the matrix

$$\begin{bmatrix} E_1 & 0 & \Omega_1 \\ 0 & E_2 & \Omega_2 \\ \Omega_1 & \Omega_2 & E_c \end{bmatrix}. \quad (2.16)$$

In this matrix, E_1 (E_2) is the energy of HHX (LHX). $E_c = \frac{\hbar^2 k_{\parallel}^2}{2m_{\text{cav}}[E_c(k_{\parallel}=0)/E_1]}$ is the energy of the cavity resonance, where m_{cav} is an effective mass. Ω_1 (Ω_2) is the coupling strength between cavity and HHX (LHX). Least squares fitting is applied to $E_{LP}(k_{\parallel})$ of spectra measured at different detunings, with $E_{1,2}$, $\Omega_{1,2}$, m_{cav} , and each $E_c(k_{\parallel} = 0)$ as fitting parameters [Fig. 2.18(d)]. The measured exciton energies and the estimated values of the other parameters are used as initial values for the fitting after adding random variations. The fitting result is consistent for the normal mode splitting between HHX and cavity $2\Omega_1 = 8.46$ meV, and the cavity effective mass $m_{\text{cav}} = 3.4 \times 10^{-35}$ kg. In the remaining part of this section, detuning will refer to the energy difference between the cavity and HHX at zero wave vector. Notably, under pulsed excitation with very high pump power, the dispersion of the PL emission overlaps well with the estimated cavity resonance [Fig. 2.18(b)]. However, the following results will be focused on continuous wave excitation, the same as used with the grating cavities.

With a non-resonant pump laser at the wavelength of the first reflection minimum above the DBR stop band, focused to a spot of about 3 μm diameter on the surface of the sample, there are laser-like transitions as the pump power increases. Two types of these transitions are observed depending on the cavity-exciton detuning. For near zero and positive detuning, while the LP dispersion remains visible up to threshold power, near the threshold emission appears following the cavity dispersion which then lases at the threshold [Fig. 2.19(a)]. The energy of the emission intensity peak abruptly changes from the polariton energy to the cavity energy. This behavior has been reported in the literature and categorized as a transition to the weak-coupling regime and an onset of conventional photon lasing (e.g. Refs. [144, 146]). For large negative detuning, lasing happens on the polariton branch but at non-zero wave vector [Fig. 2.19(b)]. This is also widely reported in the literature and sometimes named a kinetic condensate [158, 159, 160, 44, 43, 161, 145, 162]. For this, polaritons are created at the position of the pump, where the high density of large-wave-vector reservoir

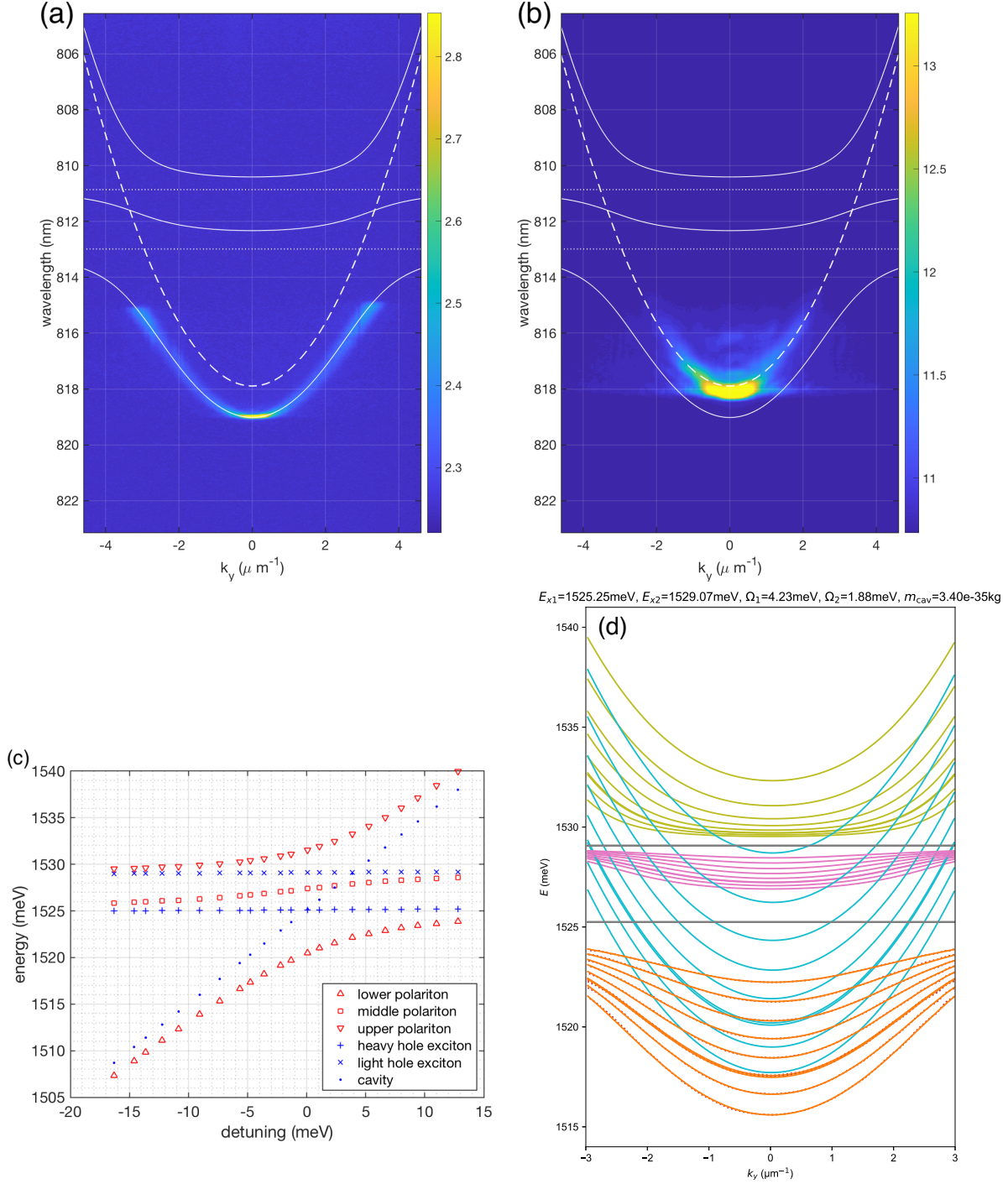


Figure 2.18: **Estimating the coupling strength and detuning of the DBR sample.** (a) PL spectra under pulsed pump at low pump power (297 μW) with the color showing intensity in log scale. (b) PL spectra at high pump power (359 mW) at the same position on the sample as (a). In (a) and (b) the estimated exciton, cavity, and polariton energies are marked with dotted, dashed, and solid lines respectively. (c) The estimated exciton, cavity, and polariton energies at zero wave vector as a function of detuning. (d) Least squares fitting of the LP dispersion. The gray and cyan solid lines are the estimated exciton and cavity energies. The orange, pink, and yellow solid lines are the estimated polariton energies, and the orange dotted lines are the measured LP energies. The fitting parameters are given in the subfigure title.

excitons provides the polaritons with an effective potential energy via repulsive interaction. There is a conversion of that potential energy to kinetic energy while the polaritons propagate away from the pumping spot, resulting in emission at a higher energy and momentum.

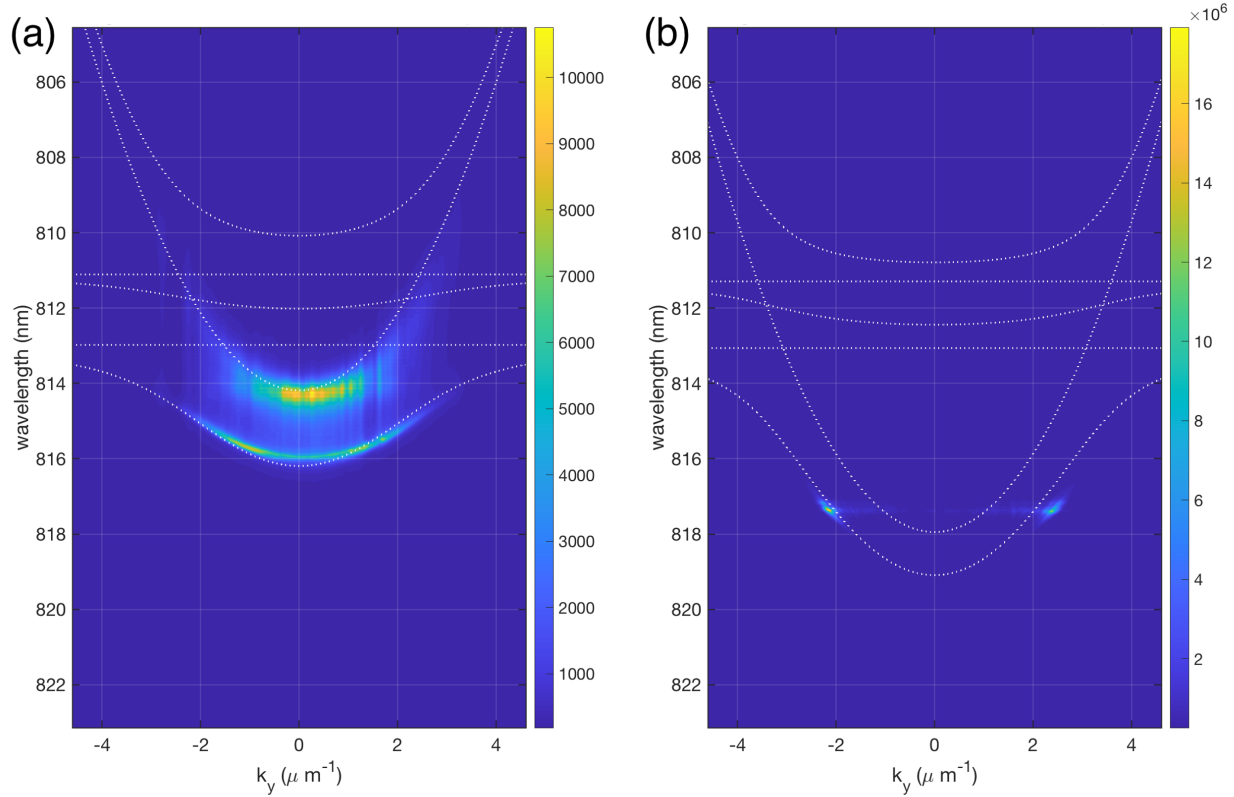


Figure 2.19: **PL spectra under CW pump with a 3 μm spot.** (a) At about -2.2 meV detuning and 24.8 mW pump power, emission appears following the cavity dispersion, with the LP still visible. (b) At about -9.2 meV detuning and 50.1 mW pump power, lasing occurs at non-zero wave vector on the polariton dispersion. In (a) and (b) the white dotted lines mark the estimated energies at low pump power, at $k_y = 0$ from low to high energy: LP, cavity, HHX, MP, LHX, UP.

The results are very different if a ring-shaped pump profile is used instead of a tightly-focused spot. Non-resonant ring-shaped pumps have been widely used as optical traps for polaritons because of the potential barrier formed along the ring by the repulsion from reservoir excitons created there [163, 164, 55, 139, 53, 133, 140, 165]. At the same time, because the propagation distance of the excitons is shorter compared to small-wave-vector polaritons, the interaction with the reservoir excitons is minimized for polaritons that flow to the center of the ring. In the following experiments,

a ring-shaped pump profile of $30\text{ }\mu\text{m}$ diameter is created with an axicon in the path of the pump laser [Fig. 2.20]. A spatial filter is used in the collection path to collect only the emission from within the ring. Laser-like transitions are again observed for a wide range of detunings, between at least -8 meV and 4 meV . At high pump powers for all these detunings, the lasing emission is from a single $k_{\parallel} = 0$ mode with a narrow linewidth below the resolution limit of the spectrometer. Outside this range, the lasing threshold is not reached with the available pump power for large positive detunings, or lasing happens in modes with multi-peak k -space distributions or in multiple modes for large negative detunings. However, within the detuning range of the single-mode $k_{\parallel} = 0$ lasing, the spectral features also depend on the detuning below the threshold. In general, for near zero and positive detuning, as shown in Fig. 2.21(a-c), the linewidth is significantly broadened, resembling a transition to the weak-coupling regime [40, 141]. For negative detuning, as shown in Fig. 2.21(d-f), the linewidth remains narrow. This qualitative difference is similar to what is observed for the grating-based cavities like those in Fig. 2.5 and Fig. 2.6.

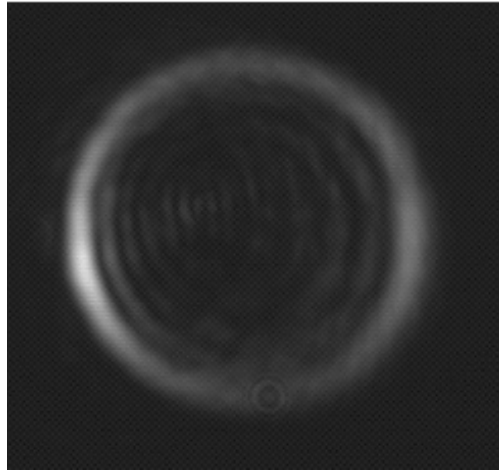


Figure 2.20: **Image of the ring-shaped pump on the sample.**

A summary of the detuning and pump power dependence data is given in Fig. 2.22, including the emission intensity, energy, and linewidth of the lowest state. The lasing transition shows up as a sharp increase in emission intensity and an abrupt narrowing of linewidth. Due to fluctuations in the pump power and spatial profile, or possibly to instabilities of the polariton system itself, at some pump powers the lasing state is not stable and the system switches between the lasing and

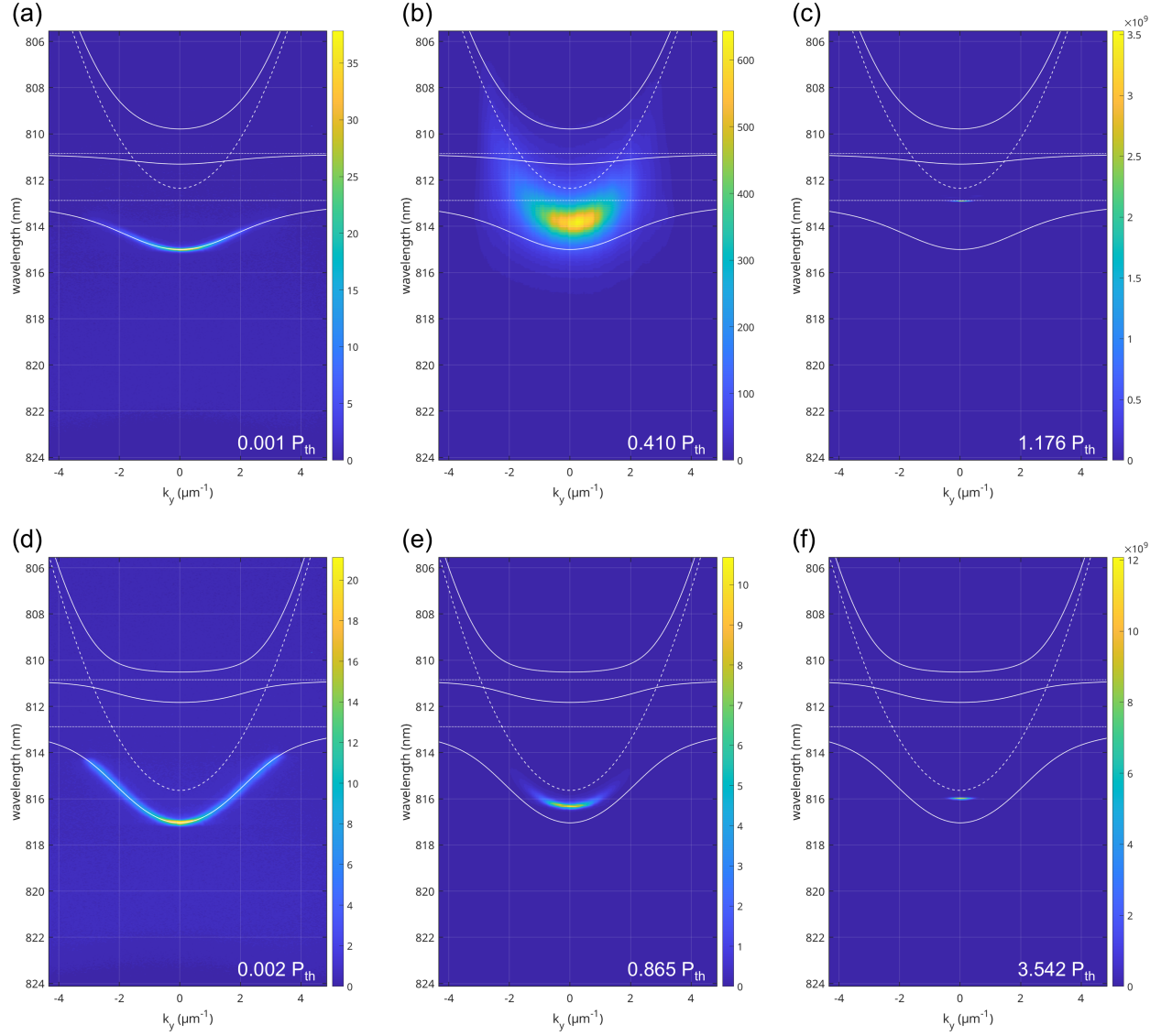


Figure 2.21: **PL spectra under CW pump showing two types of lasing transition.** (a-c) At 0.99 meV detuning, the emission peak becomes significantly broadened before threshold. (d-f) At -5.16 meV detuning, the emission peak remains narrow in linewidth. In all subfigures the pump power is labeled in units of threshold power (P_{th}). The white dotted, dashed, and solid lines mark the estimated energies at low pump power, of excitons, cavity, and polaritons respectively.

non-lasing state alternatively with a timescale of about 0.1 s. In this case multiple spectra are taken at each of these pump powers and then the threshold is defined as the minimum power where the system no longer switches back to the non-lasing state. Fig. 2.22(a) shows that lasing occurs at increasing pump power with increasing detuning. Fig. 2.22(b) shows the energy of the lowest state emission, which continuously blueshifts both before and after the threshold for all the detunings. The linewidth, shown in Fig. 2.22(c), remains very small for the negative detuning, indicating a well-defined bound state, while going through large broadening before threshold for near zero and positive detunings, indicating the loss of the bound state. There is not, however, a clear boundary between the two types of behavior. Fig. 2.22(d) summarizes various energies and linewidths as a function of detuning. The amount of blueshift up to threshold power increases with detuning, and the maximum linewidth follows the same trend. However, if the maximum linewidth is compared to the polariton linewidth measured at very low pump power, which does not change significantly with detuning, it is found that the maximum broadening is similar to the low power linewidth for negative detunings only and increases to more than one order of magnitude larger than the low power linewidth for positive detunings. These features are again similar to the grating-based cavities [compare Fig. 2.15(a)]. Yet, the energy difference between the cavity and the emission at threshold does not agree as well with results from the grating-based cavity. Although there is a decreasing trend with increasing detuning, the energy difference does not reach near zero even at the largest positive detuning but remains a finite positive value. A second threshold is not observed within the available range pump power, which is up to 3.5 times the threshold power for the negative detunings.

The PL spectral features suggest that the sample in the ring-shaped pumping configuration may have lasing mechanisms similar to that of the grating-based cavities. However, the absorption spectra of the underlying carriers is not directly observable, and therefore it is still unknown if there is population inversion while the system is lasing. Another relevant quantity that can be measured now, due to the increased size of the system, is the effective mass of the polariton dispersion. It indicates the photon fraction of the polariton state [Eq. 1.5] and is expected to drop to the bare

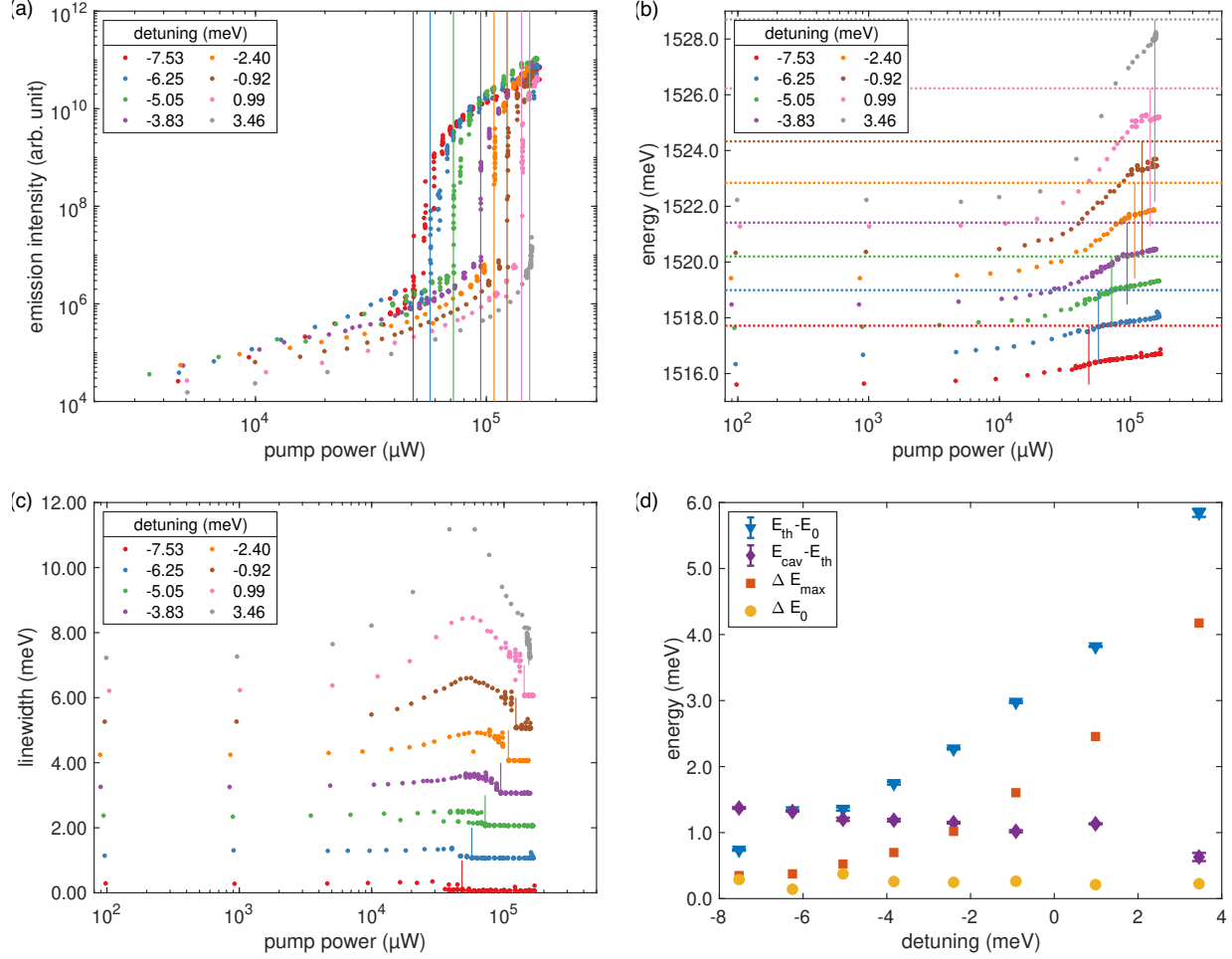


Figure 2.22: Summary of the detuning dependence of the PL spectra features. (a) Emission intensity, integrated within $|k_y| < 0.1 \mu\text{m}^{-1}$ in wave vector and twice the linewidth in energy, in log scale. (b) Emission energy at zero wave vector. (c) Emission linewidth at zero wave vector. The curves have a vertical offset of 1 meV after each detuning. In (a-c) the threshold pump power of each detuning (P_{th}) is labeled with a vertical line in the same color as the data points. (d) The amount of blueshift at P_{th} (blue triangles), the energy difference between cavity and emission peak at threshold (purple diamonds), the maximum linewidth (orange squares), and the linewidth at the low pump power limit (yellow circles) as functions of detuning. The data points of energies at P_{th} represent the mean of the corresponding values within 4 mW range about P_{th} and the error bars represent the standard deviation.

cavity value if the system becomes weakly-coupled. Excitons in the weak-coupling regime in the current system are not expected to reach a BEC, and therefore the lasing in the weak-coupling case should require population inversion. The energy-wave vector dispersion of the LP, or other possible modes it subsequently evolves into as the pump power increases, can be extracted from the measured spectra currently up to threshold power. Above threshold the emission intensity at $k_{\parallel}$ is too strong, and the dispersion would require special experiment techniques to measure [135, 165]. The dispersion curves within $|k_{\parallel}| < 1.5 \mu\text{m}^{-1}$ are then fit to a quadratic function to determine the effective mass and the result is shown in Fig. 2.23. For the most negative detuning, the effective mass remains about the same until the pump power is approaching threshold, where it starts to decrease. For positive detuning, the effective mass increases slightly and then sharply decreases. Although the decreasing effective mass indicates the change of the nature of the emission states, it does not reach the bare cavity value for any of these detunings. Furthermore, the effective mass just before threshold increases with detuning. For positive detunings there is additionally a small increase with pump power after the preceding sharp drop, indicating flattening of the dispersion curve, which might be related to modifications due to the non-equilibrium nature of the system or to localization [166, 51]. Again, there is no clear boundary between the different features and they change gradually with detuning. The effective mass results are currently not well understood and will require more careful analysis.

2.10 Summary and Future Work

In conclusion, a polariton lasing system is demonstrated, which, although having spectral features commonly identified in a polariton BEC, is shown to take place above the Mott density. This is proved with the measurement of optical gain of the underlying carriers, made possible by the polarization-selective grating-based microcavity. Distinct from a photon laser, it has spectral features including emission linewidth that remains narrow and emission energy well below the cavity or exciton resonance energies and without sudden shifts, suggesting a possible bound state that can be evolved to continuously from polaritons at low density. The lasing state becomes similar

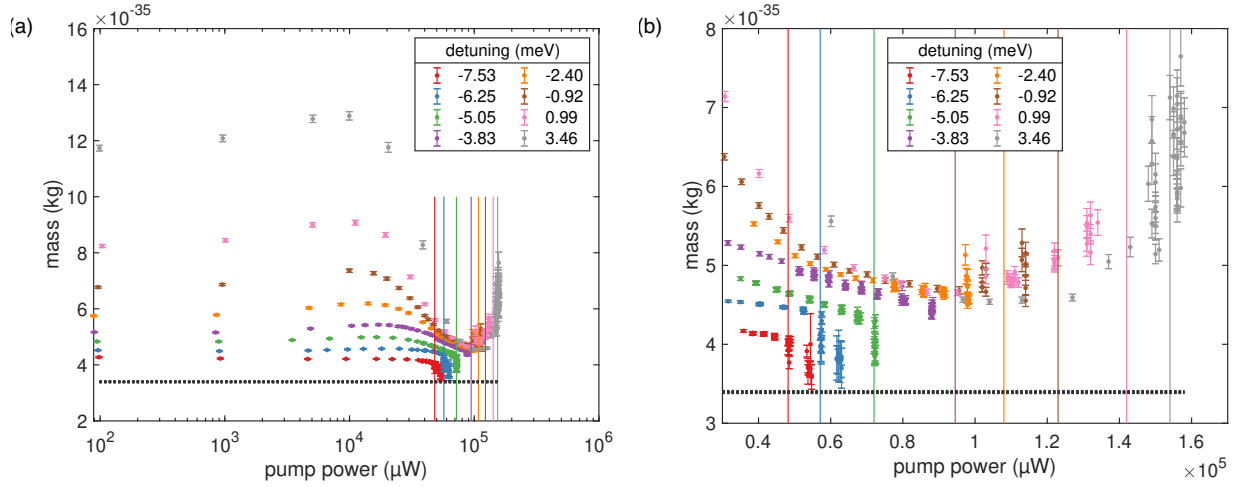


Figure 2.23: **The change in polariton effective mass with pump power.** (a) The pump power dependence of the polariton effective mass obtained with quadratic fitting of the dispersion curves within $|k_{\parallel}| < 1.5 \mu\text{m}^{-1}$, at various detunings. The error bars represent the uncertainty of the fitting result calculated from the diagonal elements of the covariance matrix of the fitting parameters. The effective mass of the cavity resonance dispersion is marked with the horizontal black dotted line. The threshold pump power of each detuning (P_{th}) is labeled with a vertical line in the same color as the data points. (b) The same data as in (a) but limited to near P_{th} and with linear scale in power.

to a photon laser as detuning increases.

The experimental observations are compared to a fermionic many-particle theory that extends the ideal BCS theory to an open, pumped and dissipative system at nonzero temperature. It reveals at the microscopic level that the polariton laser may involve the electron-hole interactions typical of the formation of a polaritonic BCS state and that it may have an electron distribution function and interband polarization that are qualitatively similar to the zero-temperature polariton BCS state.

Furthermore, with a conventional DBR microcavity under ring-shaped pump, similar lasing emission characteristics are observed, as well as the detuning dependence, suggesting possibly similar microscopic origins. The emission spectral features also have a smooth crossover as detuning changes.

Future experimental work can be directly probing the BCS gap using intraband THz spectroscopy [149, 167]. Other possible approaches include an external magnetic field [151, 150]. While current results hint at possible generalization to regular DBR cavities, more measurements and analyses

are still be needed to confirm it. It will also be interesting to realize a polariton BEC in the same system and to observe the crossover among these phases, or to determine whether a non-Hermitian phase transition may take place [168].

CHAPTER 3

Tuning Polariton Properties with Subwavelength Grating

3.1 Linewidth Tuning and Non-Hermitian Physics

Exciton-polaritons are formed by strong-coupling between a photonic cavity mode and an exciton mode. This composite system is intrinsically driven-dissipative, often requiring treatment with non-negligible pump and decay terms similar to pure optical systems. Compared to typical optical systems, the exciton component introduces a much stronger nonlinearity. This makes it a particularly interesting system for studying the topic of parity-time (PT) symmetry and non-Hermitian physics, especially the interplay with nonlinearity-induced features such as superfluidity [9, 11, 169].

Non-Hermitian physics is a framework in which a non-Hermitian Hamiltonian is used to describe a system [170]. It has been utilized to model optical systems, as they naturally involve loss and gain which can be represented as complex diagonal terms in the Hamiltonian [171]. These analyses are often based on, for example, the paraxial equation of wave propagation with complex potential or on coupled mode theory [172]. Such a system can have a complex eigenvalue spectrum. Particularly intriguing are singularities of the spectrum in the parameter space, also called the exceptional points, where eigenstates coalesce [173]. Under this framework, a range of novel phenomena related to exceptional points have been predicted and demonstrated to occur, including uni-directional propagation [174, 175], orbital angular momentum beam emission [176], coexistence of lasing and coherent perfect absorption [177, 178], and enhanced sensing [179]. The intersection between non-Hermitian physics and topological physics has also attracted much attention [180].

There are different approaches of incorporating non-Hermitian models in polariton systems. One such approach is centered on the coupling between the underlying cavity and exciton modes,

which can be subject to gain and loss. In a uniform polariton system, spontaneous condensation into different modes has been predicted to be dependent on the parameter space trajectory around the exceptional points [168], and adiabatic flipping between the photonic and excitonic modes has been envisioned near the exception points [181]. These studies provide new insight to the unique features of non-equilibrium phase transitions. Another approach utilizes localized polariton modes with coupling between them. This provides further freedom to engineer the non-Hermitian system and to access novel phenomena such as the non-Hermitian skin effect due to effective non-reciprocal hopping [182, 183, 184] and synchronization among a large array via supersymmetry selection [185].

Exploration of these phenomena often requires precise tuning of the parameters, including the real and imaginary parts of energies of the individual modes and the coupling strength between them. This can be seen in the following example: Consider a system of two modes of energies $\omega_{1,2}$, gain or loss rates $\gamma_{1,2}$, and coupling strength ν , such that the Hamiltonian is written as

$$\begin{bmatrix} \omega_1 + i\gamma_1 & \nu \\ \nu^* & \omega_2 + i\gamma_2 \end{bmatrix}. \quad (3.1)$$

The eigenvalues are

$$\lambda_{\pm} = \frac{1}{2} \left[\omega_1 + \omega_2 + i(\gamma_1 + \gamma_2) \pm \sqrt{(\omega_1 - \omega_2)^2 - (\gamma_1 - \gamma_2)^2 + 2i(\omega_1 - \omega_2)(\gamma_1 - \gamma_2) + 4|\nu|^2} \right]. \quad (3.2)$$

In the case where $\omega_1 = \omega_2 = \omega$ and $\gamma_1 = -\gamma_2 = \gamma > 0$, sometimes also called a PT-symmetric system, and assuming ν is real, this can be simplified to

$$\lambda_{\pm} = \omega \pm \sqrt{\nu^2 - \gamma^2}, \quad (3.3)$$

and the corresponding eigenvectors are $(1, \pm \exp(\mp i\theta))$, where $\sin(\theta) = \gamma/\nu$. When $\gamma < \nu$, both eigenvalues are real and both eigenvectors have equal magnitude in the two modes. When $\gamma > \nu$, the eigenvalues will have equal real parts and opposite imaginary parts, and the eigenvectors correspondingly have a larger magnitude in one of the modes than the other. At the critical point

$\gamma = \nu$, the derivatives of the eigenvalues with respect to the parameters γ and ν diverge; this point is often called the exceptional point. If the energies are allowed to be different, $\omega_1 = -\omega_2 = \delta$, and the coupling strength ν is fixed, the eigenvalues [Fig. 3.1] will be interchanged when the parameters δ and γ encircle the exceptional point. From this example it can be seen that the properties of the system change drastically near the exceptional point, and to approach it requires changing the relative magnitude between the coupling strength of modes, their detuning, and their gain or loss.

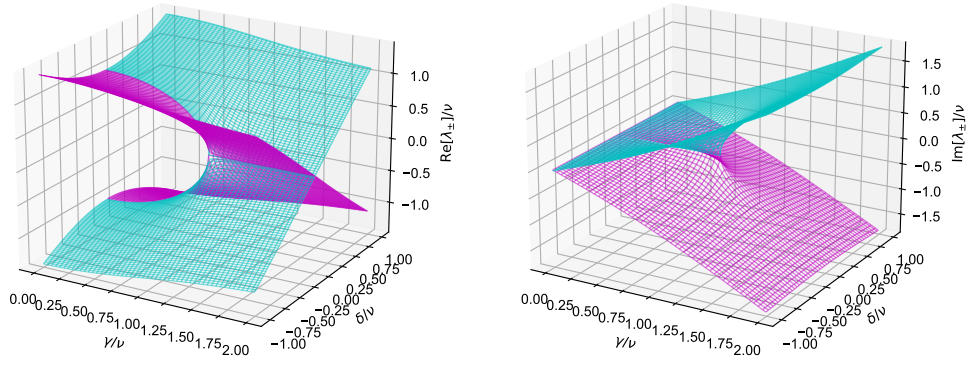


Figure 3.1: **Complex eigenvalues of the matrix in Eq. 3.1.** Here $\delta = \omega_1 = -\omega_2$, $\gamma = \gamma_1 = -\gamma_2$. The two colors represent the two solutions. There is a singularity at $\delta = 0$, $\gamma = \nu$.

Such tuning is difficult with conventional cavities made with DBR mirrors. While the cavity energy is readily tunable with a tapered cavity thickness, other parameters, such as the cavity decay rate, the energy and decay rate of the exciton, and the exciton-photon coupling, are generally not tunable in the same sample. Even the cavity energy cannot be tuned at the same position on the sample if the cavity thickness is fixed. Measurements of exceptional points and related topological invariant have been demonstrated [186], while having to rely on the anisotropy and the variation of TE-TM splitting in the wave vector space, fixed by the cavity materials. Creating a polariton lattice, by etched micropillars or patterned metal layer deposited on top of the cavity [12], introduces variable coupling between localized polariton modes, yet it is challenging to decouple the energy and linewidth of the modes. Alternatively, spatially structured non-resonant optical pumps have also been used to introduce a potential landscape and provide polariton gain, leading to observations such

as a topological Berry phase [187], chiral state emission [188], condensation in topological defect state [189], and optical switching of condensates [190]. However, the pump introduces non-uniform and nonlinear modulations of multiple parameters of the system simultaneously. Understanding the system properties and independent control of the real and imaginary parts of the eigenenergy often require comparison between experiments and simulation for each device and pump configuration, making both *a priori* predictions and extensions to more complicated or large-scale lattices difficult.

In this chapter, a method to independently control both the energy and the linewidth of the photonic cavity mode, and consequently the polariton mode, will be demonstrated. It is based on a hybrid microcavity with an SWG and a DBR as the two reflectors [120]. The reflectivity and reflection phase of the grating, which affect the cavity decay rate and the resonant energy respectively, can be tuned via the geometric parameters of the grating. The complex eigenenergy of the polariton is determined in a completely passive way during the fabrication of the device, which allows the device to be compatible with simple spatially uniform optical pumping or even electrical pumping. The gratings also define the in-plane spatial extent of the polariton mode, thereby allowing the creation of multiple localized modes with controllable coupling strengths that could be potentially extended to large scale devices [122, 123]. The polariton mode is linearly polarized and the polarization is determined by the orientation of the grating. Devices are fabricated and characterized, showing a tuning range on the order of a few meV for the eigenenergy and from sub-meV to a few meV for the linewidth, appropriate for models that require the energy, the linewidth, and the coupling strength to be comparable [168, 181, 191].

3.2 Monolithic Grating and Fabrication

The gratings used in this chapter are slightly different from the ones in the last chapter. The high-refractive-index grating bars are not suspended, but remain attached to the underlying layer. These structures are sometimes called monolithic high-contrast gratings (MHCGs), and have been previously studied as reflectors [192, 193, 194, 195] and used in polariton cavities [121]. Compared to suspended gratings, an MHCG is less prone to damage due to strain and vibration, and can be

made in larger sizes. Most importantly here, the geometric parameters can be controlled with a higher precision while using a simple fabrication procedure.

The mechanism of the high reflectance of an MHCG is similar to the suspended grating, and can be viewed as the result of the cancellation between internal resonances. But the underlying medium, which is on the side of light incidence for cavities, may not have a refractive index low enough to eliminate higher diffraction orders in contrast to the suspended gratings. In principle, the zeroth order reflectivity will be diminished due to the loss of power into higher diffraction orders. However, some of the high reflectance peaks do not change much with the refractive index of the substrate medium, as shown in Fig. 3.2(a), the zeroth order reflection spectra with increasing substrate refractive index. Here, while the high reflectance peak near 600 nm wavelength strongly depends on the substrate index and does not exist above the first order diffraction cutoff, the high reflectance peak near 800 nm wavelength does not shift much with the substrate index, and persists above the cutoff, even though higher order diffraction becomes possible. Intuitively, this can be understood by observing the field distribution in the structure. Fig. 3.2(b) and (d) show the dielectric functions of MHCGs with substrate refractive index of 1 and equal to the grating bars, and Fig. 3.2(c) and (e) show the field amplitudes in the two cases when light in the wavelength range of the persistent high reflectance is normally incident from the substrate (the lower side). In the case where the substrate and the grating bars have equal refractive indices [Fig. 3.2(d, e)], the first order diffraction is allowed. In Fig. 3.2(c), the superposition of the excited resonances inside the grating approximately forms a node near the interface between the grating and the substrate, and the field is almost constant at this interface. This is in contrast to the interface between the grating and the upper medium, where the zeroth order Fourier component is zero, but the field still has large oscillations. When the substrate index is increased from 1, because the resonances have low field distribution inside the substrate, their energy and the field profile do not change significantly [Fig. 3.2(e)]. The node in the field distribution remains near the lower interface, and therefore the high reflectance is preserved. A more rigorous analytical explanation based on impedance matrix can be found in Ref. [196].

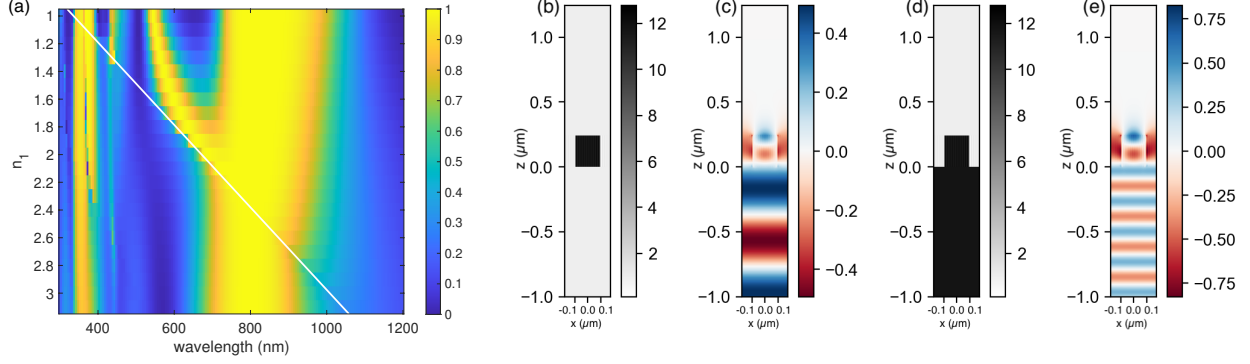


Figure 3.2: Mechanism of the high reflectivity of MHCG. (a) Simulated reflection spectra of an MHCG as a function of substrate refractive index n_1 , for TM-polarized light at normal incidence. In this example, the environment refractive index is 1 and the refractive index of the grating bars is 3.58. The period of the grating is 336 nm, the thickness is 225 nm, and the duty cycle is 0.6. The white solid line is the cutoff of the first order diffraction in substrate. (b) The cross-sectional view of the dielectric function $\varepsilon = n^2$ and (c) the field amplitude distribution E_x when light is incident from the substrate side of an MHCG. In this example, the environment refractive index and the substrate refractive index are both 1 and the refractive index of the grating bars is 3.43. The grating is periodic in x -direction, with period 351 nm, thickness 242 nm, and duty cycle 0.54. (d-e) Same as (b-c) but with the substrate refractive index equal to that of the grating bars.

For the experiment, the grating-based cavity devices are fabricated on GaAs-based epitaxial wafers. These wafers have 12 GaAs/AlAs QWs, each of thickness 12 nm, distributed in three groups at the three highest anti-nodes of the resonant mode of an AlAs $3\lambda/2$ -cavity. Three 4 nm AlAs spacer layers separate the QWs within each group. The lower mirror of the cavity is an $\text{Al}_{0.15}\text{Ga}_{0.85}\text{As}/\text{AlAs}$ or $\text{Al}_{0.2}\text{Ga}_{0.8}\text{As}/\text{AlAs}$ DBR with a total of 36 pairs of layers. The upper mirror is an assembly of an $\text{Al}_{0.2}\text{Ga}_{0.8}\text{As}$ grating layer, an $\text{Al}_{0.85}\text{Ga}_{0.15}\text{As}$ spacer layer, and 1.5 pairs of DBR. Each grating device is in a $25\ \mu\text{m} \times 25\ \mu\text{m}$ or $30\ \mu\text{m} \times 30\ \mu\text{m}$ square region, and many devices are made with different periods and duty cycles near the reflectivity maximum.

The procedures of the fabrication can be summarized as follows: A 300 nm layer of ZEP520A e-beam resist is spin-coated on the top $\text{Al}_{0.2}\text{Ga}_{0.8}\text{As}$ layer of the wafer. The pattern is written with e-beam lithography with a dose of about $165\ \mu\text{C cm}^{-2}$ and a shot pitch of 12 nm. The pattern is then transferred to the grating layer with BCl_3 reactive ion etching. Finally, the remaining resist is removed by oxygen plasma. There are several notable issues in the fabrication process. First,

the etching should stop at the second layer, which is $\text{Al}_{0.85}\text{Ga}_{0.15}\text{As}$. A selective etching method using a combination of BCl_3 and SF_6 that can etch GaAs and stop on AlGaAs has been reported [197, 198]. However, since this method relies on the passivation of Al for the etching stop, when it is applied to the present $\text{Al}_{0.2}\text{Ga}_{0.8}\text{As}/\text{Al}_{0.85}\text{Ga}_{0.15}\text{As}$ structure, the selectivity drops sharply due to the nonzero Al fraction. The selectivity achieved on current $\text{Al}_{0.2}\text{Ga}_{0.8}\text{As}/\text{Al}_{0.85}\text{Ga}_{0.15}\text{As}$ samples without resist is only about 2 : 1 even with optimized parameters. The improvement over non-selective BCl_3 etching does not outweigh the undesirable side effects such as slanted sidewalls, even when a two-step process with non-selective etching first and ending with selective etching [121] is used. In contrast, non-selective etching with calibrated etch time has produced more consistent results and has been used in this work. Second, the etching rate is also not uniform among different feature widths, known as aspect ratio dependent etch-rate [199]. This has been partly circumvented by using thinner resist layer of about 150 nm and limiting the range of feature width used in a single etch. Finally, there is some residual e-beam resist even though most of the resist can be removed by either oxygen plasma or commercial PRS-2000 solvent. While other methods of removing the resist might produce a cleaner outcome, presently the residue is mainly distributed near the corner of the etched features within about $4\text{ }\mu\text{m}$ and should not affect larger devices too much. Fig. 3.3 shows examples of MHCG fabricated with this process.

3.3 Tuning Mechanism

It has been discussed in the previous section that the high reflectivity of the grating mirror relies on the cancellation of the resonances of the grating structure. The resonance energy and the field amplitude distribution of each of the modes are sensitive to the geometry of the grating bars with different dependencies. Therefore, a change in the period or the duty cycle of the grating results in an overall change of the amplitude and phase of the reflected field. This makes it possible to tune the energy and linewidth of the cavity resonance locally.

To evaluate the accessible range of tuning, numerical simulations¹ with the RCWA method [154]

¹The numerical solver from Ref. [200] is used here.

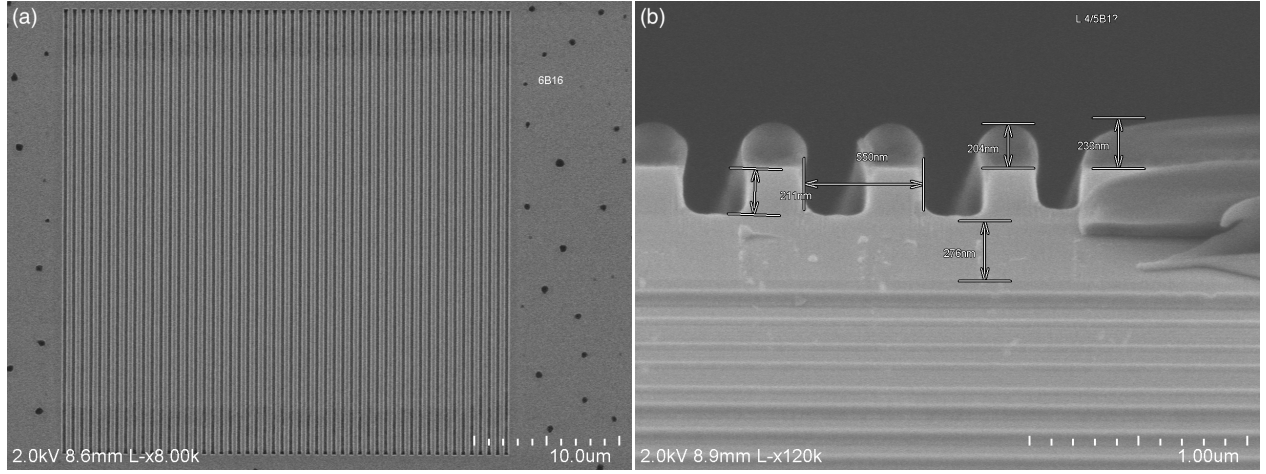


Figure 3.3: **SEM images of MHCG.** (a) Image of a $30\text{ }\mu\text{m} \times 30\text{ }\mu\text{m}$ grating. (b) Cross-sectional image of a grating after etching, with the remaining e-beam resist still on top of it.

are first performed for a realistic grating mirror structure. The mirror consists of an $\text{Al}_{0.2}\text{Ga}_{0.8}\text{As}$ grating on top of an $\text{Al}_{0.85}\text{Ga}_{0.15}\text{As}$ layer, followed by a 1.5-pair DBR, with the DBR side facing an AlAs cavity. Fig. 3.4(b, c) shows an example of the zeroth-order reflectivity and reflection phase near a high-reflectivity region. The reflection is computed for 800 nm light with the electric field polarized perpendicularly to the grating bars and normal incidence from the AlAs side. For the one-dimensional gratings, the reflection properties can typically only be optimized for one of the orthogonal linear polarizations and not both simultaneously. In this example, for the polarization parallel to the grating bars the reflectivity in this parameter range is only about 0.5. The grating has a thickness of 242 nm and is optimized for high reflectivity at 800 nm. The period Λ and the duty cycle η are the geometric tuning parameters and the highest reflectivity is obtained at $(\Lambda_0, \eta_0) = (351\text{ nm}, 0.54)$. The thickness of the $\text{Al}_{0.85}\text{Ga}_{0.15}\text{As}$ layer is chosen so that the reflection phase is π at the highest reflectivity.

As (Λ, η) are tuned away from (Λ_0, η_0) in the two-variable parameter space, the reflectivity decreases [Fig. 3.4(b)] while the reflection phase [Fig. 3.4(c)] changes monotonously in the vicinity. Therefore, by selecting certain combinations of period and duty cycle, it is possible to attain certain reflectivity and phase separately. The available ranges of the reflectivity and phase are not

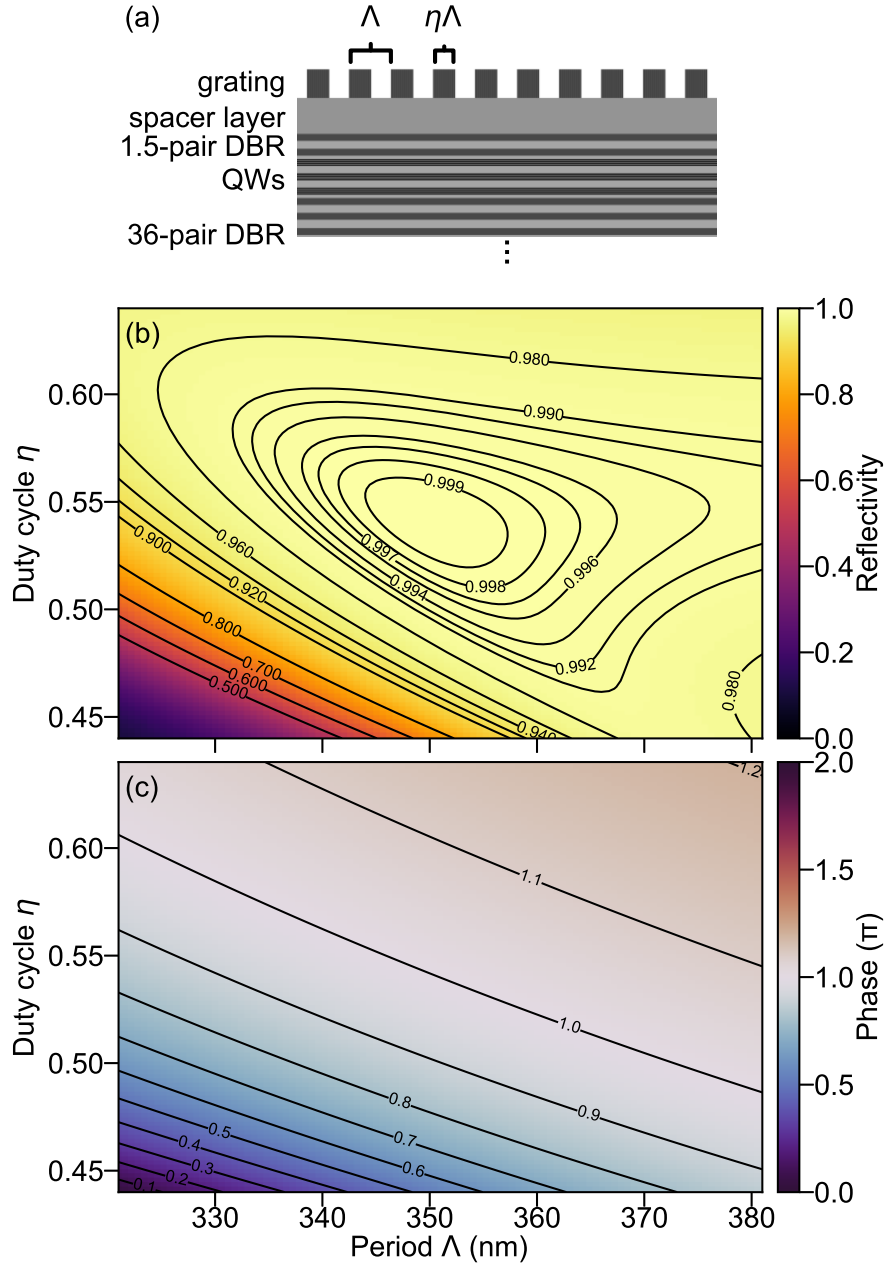


Figure 3.4: **Reflectivity and reflection phase of a grating-based mirror.** (a) A schematic of the cross-sectional profile of the microcavity. (b-c) Simulated zeroth-order reflectivity (b) and reflection phase (c) of a grating-based mirror as a function of the period and the duty cycle of the grating, with 800 nm light polarized perpendicularly to the grating bars and normal incidence from the AlAs side.

independent. With Fig. 3.4(b, c) as an example, given a fixed reflectivity, the corresponding (Λ, η) can only move on a contour line, corresponding to a limited range of phase. However, as will be shown later, the ranges are often adequate.

In experiment, the thicknesses of the layers of the wafer as well as the parameters of the grating may differ from the design, which can be calibrated through iterations between simulation and measurement of the energy and linewidth.

3.4 Experimental Demonstration of Linewidth Control

Fig. 3.5 shows photoluminescence (PL) spectra of typical devices in Fourier-space, with a polarizer perpendicular to the grating bars. The devices are pumped with a non-resonant CW laser focused to a $2\text{ }\mu\text{m}$ spot in the center of the grating. The sample is kept at 5 K temperature. Strong-coupling is evidenced by the emission from LP and UP branches for both devices in display. There is emission from near the exciton energy, due to the inhomogeneous broadening of the exciton [201] or emission from excitons with large wave vectors through first order diffraction of the grating. For the orthogonal polarization, the excitons remain in the weak-coupling regime.

These two devices provide an example of local linewidth control by the grating parameters. Their design parameters are $\Lambda = 324\text{ nm}$, $\eta = 0.763$ [Fig. 3.5(a)] and $\Lambda = 348\text{ nm}$, $\eta = 0.763$ [Fig. 3.5(b)]. The resulting LP modes have nearly the same energy at zero wave vector at $(1549.98 \pm 0.02)\text{ meV}$ and $(1549.99 \pm 0.01)\text{ meV}$, but the linewidths are vastly different at $(1.67 \pm 0.07)\text{ meV}$ and $(0.67 \pm 0.01)\text{ meV}$ [Fig. 3.5(c)]. The Rabi splitting is $\hbar\Omega_R \approx 10\text{ meV}$ at zero wave vector, computed from the exciton energy measured in the unetched area near the device and both the UP and the LP energies. The dispersions of the cavity, and thus of the polaritons, have a small deviation from that of a conventional DBR-DBR microcavity, whose cavity resonance dispersion is approximately quadratic near zero wave vector, because the grating reflection phase also depends on the wave vector. The polariton dispersions of conventional DBR-DBR microcavities are calculated with a coupled oscillator model and overlaid on the spectra for comparison, with parameters that result in the same exciton and polariton energies at zero wave vector as the

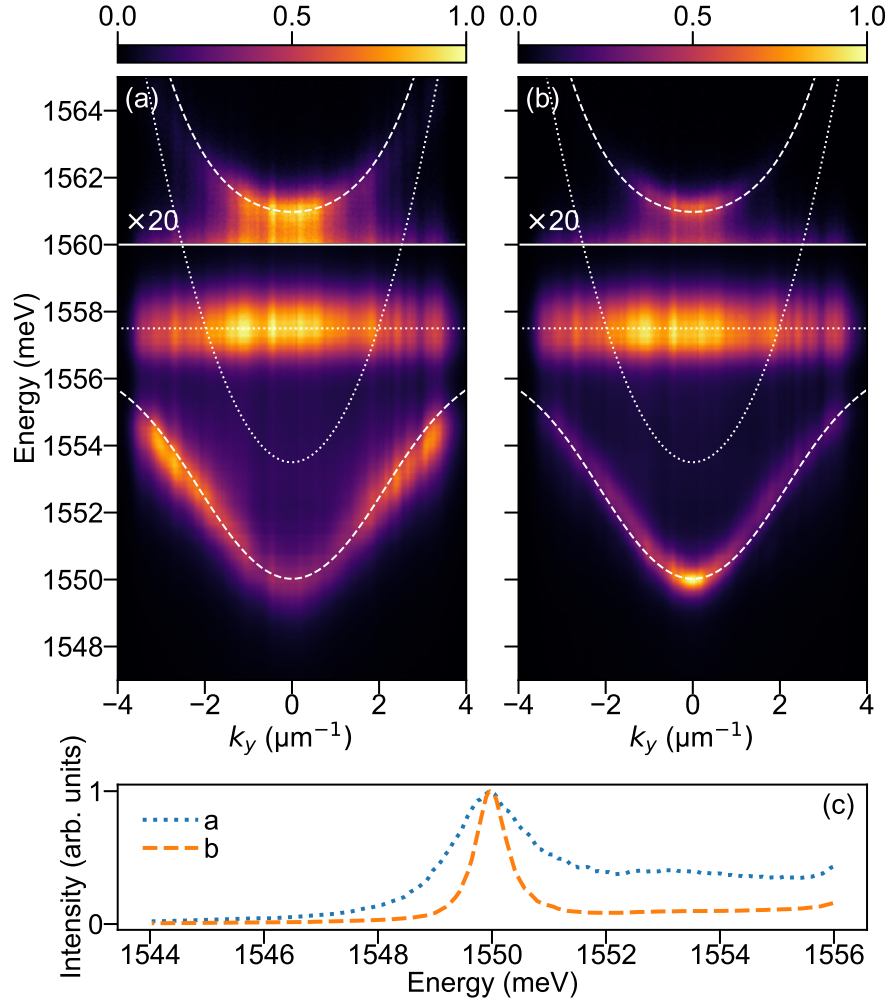


Figure 3.5: **Fourier-space PL spectra of typical devices showing equal LP energy but different linewidths.** (a) Grating with designed period 324 nm and duty cycle 0.763. (b) Grating with designed period 348 nm and duty cycle 0.763. In subfigures (a) and (b), the horizontal axes are the wave vector parallel to the grating bars (k_y). The spectra are collected with a polarizer perpendicular to the grating bars. The dashed lines are the polariton dispersions of a conventional DBR-DBR microcavity for comparison, calculated with a coupled oscillator model assuming quadratic cavity resonance dispersion and constant exciton energy for the relevant wave vectors. The parameters in the calculation are chosen to obtain the same exciton and polariton energies at zero wave vector as the measured devices, and the uncoupled exciton and cavity energies are marked with the dotted lines. (c) Line spectra near the LP energy corresponding to (a) and (b), integrated within $k_y < 0.1 \mu\text{m}^{-1}$ and each normalized to its maximum value in the energy range.

measured devices and assuming a quadratic cavity resonance dispersion.

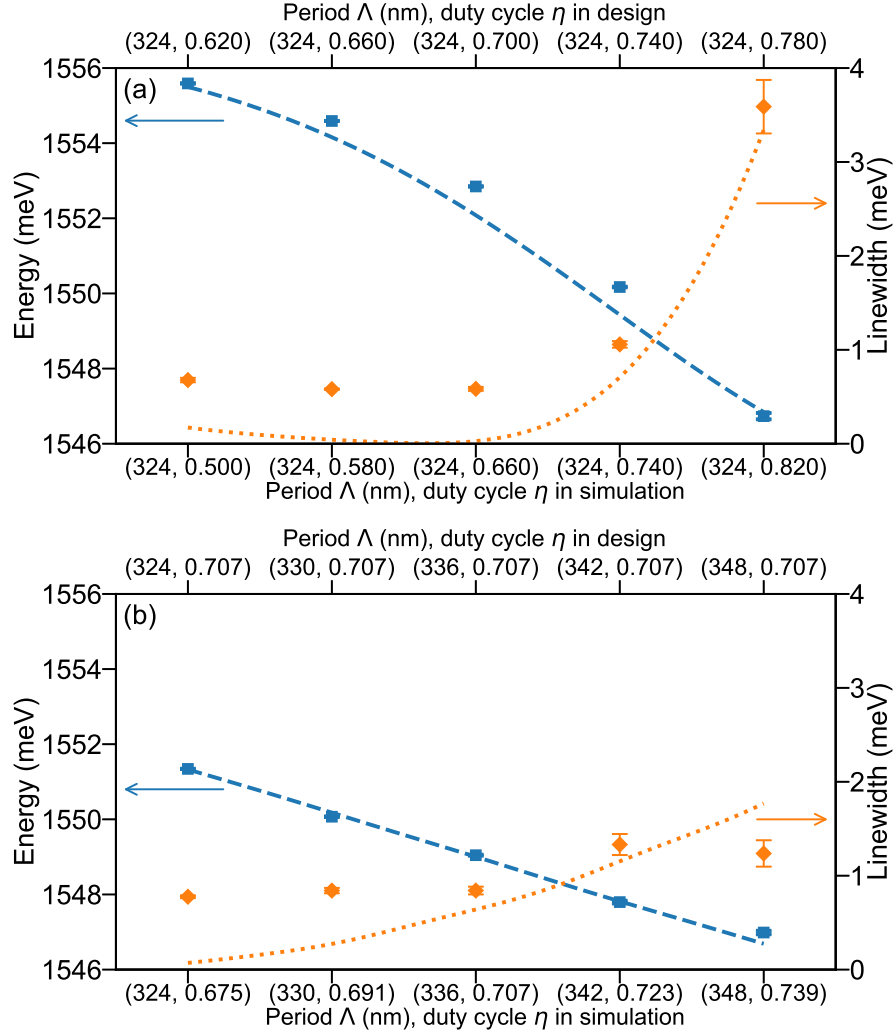


Figure 3.6: **Measured and simulated lower polariton energy and linewidth at zero wave vector for two series of gratings.** The two series are shown in (a) and (b) respectively. (a) shows gratings designed to have the same period and varying duty cycles, while (b) shows varying period and the same duty cycle. The upper horizontal axis labels the designed parameters while the lower horizontal axis labels the parameters used in simulation. The experimental energy (blue square markers) and linewidth (orange diamond markers) values are obtained by curve fitting of the PL spectra with Lorentzian peaks and the error bars represent the uncertainty of the corresponding fitting parameters calculated from the diagonal elements of the covariance matrix of the parameters. The simulated energy (dashed lines) and linewidth (dotted lines) values are obtained by curve fitting of the simulated reflection spectra with Fano lineshape.

To evaluate experimentally viable step sizes of the individual parameters, arrays of gratings

are fabricated, with a range of different periods Λ and duty cycles η . The parameter values measured with a scanning electron microscope (SEM) are then compared with the design. The measured polariton PL energy and linewidth of these devices are also compared with the values from RCWA-simulated reflection spectra [Fig. 3.6]. All RCWA simulations are performed with 41 basis functions and numerical convergence is verified.

The period Λ of the fabricated devices follows the design very well apart from a $(2.8 \pm 0.4)\%$ offset above the designed values. This is consistent among devices with different parameters and the offset is likely from an overall error in the calibration of the magnification of either the lithography or the measurement. The measured duty cycle η sometimes has a larger deviation from the design, due to the limitation of beam positioning accuracy of the e-beam lithography and due to insufficient anisotropy of the etching process. For example, when the duty cycle is kept the same in the design and only the period is changed (near 340 nm), the duty cycle can still vary by 0.07 (about 24 nm). If the duty cycle is also varied in design, the discrepancy can be even larger, though the measurement uncertainty of the duty cycle is limited by the SEM to about 20 nm.

To calibrate the actual duty cycle more accurately, the measured energy and linewidth of the devices are compared with RCWA simulations where the period is fixed at the designed value and the duty cycle is allowed to be scaled linearly with the designed values of both period and duty cycle. As shown in Fig. 3.6, the measured linewidth follows qualitatively the simulated results but are generally larger than the simulated linewidth. This is expected as the linewidth of the device is susceptible to imperfections from fabrication, such as sidewall roughness and surface scattering. The finite size of the gratings can also contribute to the broadening, although it is not expected to be dominant here because the size of the devices is larger than 50 periods of grating [202, 203]. On the other hand, the polariton energies show excellent agreement between simulation and measurements.

From this result a step size of 6 nm for the period and 0.08 (26 nm) for the duty cycle is estimated. For these devices, the step sizes translate to polariton energy resolution of about 1 meV and linewidth resolution down to 0.5 meV. By mixing changes in both parameters, it is possible to obtain finer steps of polariton energy or linewidth. It is expected that the duty cycle step size can

be further reduced to < 6 nm with improvements to the fabrication process.

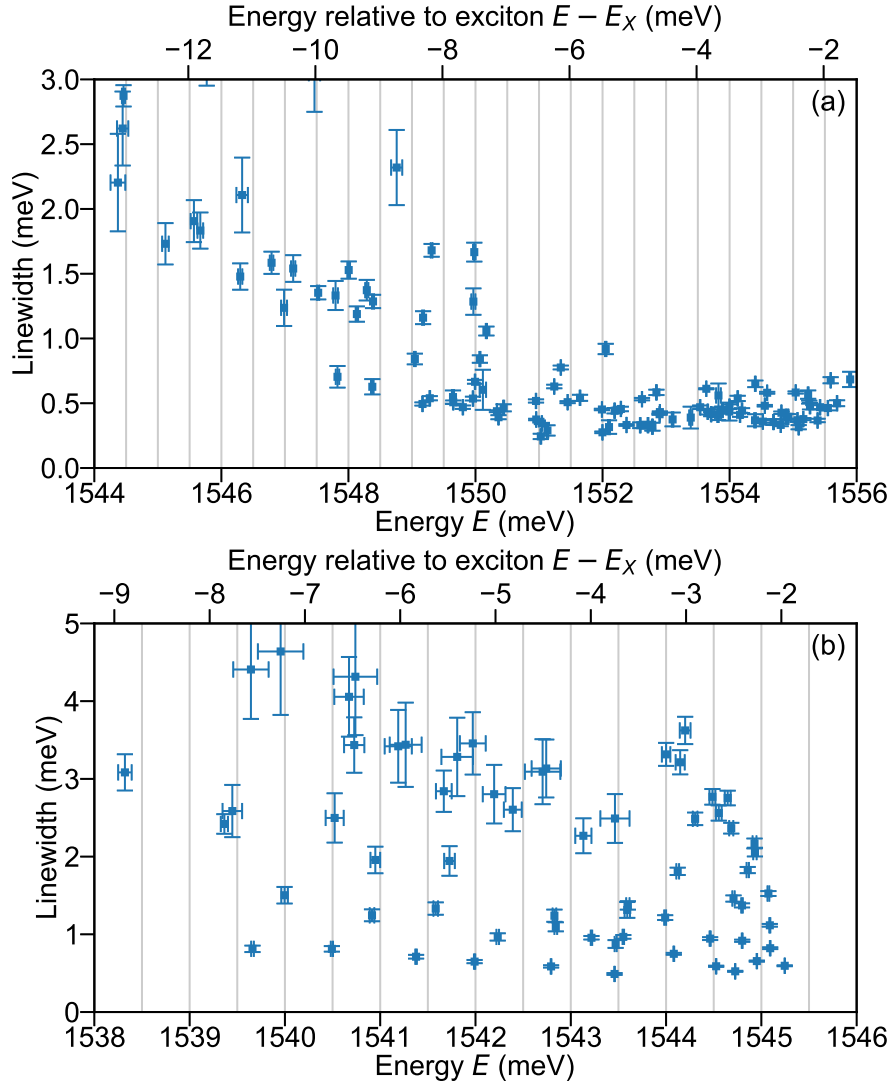


Figure 3.7: **A collection of measured PL energy and linewidth of LP modes at zero wave vector of gratings with various parameters on two different sample pieces.** (a) The gratings have designed periods around 336 nm and duty cycles around 0.72. (b) The gratings have designed periods around 550 nm and duty cycles around 0.50. Gray vertical lines mark 0.5 meV energy spacing. The experimental energy and linewidth values are obtained by curve fitting of the spectra with Lorentzian peaks and the error bars represent the uncertainty of the corresponding fitting parameters calculated from the diagonal elements of the covariance matrix of the parameters.

To evaluate how broadly the energy and linewidth can be tuned separately, arrays of gratings are fabricated with varying Λ and η on $5 \text{ mm} \times 5 \text{ mm}$ wafer pieces. Fig. 3.7(a, b) shows the measured

PL energy and linewidth of LP modes at zero wave vector from the gratings on two sample pieces respectively.

In general, the ranges of energy and linewidth are limited by the choice of the other parameter. To obtain a certain detuning, for example, will require the parameters to stay on a curve in the parameter space that gives the grating mirror the correct reflection phase for that cavity energy, and the reflectivity can only take values on that curve, limiting the linewidth. On the other hand, the reflectivity needs to be kept high enough for strong-coupling, which limits the range of detuning. For the piece shown in Fig. 3.7(a), the energy range spans more than 10 meV. For energies below 1550 meV, the linewidth range spans over 1.5 meV. For energies above 1550 meV, the linewidth is narrower and ranges between 0.24 meV and 0.92 meV. The reduced linewidth range is due to the more positive detuning and the smaller photonic fraction at these energies, as the linewidth variation comes from the photonic mode. Results from a similar wafer piece is shown in Fig. 3.7(b), where high-reflectivity is found in the parameter region near $\Lambda \sim 550$ nm and $\eta \sim 0.5$. In this piece, for more than 5 meV of energy range, the linewidth can be tuned by more than 2 meV.

With these records as a calibration, a practical procedure can be followed to design and fabricate new devices that require local modes with different complex eigenenergies: first determine the target energy and linewidth, and then select the corresponding parameters that achieve those values from the calibration results.

3.5 Coupled Devices and Future Work

Finally, with the tuning capabilities demonstrated, the possibility of realizing a simple design of non-Hermitian devices that consist of two coupled local polariton modes is considered. For such devices, it is often required that the coupling strength is comparable to the difference between the imaginary parts of the energies. It has been shown that SWG-based cavities can confine polariton modes in-plane to smaller than $10\ \mu\text{m}$, and that these modes can be coupled to form dimer-like modes and one-dimensional chains [120, 122]. The coupling strength between neighboring modes is on the order of 0.5 meV [123], comparable to the tuning range of the linewidth in the present case.

In Fig. 3.7 vertical lines with 0.5 meV spacing are added in the background to show the available choices of linewidth within 0.5 meV energy bins.

Preliminary results from the attempt to make coupled-mode devices with MHCG are presented in Fig. 3.8. In this example, the localized modes are formed with gratings each in a circularly-shaped area. Two of these localized modes are placed together, partly overlapping. They are embedded in a background of gratings in the perpendicular orientation. The design of a device with circles of 4 μm diameter and a center-to-center distance of 2.4 μm are shown in Fig. 3.8(a), and the corresponding SEM image is shown in Fig. 3.8(b). The PL of these devices is measured under non-resonant CW laser excitation with a large beam spot covering the entire device. At low pump power, the polariton emission within the localized modes is weaker than the exciton emission outside this area, although the background gratings in the perpendicular orientation partly suppress the exciton emission intensity. At high pump power, however, the polariton emission within the localized modes dominates the spectra. Fig. 3.8(c-d) shows the real-space and Fourier-space spectra of a device with 5 μm diameter circles and 4.5 μm center-to-center distance, measured in the direction parallel to the line through the centers of the circles, and with polarization perpendicular to the grating bars. Two eigenmodes are visible, the one at lower energy being a symmetric mode, and the one at higher energy being an antisymmetric mode, as evidenced by a node at wave vector $k_{\parallel} = 0$. The energy separation between the eigenmodes is about 0.5 meV. Presently, the coupled-mode devices that have been made all have the same geometric parameters for the gratings for both of the localized modes. Future work could include using different parameters to achieve different linewidths and energies for the two uncoupled modes, or even more complicated schemes.

In summary, grating-based microcavities can be used to create polariton modes with controllable energy and linewidth. The polariton energy can be tuned over a range comparable to that of the normal mode splitting, while the linewidth can be independently tuned from a fraction of a meV to a few meV. The tuning resolution of 0.5–1 meV is mainly limited by the e-beam lithography resolution of 24 nm. Improvements in the fabrication accuracy, especially in the e-beam lithography, will enable finer resolution. The demonstrated system is scalable to large lattices. The ability to

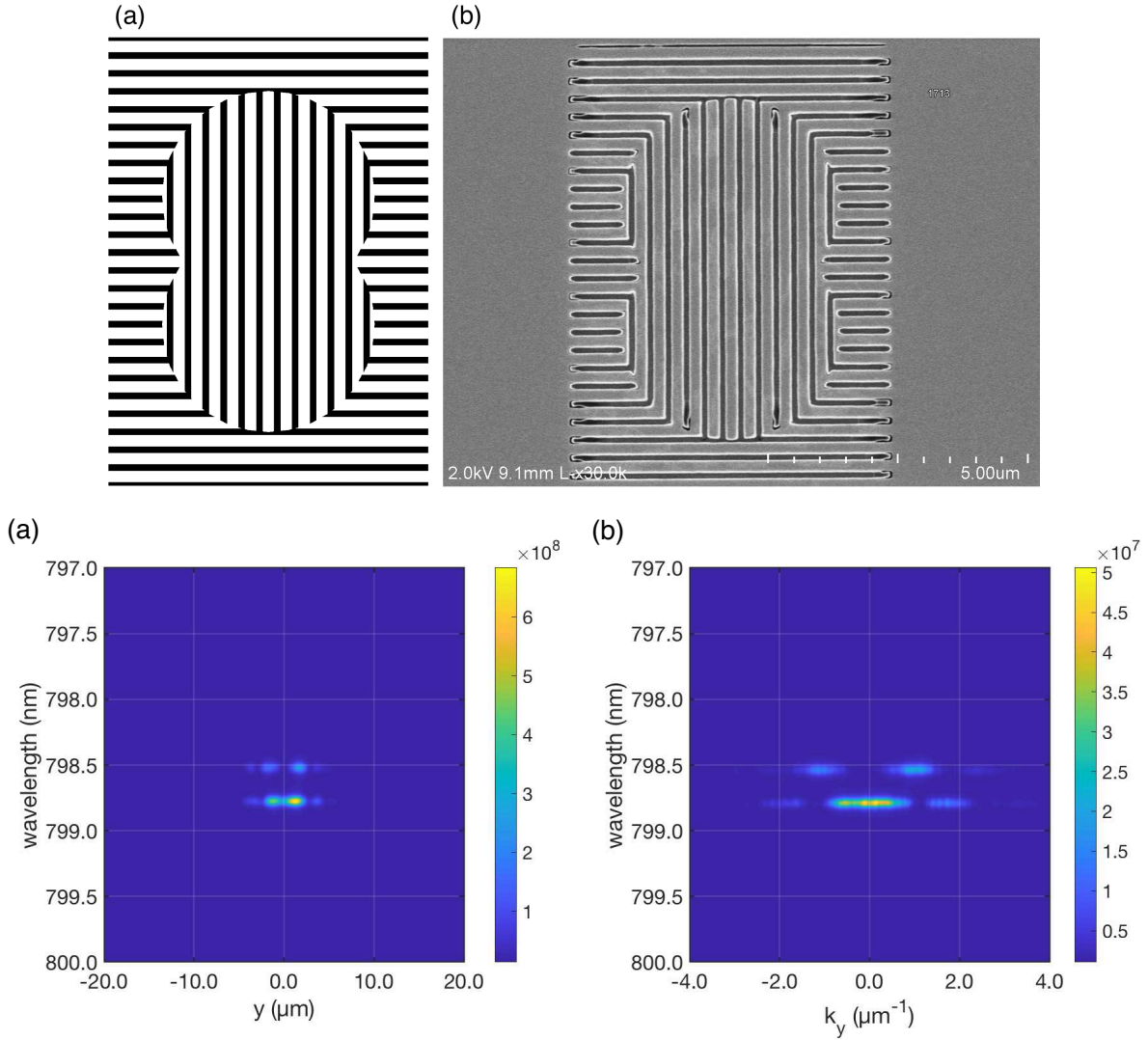


Figure 3.8: **Coupled grating-based polariton devices.** (a) The design of a coupled device consisting of two overlapping circularly-shaped grating-based cavities. The regions shaded in black are to be etched in the first layer of the sample. The two circular shapes are of $4\mu\text{m}$ diameter and have a center-to-center distance of $2.4\mu\text{m}$. (b) SEM image of the fabricated device corresponding to the design shown in (a). (c-d) The PL spectra of a device with $5\mu\text{m}$ diameter circles and $4.5\mu\text{m}$ center-to-center distance, under around 32 mW pump power. (c) Real-space spectra along $x = 0$ where y is the direction parallel to the line through the centers of the circles and the origin is at the center of symmetry of the device. The electric field is polarized along x . (d) Fourier-space image along $k_x = 0$, same polarization as in (c).

accurately and independently tune the energy and loss and gain of the system may facilitate the study a wide range of non-Hermitian phenomena using a polariton platform [204, 168, 181, 191], and may also enable development of single-photon or entangled-photon sources based on coupled, driven polariton systems [205, 206, 207].

CHAPTER 4

Polariton Cavity with a Fano Interference-Based Reflector

4.1 Fano Interference-Based Reflector and Cavity

The interference between a discrete resonance and a continuum is known as Fano interference and is often associated with a characteristic asymmetric lineshape [208]. Although initially considered for the scattering between electrons and atoms, it has been observed in many different fields involving wave phenomena, including optics [209, 210]. Particularly, the Fano interference between a discrete mode of a resonator in an optical path and the direct transmission continuum, which can be found in photonic crystal slabs, can create an asymmetric shape in transmission and reflection spectra that goes from 0 to 1 near the resonance energy of the discrete mode. The spectra can be modeled by a set of equations for the amplitude of the discrete mode in the resonator and the waves coupled to it [211, 212, 213, 214, 215, 216]:

$$\frac{da}{dt} = (-i\omega_0 - \gamma) a + \begin{bmatrix} d_1 & d_2 \end{bmatrix} \begin{bmatrix} s_1^+ \\ s_2^+ \end{bmatrix}, \quad (4.1)$$

$$\begin{bmatrix} s_1^- \\ s_2^- \end{bmatrix} = C \begin{bmatrix} s_1^+ \\ s_2^+ \end{bmatrix} + a \begin{bmatrix} d_1 \\ d_2 \end{bmatrix}. \quad (4.2)$$

Here, a is the amplitude of the discrete mode with resonance frequency ω_0 and decay rate γ . $s_{1,2}^+$ ($s_{1,2}^-$) are the amplitudes of waves in the media on the two sides of the resonator, propagating into (out of) the resonator, and $d_{1,2}$ are the coupling coefficients to the discrete mode. If there is no decay channel other than the coupling to the two media, then energy conservation requires $\gamma = \frac{|d_1|^2 + |d_2|^2}{2}$. $C = \begin{bmatrix} r_B & -it_B \\ -it_B & r_B \end{bmatrix}$ is a scattering matrix for the background reflection r_B and transmission t_B in the

absence of the resonator. These equations yield the solution

$$a = \frac{d_1 s_1^+}{-i(\omega - \omega_0) + \gamma}, \quad (4.3)$$

and the transmission and reflection amplitudes

$$t = -it_B + \frac{d_1 d_2}{-i(\omega - \omega_0) + \gamma}, \quad (4.4)$$

$$r = r_B + \frac{d_1^2}{-i(\omega - \omega_0) + \gamma}. \quad (4.5)$$

The transmission power coefficient is

$$T = |t_B|^2 \frac{(\epsilon - \cot \Delta)^2}{1 + \epsilon^2}. \quad (4.6)$$

This is in the form of a Fano lineshape, with detuning $\epsilon = \frac{\omega - \omega_0}{\gamma}$ and asymmetry factor $q = -\cot \Delta$ where $\Delta = \pm \sin^{-1} \frac{t_B \gamma}{|d_1 d_2|}$ and the symmetry of the mode determines the sign. The reflection power coefficient is then $R = 1 - |t_B|^2 \frac{(\epsilon - \cot \Delta)^2}{1 + \epsilon^2}$ and reaches 1 when $\epsilon = \cot \Delta$.

Fig. 4.1 shows an example of such a resonator and its reflection spectrum and phase. The discrete mode is provided by a one-dimensional Si_3N_4 grating on SiO_2 substrate [Fig. 4.1(a)]. Light with electric field polarized in the direction parallel to the gratings (TE-polarization) and incident normally from the air side of the grating can excite a resonance in the grating structure near 730 nm. The resonance is coupled to the media on both sides of the grating, and can interfere with the continuum spectrum of direct transmission through the Si_3N_4 layer. The resulting total reflection power spectrum and the corresponding reflection phase delay are shown in Fig. 4.1(b-c), and are compared with a conventional $\text{SiO}_2/\text{TiO}_2$ DBR with 12 pairs of quarter wave layers. The reflectance only has a narrow peak reaching 1 instead of the broadband near-1 reflectivity of the DBR, and the phase changes much faster near the resonance compared to the DBR.

This type of resonators can substitute for the reflectors of FP cavities [217, 218, 219, 216, 220] (the resulting cavity is called a Fano cavity in the remaining text). The resulting cavity resonance

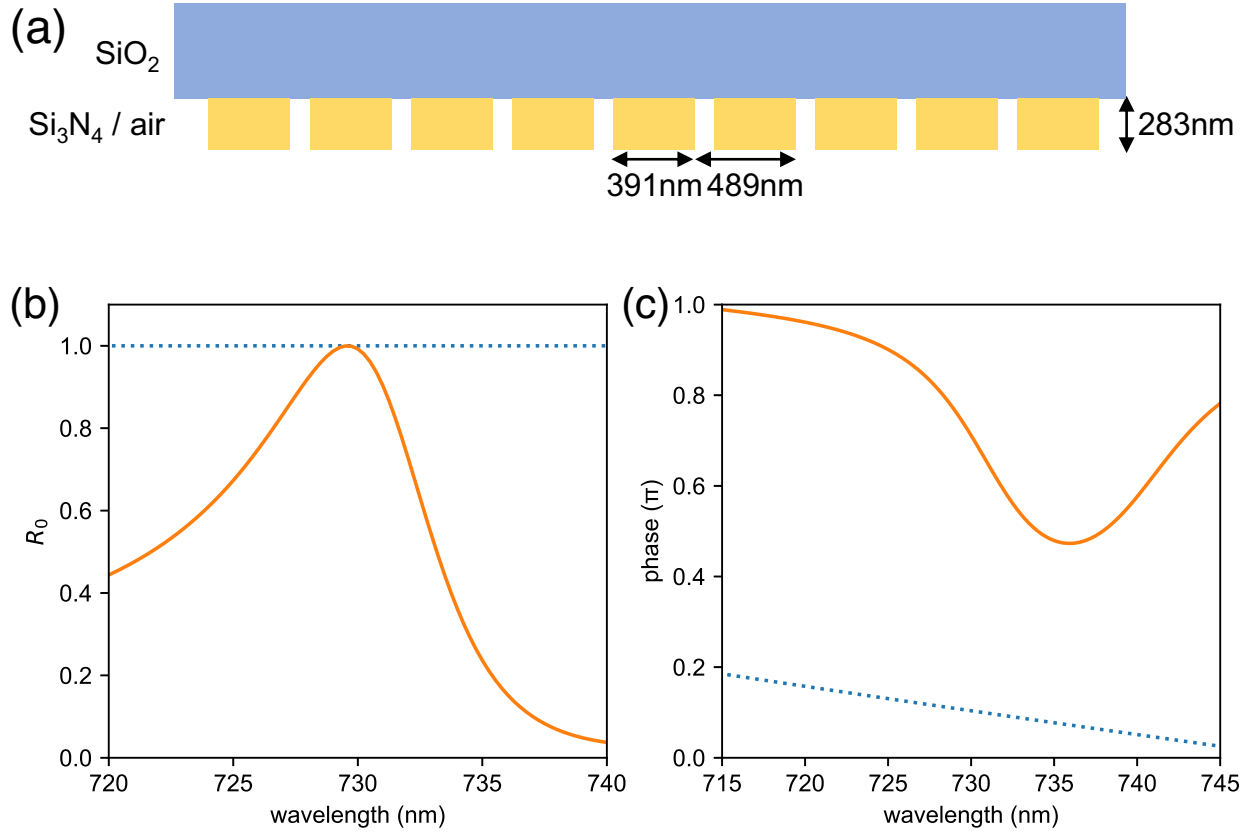


Figure 4.1: **Schematic and reflection spectrum of a Fano interference-based reflector.** (a) Schematic of a Si_3N_4 -on- SiO_2 Fano interference-based reflector. Light is incident from the air side of the grating. (b-c) The total reflection power spectrum (b) and the corresponding reflection phase delay (c) of the Fano interference-based reflector (orange solid lines) and a DBR (blue dotted lines) for comparison.

also has a Fano lineshape, as can be computed by adding the equation

$$s_1^+ = s_1^- r_1 \exp\left(\frac{2i\omega L}{c}\right) \quad (4.7)$$

to Eq. 4.3 and Eq. 4.4, where ω and c are the frequency and speed of light, L is the cavity length, and r_1 is the reflection amplitude of the reflector on the other end of the cavity, typically a DBR. There are additional different features compared to a conventional cavity with a DBR for both reflectors, mainly due to the large phase variation of the Fano interference-based reflector. The cavity resonance needs to satisfy the round-trip phase condition

$$\phi(\omega_m) = \frac{2\omega_m L}{c} + \phi_F(\omega_m) + \phi_1(\omega_m) = 2\pi m, \quad (4.8)$$

where $\phi_F(\omega)$ is the reflection phase of the Fano interference-based reflector, $\phi_1(\omega)$ the reflection phase of the other reflector, and m the mode index of the cavity resonance. With large positive $\frac{d\phi_F}{d\omega}$, the linewidth of the cavity resonance is narrowed due to enhanced $\frac{d\phi}{d\omega}$, which makes it easier to obtain a high-Q cavity [216]. However, the high-Q cavity still critically requires small detuning between the cavity and the discrete mode resonances $\delta = \omega_m - \omega_0$, as the reflectivity of the mirror drops quickly at larger detuning. The cavity resonance frequency is stabilized against cavity length variation near the cavity resonance, as the magnitude of $\frac{d\omega_m}{dL} = -\frac{2\omega_m}{c} \left(\frac{2L}{c} + \frac{d\phi_F}{d\omega_m} + \frac{d\phi_1}{d\omega_m} \right)^{-1}$ is reduced. Fano cavities have been employed in photonic crystals, with in-plane defect waveguides and side-coupled cavities providing the continuum and the discrete modes. There have been experimental demonstration of ultra-coherent lasing due to the high-Q cavity resonance and additional noise suppression by arrangement of the position of the active material tailored to the field distribution [219]. In the same type of experimental system, the strong frequency dependence of the reflectivity and therefore the strong dependence of cavity Q on detuning have been utilized to achieve passive Q-switching and a self-pulsing laser [218]. There are also theoretical proposals for vertical cavities using the same principle [216], including the study of possible suppression of Stokes or anti-Stokes scattering due to the asymmetric cavity transmission [220].

In this chapter, the possible usages and advantages of Fano cavities for exciton-polariton studies will be discussed. Realistic sample structures have been designed and simulated¹, showing the strong-coupling between QW exciton and the high-Q cavity. As a preliminary experimental attempt, fabricated reflectors are measured and empty cavities are tested in an open cavity setup.

4.2 Application to the Exciton-Polariton System

The grating reflectors in the two preceding chapters both have their own limitations. The suspended SWG in Chapter. 2 mainly suffers from its mechanical complexity and fragility. The suspended structure is prone to damage during fabrication and measurement, especially for large devices and high pump power [221]. The samples also degrade over time in long-term storage. The bending of the grating and therefore the required strain relief patterns also increase the difficulty to precisely predict and tune the resonance. The monolithic design in Chapter. 3 is more stable and precise. However, in our GaAs-based samples, at the optimal parameters the gratings are only subwavelength with respect to the air side but not to the other layers in the wafer. The additional diffraction orders make it impossible to get reflection spectra similar to those in Chapter. 2 where the absorption or gain of the carriers can be easily obtained, because most of the normally incident light is diffracted into the first orders. In PL measurements, strong exciton emission from the wave vectors corresponding to the first diffraction orders can obscure the weak polariton emission of interest. Ideally, monolithic gratings with high enough refractive index to eliminate the first diffraction orders may have the advantages of both types of gratings. But for the current AlGaAs system, a lower Al fraction will introduce absorption at the GaAs QW exciton wavelength, which lowers the cavity Q-factor.

This has led to the consideration of a reflector separated from the rest of the cavity, in an open cavity setup similar to Refs. [109, 111, 222]. This removes the restrictions on the selection of materials that is otherwise necessary to allow epitaxial growth, such that materials that have established fabrication processes to produce high quality photonic crystals such as Si₃N₄ can also

¹Simulations are performed with the numerical solver from Ref. [200].

be used. The grating reflectors used in the previous chapters are utilizing the cancellation between two broad resonances close to each other in frequency to achieve a broadband high-reflectivity. The linewidths of these resonances depend on the modulation strength of the refractive index in the grating layer. With a Si_3N_4 grating like the example in Section. 4.1, the weaker modulation strength results in narrower linewidth compared to the AlGaAs gratings, and broadband high-reflectivity is more difficult to achieve. However, as shown in Section. 4.1, even with just one resonance, it is possible to utilize the strong wavelength dependence of the reflection phase to form high-Q cavities.

A schematic of a Fano cavity consisting of a GaAs/AlGaAs wafer including a DBR reflector and GaAs QWs and a separate $\text{Si}_3\text{N}_4/\text{SiO}_2$ grating reflector is shown in Fig. 4.2(a). Since there is no limitation on the choice of the QW material and the DBR, with only small changes to the parameters of the grating to shift the frequency of the resonance, the reflector can be used for monolayer MoSe_2 on top of a $\text{SiO}_2/\text{TiO}_2$ DBR, as shown in Fig. 4.2(a). For the latter example, the wavelength of the cavity resonance and the corresponding Q-factor obtained by RCWA simulations are shown in Fig. 4.2(b). In the simulation, the grating has a thickness of 283 nm, a period of 489 nm, and a duty cycle of 0.8, same as in Fig. 4.1(a). The wavelength and the linewidth of the discrete mode resonance of the grating are 731 nm and 8.5 nm. The asymmetry factor found by curve fitting with the Fano lineshape equation is $q = -0.36$, and the background reflection is $|r_B|^2 = 0.14$. Although the Q-factor of the resonance in the grating is only about 86, combined with the other reflector, the cavity Q-factor is much higher, as shown in Fig. 4.2(b). The cavity Q-factor is strongly dependent on the detuning between the resonance in the grating and the FP mode of the cavity. The latter one can be adjusted by changing the thickness of the air gap d between the two parts of the cavity. However, Q is larger than 10^3 for about an 80 nm range of d , well accessible in experiments.

With the QW exciton modeled as a Lorentz oscillator in the simulation, mode-splitting due to the strong-coupling between the exciton and the cavity mode can be found in both the GaAs and the MoSe_2 schemes. Fig. 4.3 shows the simulated absorption spectra as a function of d , and as a function of the in-plane wave vector component k_x (k_y) when the other component k_y (k_x) is zero. Here y denotes the direction parallel to the grating bars and x the direction perpendicular to them.

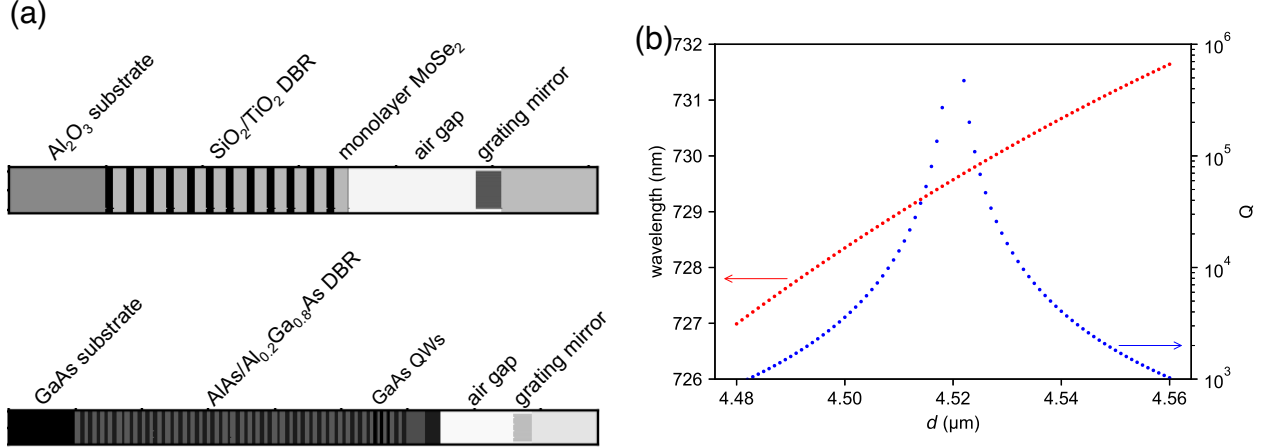


Figure 4.2: **Microcavity using the Fano interference-based reflector.** (a) Demonstration of the usage of the Fano interference-based reflector with a lower DBR and QWs of either MoSe₂ or GaAs. (b) The wavelength and Q-factor of the resonance of the cavity as a function of the thickness of the air gap d for the MoSe₂ design.

These results are obtained with exciton oscillator dielectric function $\varepsilon(E) = \varepsilon_\infty + \frac{B}{E_R^2 - E^2 - iE\Gamma}$ with realistic parameters $\varepsilon_\infty = 4.12$, $B = 1 \text{ eV}^2$, $\Gamma = 6 \text{ meV}$ [223]. The exciton energy E_R is currently adjusted in the simulation to be near the cavity resonance, but this should not affect the main result as the cavity resonance can be changed simply by proportionally scaling the thickness and period of the grating. Two branches, namely the LP and UP, are clearly visible and there is anti-crossing between them as the parameters change and the Rabi-splitting is about 7 meV. The splitting is smaller than conventional symmetric DBR cavities, partly because the electric field distribution of the cavity mode has a significant portion inside the grating layer, and partly because of the large air gap size. In the k_x spectra, the dispersions of the modes are strongly affected by the dispersion of the grating resonance. There is a cutoff line at small k_x , below which the first order diffraction into the SiO₂ is suppressed. In the k_y spectra, the dispersions resemble that of a DBR cavity.

Beyond the common features shared with other grating reflectors, it would be of interest to find out how the strong frequency dependence of the Fano cavity can affect polariton properties, especially when the nonlinearity of the exciton or polariton can effectively shift the detuning.

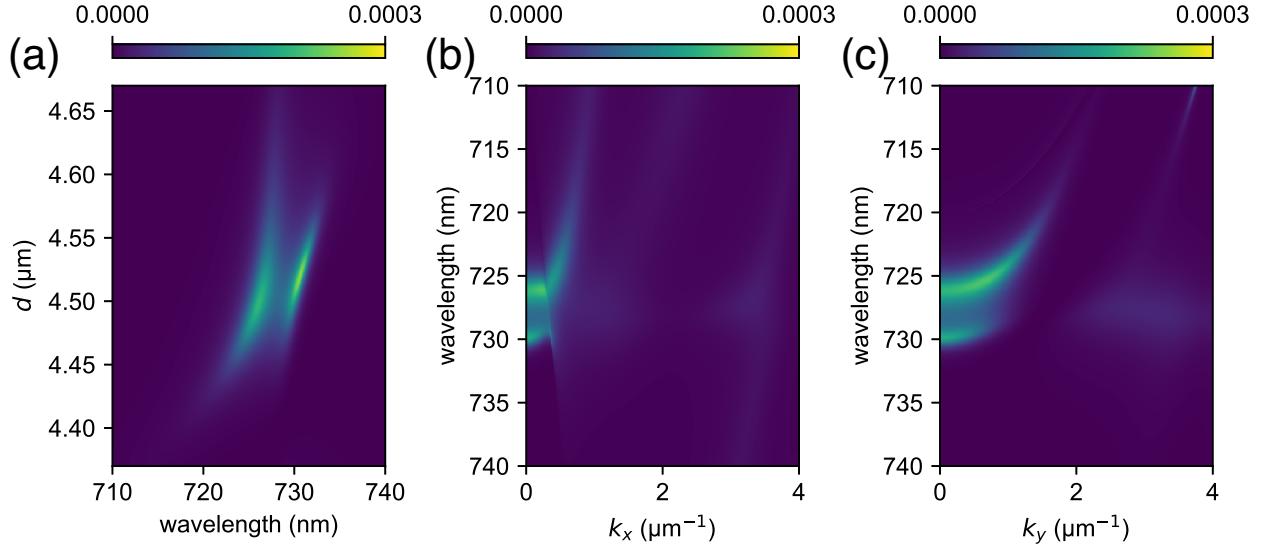


Figure 4.3: **Strong-coupling between exciton and photon with the Fano cavity.** Simulated absorption spectra of the cavity with MoSe₂ inside, as a function of (a) the air gap thickness, (b) k_x along $k_y = 0$, and (c) k_y along $k_x = 0$. Here x is the direction of the periodicity of the grating and the electric field is polarized in the y -direction.

4.3 Sample Characterization

Samples of the Si₃N₄ gratings with different periods and duty cycles have been fabricated by our collaborators Frédéric Peyskens and Muhammad Muneeb. However, the samples are different from the design in that the substrate is not SiO₂ but Si, with a 3.2 μm SiO₂ layer in between the grating and the substrate. The main characteristics of the grating reflector are not affected, other than small changes to the lineshape due to a background FP pattern from the thin SiO₂ layer. Fig. 4.4 shows two examples of the reflection spectra of these gratings, one for TE-polarization and another for TM-polarization. Comparing with simulation results, it can be seen that the Fano lineshape including the near-unity reflectivity peak is well reproduced. The series of line features with negative slope at larger k is from the FP pattern folded back due to the grating.

To build a cavity with the grating reflector and a DBR, an open cavity setup [109, 111, 222] is used. Fig. 4.5(a) shows the schematic of the setup. A piezo-driven stage that can move vertically is fixed inside a metal frame. The top surface of the metal frame has an opening at the center. One

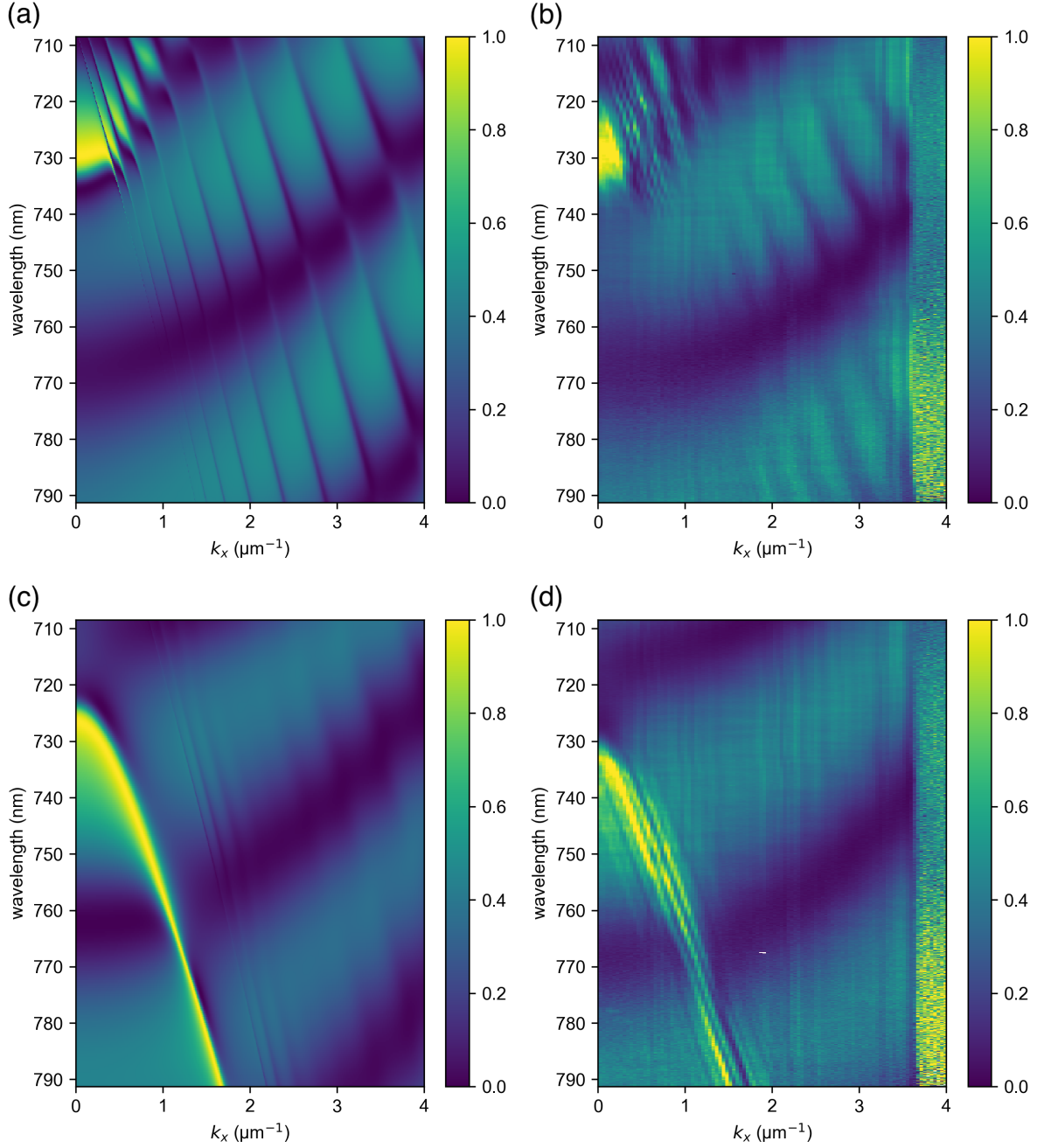


Figure 4.4: **Simulated and measured angle-resolved reflection spectra of the reflectors.** (a) and (c) are simulations and (b) and (d) are experimental data at room temperature. (a) and (b) are for electric field polarized in y and (c) and (d) are for electric field polarized in the x -direction. The period Λ and duty cycle η used in simulation or in the design of the fabricated devices are: (a) $\Lambda = 489$ nm, $\eta = 0.8$, (b) $\Lambda = 489$ nm, $\eta = 0.804$, (c) $\Lambda = 460$ nm, $\eta = 0.8$, (d) $\Lambda = 469$ nm, $\eta = 0.815$.

part of the cavity can be mounted on top of the stage and the other part can be mounted on top of the metal frame. The metal frame is then fixed to a three-dimensional piezo-driven stage and the entire assembly can be kept in a cryostat for measurement. By applying electric pulses to the piezo-stage inside the metal frame, the distance between the two parts of the cavity can be adjusted. By mounting the upper part on the metal frame with flexible aluminum tape, the upper part can self-adjust to become parallel with the lower part when they are close, which can help minimize the distance. The minimum distance achieved so far with this setup and two DBR reflectors is about $2.3\text{ }\mu\text{m}$, calculated from the cavity mode spacing. PL and reflection measurements can be done from the top side if the upper part has a transparent substrate. With two DBR reflectors, each with 12 pairs of $\text{SiO}_2/\text{TiO}_2$ on a sapphire substrate, and a MoSe_2 monolayer in between, the strong-coupling between the exciton and the cavity mode is observed, as shown in the PL spectra in Fig. 4.5(b-e).

The reflection spectrum of a Fano cavity consisting of the grating reflector and a $\text{SiO}_2/\text{TiO}_2$ DBR is measured. Because the cavity resonance is expected to be much narrower than the spectral resolution of the spectrometer, it shows up as a very small modulation in the spectrum. A differential spectrum is computed to show the resonance clearly: first measure a spectrum R_1 , slightly change the cavity length and measure again to get another spectrum R_2 , and then compute the normalized difference $dR = (R_1 - R_2)/R_1$. The cavity resonance will show up as two replicas of different sign slightly offset in wavelength, as shown in the example of an open DBR-DBR cavity in Fig. 4.6(a). The differential spectrum of the grating-DBR cavity is shown in Fig. 4.6(b), with an attempted reproduction in simulation in Fig. 4.6(c). Here, a small modulation of reflectivity is visible at $k_x = 0$ and near the expected wavelength, although having a much broader linewidth. The cavity resonance energy is expected to shift periodically versus the air gap thickness. Fig. 4.6(d) shows the line spectra at $k_x = 0$ for several different d , though the precise values of d have yet to be calibrated.

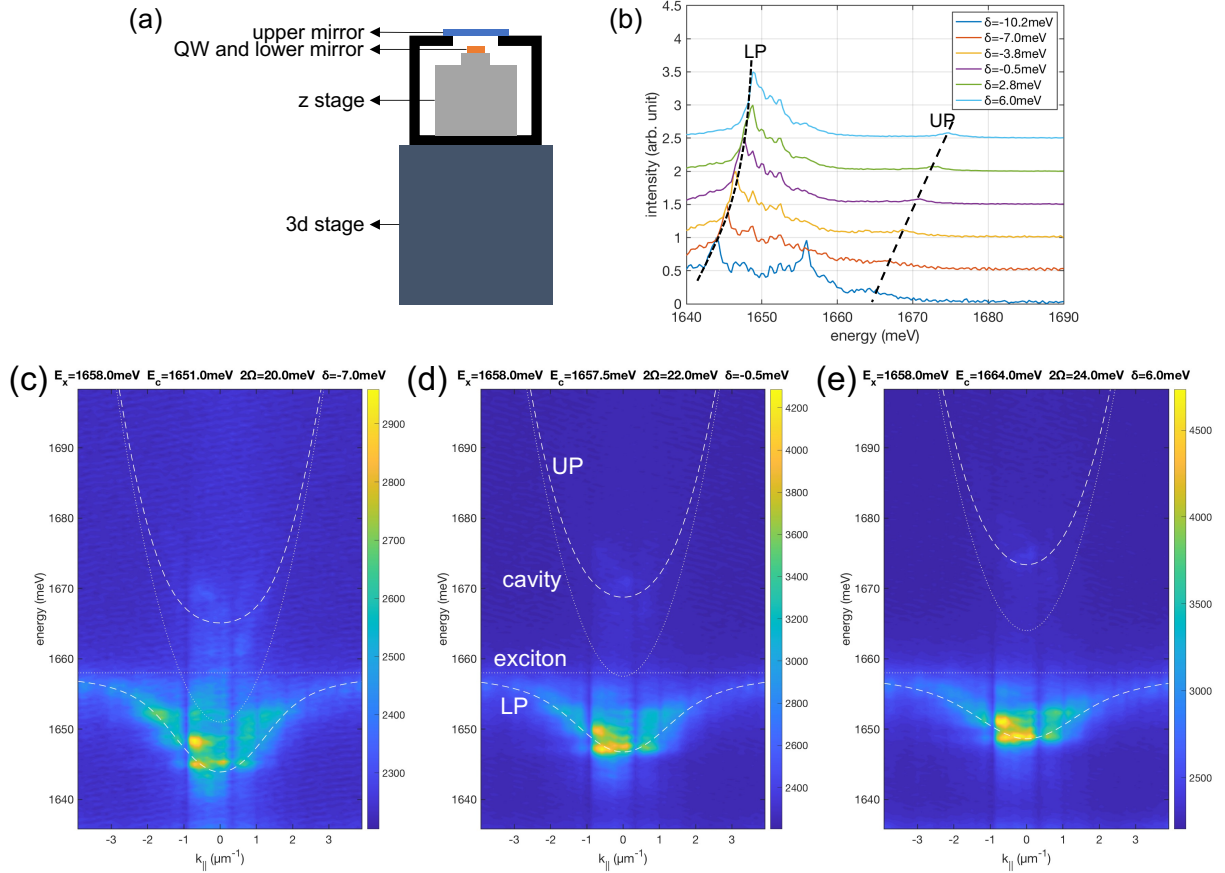


Figure 4.5: Open cavity experimental setup. (a) Schematic of the open cavity experimental setup. (b-e) PL spectra demonstrating strong-coupling between MoSe₂ exciton and a DBR-DBR cavity, at 4 K temperature. (b) Spectra at zero in-plane wave vector for various detunings. There is an offset between different detunings and the shift of the LP and UP peaks is marked with the black dashed lines. (c-e) Angle-resolved spectra at three different detunings. The estimated values of exciton energy E_x , cavity resonance energy at zero wave vector E_c , Rabi splitting 2Ω , and detuning δ are given in the subfigure titles. The estimated dispersion curves are labeled with dotted lines for the exciton and cavity resonances, and with dashed lines for LP and UP.

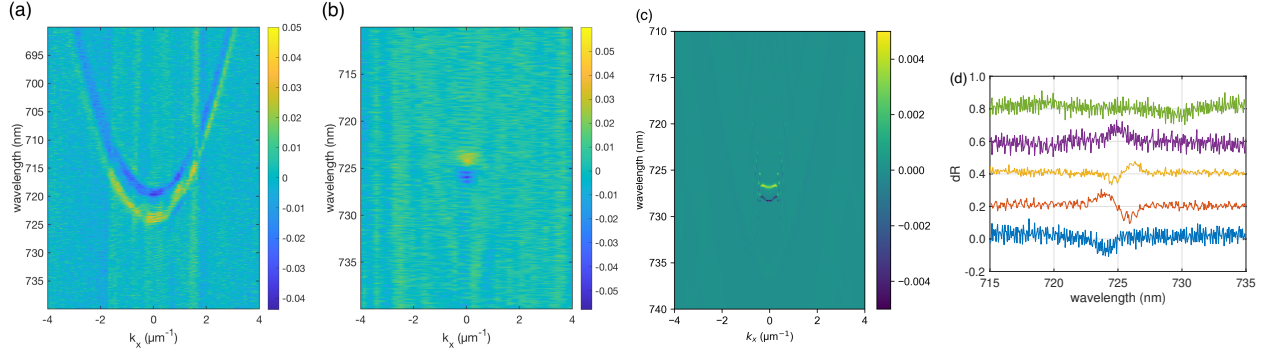


Figure 4.6: Differential reflection spectra of the Fano cavity. (a) Experimentally measured reflection spectrum of a DBR-DBR cavity as a reference, at 4 K temperature. The differential spectrum is the difference between two reflection spectra with slightly different air gap thicknesses, normalized to one of the spectra. Therefore, the resonance shows up twice with slightly different wavelength and opposite sign. (b) Experimentally measured reflection spectrum of a DBR-Fano mirror cavity, at room temperature. The grating is designed to have period $\Lambda = 483$ nm and duty cycle $\eta = 0.812$, and the electric field polarization is parallel to the grating bars. (c) Simulated reflection spectrum of a grating-DBR cavity with grating parameters $\Lambda = 489$ nm and $\eta = 0.8$. (d) Experimentally measured spectra integrated within $|k_{\parallel}| < 0.5 \mu\text{m}^{-1}$, while changing the air gap thickness in consecutive steps.

4.4 Future Work

In summary, this chapter introduces the application of a Fano-interference-based grating reflector in cavities for exciton-polariton studies. This type of reflector is expected to give a high Q-factor to the cavity and have strong wavelength dependence. Numerical simulations show strong-coupling between the exciton and cavity modes, even with larger cavity length. Preliminary experimental results are presented, such as the reflection spectra of the grating reflectors, which agree well with the numerical results. An empty cavity using this type of reflector is built with an open cavity setup, and the measured reflection spectra show features that may be from the expected cavity resonance.

As this is an unfinished work, there are many possible future directions. The immediate next steps can be improving the characterization of the empty cavity to verify the high Q-factor and to experimentally study the effect of the detuning between the cavity and grating resonances, or including a monolayer two-dimensional semiconductor inside the cavity to test if there can be strong-coupling. Alternatively, if the grating is fabricated on a transparent substrate such as fused

silica, it can be used with GaAs QW samples, which are currently better understood in terms of phenomena like polariton condensation, without involving other techniques like removing the substrate from the GaAs wafer. Conceptually, the grating reflector provides a good opportunity to engineer the dispersion of the cavity. With the TM-polarized design shown in Fig. 4.4(c-d), the resulting cavity mode has negative effective mass in the k_x direction. More complicated dispersions can be generated, for example, by tuning the grating parameters to overlap different photonic bands. Particularly, the interplay between the strong frequency dependence of the reflector and the nonlinearity of polaritons may enable new devices. Blueshifting of excitons or polaritons can lead to a change in the detuning and cavity loss, and may produce Q-switching-like behavior.

CHAPTER 5

Conclusion

In this dissertation, research on exciton-polaritons in semiconductor microcavities has been presented, including the identification of different phases or lasing mechanisms at high excitation density, and the usage of grating-based cavities to engineer the photon component of the polariton.

The first results are around the observation of a lasing transition that shows the predicted features of a polariton BCS phase. Since the early discoveries of BECs in the exciton-polariton system, which is open-dissipative and involves both bosonic and fermionic components, the studies of various possible phases of this system, including BECs, BCS states, and conventional photon lasing, as well as the crossover between them, have attracted both theoretical and experimental interest. The identification of these phases requires measurement of not only the cavity photon component, but critically also the electronic medium. By utilizing the polarization selectivity of a grating-based microcavity, the reflection spectra of the underlying excitons or electron-hole pairs are monitored simultaneously to a laser-like transition that occurs for the polaritons. The transition exhibits features in emission spectra that are typically associated with polariton BEC, such as a continuous blueshift before and after the threshold and a linewidth much smaller than both the amount of the blueshift and the difference between the cavity and low-density polariton modes. The reflection spectra measurements, however, show that the system is already above the Mott transition density, and the onset of lasing is accompanied by optical gain. Through comparison with a microscopic theory, this lasing state can be explained as a polariton BCS-like state where the electron-hole pairs no longer form bound excitons but only have weak k -space paring. In stark contrast, with an increase of the detuning between cavity and exciton, the lasing

transition happens only after significant broadening of the linewidth and blueshift close to the cavity energy, commonly associated with conventional photon lasing. Moreover, with another microcavity sample with regular DBR reflectors and ring-shaped optical pumping, the measured emission spectra show qualitatively similar trends suggesting that the BCS-like phase is potentially not limited to the specialized grating cavity system, although the reflection spectra measurements are not yet possible. In both types of samples, a clear boundary is not found between the two phases, suggesting a possible crossover.

Future work is still required to provide more direct evidence for the polariton BCS phase, such as the measurement of the pair-breaking gap energy, by THz spectroscopy or other methods. The conditions needed for the observation of each of the phases and the crossover between them are also unclear so far and call for more investigation. Particularly, a polariton BEC is not found in the current suspended grating-based microcavities.

From the technological aspect, the grating reflectors provide exceptional possibilities of engineering the photonic mode of the polariton, including its energy, decay rate, mode profile, polarization, and dispersion. Specifically, the grating-based cavities with a monolithic design are demonstrated experimentally to achieve high precision in tuning the energy and linewidth of the cavity resonance separately and defining localized modes with coupling between them. This capability may help the study of phase transitions in non-Hermitian systems, predicted to occur for exciton-polaritons, as well as realizing large scale devices such as lattices that utilize non-Hermitian physics. A different type of grating reflector relies on the Fano interference between an internal resonance and the background transmission to attain high reflectivity and strong frequency dependence. Numerical simulations are performed which show that this type of reflectors can replace a mirror of a polariton cavity and provide high cavity Q-factor and maintain strong-coupling. Experimentally, this cavity structure without quantum wells inside is tested with an open cavity setup with the two reflectors separated and their distance adjustable. Preliminary results show the signature of the resulting cavity resonance. This may address several issues found in the previous systems like mechanical stability or extra diffraction orders, to provide better experimental access to the different phases of

the polariton system.

With these experiments demonstrating basic functionalities of the grating-based cavities, future work can be performed either improving these techniques, such as increasing the tuning precision, or using them to solve real problems, such as selecting the energy and linewidth tuning parameters to observe different types of non-Hermitian phases, or using the frequency dependence of the mirror to modulate polariton lasing.

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