Strong Coupling and Magnetic Field Effects in Microcavity Light Sources

by

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To my mentor, Swami Sivapradananda (Saikateshda), who taught me the essence of 'simple living and high thinking' and used to quote: "manifest plainness, embrace simplicity, reduce selfishness, have few desires" and to my parents, for their unconditional love and support
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The realization of an exciton-polariton laser in the strong coupling regime promises an ultra-low threshold light source. In large bandgap semiconductors such as GaN or ZnO, it is possible to observe polariton lasing at room temperature because of the large exciton binding energy and small exciton radius, providing a very large critical temperature for polariton condensation, a necessity for high-temperature polariton lasing and Bose-Einstein condensation. A single GaN nanowire of appropriate diameter, grown defect-free on silicon with a very low background carrier concentration was embedded in a dielectric microcavity to realize a high temperature polariton laser. Relevant experiments were performed and polariton lasing was observed at 300 K with an optical pumping threshold power as low as $P_{th} = 92 \text{nJ/cm}^2$ and emission linewidth of 1.14 meV. Several control experiments confirm condensation towards $k_\parallel \sim 0$ and coherency of the emission. Conventional photon lasing in the same device with carrier population three orders of magnitude higher inversion is observed. Similar measurements were carried out with a single ZnO nanowire embedded in a dielectric microcavity. A very large Rabi splitting of 103 meV is obtained from the polariton dispersion and polariton lasing was observed at room temperature with a low threshold of 1.63 $\mu$J/cm$^2$, which corresponds to a polariton density an order of magnitude smaller than that for the Mott transition. The momentum distribution of the lower polaritons shows evidence of dynamic condensation and the absence of relaxation bottleneck. The polariton relaxation dynamics were investigated by time-resolved measurements, which showed a progressive decrease in the polariton relaxation time with increase in polariton density, to a value smaller than the radiative lifetime of polaritons at $k_\parallel = 0$.

Bose-Einstein condensation was also observed at room temperature in thermal equilibrium with the lattice. In the first experiment, the polariton scattering rates were engineered
by changing the detuning and the temperature (25 to 100 K corresponding to a change in detuning from -3.0 to 4.2 meV). The best results are observed at 85K with a positive detuning of 2.3 meV. The temperature of the lower polaritons $T_{LP}$ becomes almost equal to the temperature of the lattice $T_{latt}$ and the polariton relaxation time is smaller than the polariton lifetime at $k_{||} \sim 0$ by a factor of 2. In the second experiment, a spatial potential trap is formed in a 6.0 μm Al(Ga)N nanowire by varying the Al composition along its length during epitaxial growth. Excitation is provided at the Al(Ga)N end of the nanowire and polariton emission is observed from the lowest bandgap GaN region of the nanowire. Comparison of the results with those measured in an identical microcavity with an uniform GaN nanowire and having an identical exciton-photon detuning suggests evaporative cooling of the polaritons as they are transported across the trap in the Al(Ga)N nanowire. Measurement of the spectral characteristics of the polariton emission, their momentum distribution, first-order spatial coherence and time-resolved measurements of polariton cooling provide strong evidence of the formation of an equilibrium Bose-Einstein condensate in the GaN region of the nanowire at room temperature. An equilibrium condensate is not formed in the GaN nanowire dielectric microcavity without the spatial potential trap.

While most of the polariton laser systems have been optically pumped, an electrically injected polariton laser has not been experimentally demonstrated. In the present study, polariton lasing from a GaAs-based quantum well microcavity diode under electrical injection was observed for the first time. This is achieved by a combination of modulation doping of the wells, to invoke polariton-electron scattering, and an applied magnetic field to enhance the exciton-polariton saturation density. These measures help to overcome the relaxation bottleneck and to form a macroscopic and degenerate condensate. The experiments were performed at 30 K with an applied field of 7 Tesla. The threshold current density was 12 A/cm$^2$ and the minimum
measured linewidth was 0.6 meV. A second threshold, corresponding to photon lasing with population inversion, is also observed from the same diode at a current density ~3 orders of magnitude higher than the polariton lasing threshold. First order coherence measurements reveal formation of local condensates of size ~ 10 μm while polarization measurements show a buildup of both linear and circular polarization above the polariton lasing threshold.
Chapter 1
Introduction

1.1 Background

Semiconductor microcavities in the strong light-matter coupling regime have been attracting an ever growing interest for almost two decades. These structures are rather simply described, though: two semiconductor distributed Bragg reflectors (DBR) sandwiching an active layer made of bulk semiconductor, quantum wells (QWs) or quantum dots (QDs). Nevertheless, since the first measurement of the normal mode splitting between the excitonic resonance of the QW and a GaAs-based microcavity optical mode by C. Weisbuch et al. [1] in 1992, numerous publications have explored the extremely rich physics of these hybrid light matter waves.

The eigen modes of the system, the exciton polaritons (also called microcavity polaritons, in order to differentiate them from bulk exciton polaritons) are indeed a very unique feature. Part excitonic and part photonic, these two-dimensional quasi-particles are gathering advantages of their two components. From their light part, and a one-to-one coupling to the extra-cavity field, with conservation of in-plane momentum, energy and phase [2], exciton polaritons can be easily optically injected, manipulated, and detected. They also inherit the very small effective mass of the cavity photon. Through their excitonic part, polaritons can interact with each other, paving the way to non-linear and parametric processes. Eventually, exciton polaritons are composite bosons, as long as their excitonic oscillator strength is not screened.
The first demonstration of the potential of polaritons was with the parametric amplifier [3, 4, 5], which exploited at the same time the polariton-polariton interaction for scattering from an excited state into a pair of signal and idler states, and the bosonic stimulation to amplify a weak probe pulse resonant with the signal state. The strong non-linearities inherited from the exciton component were also exploited to demonstrate optical bistability, under resonant optical excitation of the polariton modes [6]. Under non-resonant excitation (i.e. creating hot electron-hole pairs above the photonic band gap of the DBR), several groups have reported stimulated relaxation mechanisms towards the bottom of the polariton dispersion [7, 8]. Along this line, polariton lasers, formed by bosonic final state stimulation, have emerged as a new source of coherent light with a threshold significantly lower than that of conventional semiconductor lasers. Polariton lasing was reported in GaAs-quantum well (QW) microcavities at a threshold density two orders of magnitude lower than that of photon lasing in the same device. In 2006, stimulated relaxation towards the macroscopically occupied lowest energy state was shown to be accompanied with the appearance of long range order within the polariton gas in a CdTe microcavity [9], allowing the recognition of the process as a phase transition to a non-equilibrium Bose-Einstein condensate (BEC). This phase transition differs from atomic condensates by its non-equilibrium nature and the loss of the polaritons as photons; hence the condensate needs to be constantly replenished by the laser pump. The other main difference, which is a major experimental advantage, comes from the very small polariton effective mass, allowing for the phase transition to be observed at cryogenic temperatures. Numerous investigations have followed the demonstration of polariton BEC, such as the observation of quantized vortices in the condensed phase [10, 11], and the demonstration of spontaneous coherent oscillations in coupled polariton condensates [12]. There are proposals to use polaritons
for the generation of single photons [17, 18], or for new kind of logical elements exploiting the polarization (or spin) degree of freedom of the polariton field [19, 20].

1.2 Motivation for Present Work

All the experiments performed in GaAs or CdTe based microcavities have been conducted at cryogenic temperatures, as the exciton binding energy $E_B$ is in the order of 10 meV in GaAs [21], and 25 meV in CdTe [22], to be compared to the thermal energy $k_B T$ at room temperature ($\sim 26$ meV) and liquid helium temperature ($\sim 0.5$ meV). However, while cryogenic temperatures are much more accessible than the nanokelvins required for atomic BEC, they hinder the implementation of practical room temperature devices. Therefore, the development of high exciton binding energy materials holds great promise for the practical implementation of polaritonic devices. In this respect microcavities based on GaN ($E_B \sim 40$ meV) [23], ZnO ($E_B \sim 60$ meV) [24, 25] or organic materials ($E_B \sim 0.2-0.5$ eV) [26, 27] have become very attractive. Moreover, these materials feature large exciton oscillator strength, which is essential for the robustness of the strong coupling regime. Polariton lasing at room temperature was reported in bulk organic microcavities, and in bulk and multiple-quantum-well (QW) GaN microcavities. However, in III-nitride microcavities, only a modest improvement was obtained compared to weakly-coupled GaN QW devices. Furthermore, in III-nitride bulk-microcavity and QW-microcavity structures the media and photon coupling is significantly reduced by a large built-in polarization field; the optical quality and reproducibility of the devices are often compromised by relatively large compositional inhomogeneities and structural defects.

It is worth mentioning that non-resonant pumping of the microcavity diodes has recently been demonstrated by means of electrical injection [28 - 30], which is another step towards
practical applications. However, electrically injected polariton lasers had not been demonstrated to the best of our knowledge and the reason cited for GaAs microcavities is the relaxation bottleneck encountered by the LPs when the dispersion characteristics in the LP branch transitions from exciton-like to photon-like. Finally, it is important to state that inversionless ultra-low threshold polariton lasers operating with electrical injection at room temperature will be useful in applications currently limited by energy consumption, such as ultrafast optical communication networks and medical diagnostics. As heat production has become a bottleneck for highly integrated devices and circuits, reducing the energy threshold of integrated semiconductor lasers has become ever more important.

### 1.3 Dissertation Overview

The present dissertation is an experimental study on strong coupling effects in microcavities containing a variety of materials, quantum confined systems and nanostructures. We have used GaN or ZnO nanowires to achieve exciton-polariton condensation and ultra-low threshold lasing at room temperature. These nanowires grow on Si and are relatively free of extended defects and have a very small polarization field and a high internal quantum efficiency. We employed a single nanowire strongly coupled to a dielectric microcavity – the emitter is just 1 μm$^2$ in area, but it still gives room temperature lasing at a threshold current density which is three orders of magnitude lower than any other existing laser.

Dynamic Bose-Einstein condensation in a single GaN nanowire-dielectric microcavity has been demonstrated by engineering the exciton-photon detuning and the temperature and the system can attained a state close to thermal equilibrium at a temperature of 85K with a positive detuning of 2.3 meV. The temperature of the lower polaritons $T_{LP}$ becomes almost equal to the
temperature of the lattice $T_{\text{latt}}$ and the polariton relaxation time is smaller than the polariton lifetime at $k_{\parallel} \sim 0$ by a factor of 2. Also, Bose-Einstein condensation at room temperature has been demonstrated in a graded Al(Ga)N nanowire-microcavity employing a potential trap. With the nanowire selectively excited at the high bandgap Al$_{0.05}$Ga$_{0.95}$N end, the exciton-polaritons which are more photon-like relax to lower energies by scattering as they drift and diffuse to the central GaN region where they are less photon-like. The polariton-phonon scattering rate will gradually increase due to the increasing exciton-photon detuning in the microcavity. At the same time the higher energy polaritons (at higher $k_{\parallel}$ values) will be lost as photons, thereby providing evaporative cooling.

Electrically injected polariton lasing has also been demonstrated in a multi-quantum well microcavity with modulation doping in the barrier region in the presence of a magnetic field. Modulation doping introduces electron-polariton scattering that has a large matrix element and can take away more energy for a given momentum, since the electron effective mass is less than the exciton mass. Thus LP relaxation to lower energy state requires fewer scattering events and the bottleneck is reduced if not suppressed. Application of a magnetic field reduces the exciton Bohr radius and increases the Rabi splitting. Thus the strongly-coupled system becomes more robust and the exciton saturation density increases by at least an order of magnitude, allowing non-linearity due to polariton-polariton scattering and consequent quantum degeneracy to appear in the lowest energy state before the system can transition over to the weak coupling regime. The device reports electrically injected polariton lasing, for the first time in a solid-state system.
1.4 Thesis Organization

The thesis describes an experimental study on the application of exciton-polaritons and strong coupling effects to ultra-low threshold lasers. The dissertation is organized as follows: In Chapter 2 I review the basic physics of semiconductor microcavities. In Chapter 3, I have described the experimental measurement and sample and device fabrication techniques. In Chapter 4 I present experimental results of room temperature polariton lasers based on a single GaN or ZnO nanowire embedded in a dielectric microcavity. In Chapter 5 I describe techniques to realize a practical electrically injected polariton laser. The investigation includes studying the effects of magnetic field and modulation doping in the active region. In Chapter 6, I describe the attainment of an equilibrium polariton BEC at 85K by varying the detuning and temperature in a single GaN nanowire-microcavity device and also at room temperature by designing a potential trap with a graded Al(Ga)N nanowire. The results are summarized in Chapter 7, with suggestions for future work. Details of important calculations, simulations, and theories are attached in the Appendix.
Chapter 2

Microcavity Exciton-Polaritons

The existence of normal-mode coupling between light and elementary electronic excitations (excitons) in bulk semiconductors was predicted by Hopfield [31] in 1958. In Hopfield’s theory, because of a periodic energy exchange interaction between photons and excitons due to translation invariance symmetry, the elementary excitations of a bulk semiconductor are mixed exciton-photon states called exciton-polaritons. In this chapter the different elements which constitute a semiconductor microcavity and how strong light-matter coupling leads to the formation of exciton polaritons are described.

2.1 Excitons in Semiconductors

2.1.1 Bulk Excitons

In direct gap semiconductors, the promotion of an electron to the conduction band leaves a hole in the valence band. The Coulombic interaction between the oppositely charged electron and hole leads to the formation of a bound state called an exciton. This elementary excitation appears as a new line in the absorption spectrum, separated from the band gap $E_g$ by the exciton binding energy $E_b$. The exciton can be seen as a hydrogenic quasi-particle, whose binding energy is analogous to the hydrogen Rydberg energy, although much smaller [32]. Semiconductor excitons are generally of the Wannier type, meaning that the electron-hole relative wavefunction is delocalized over many lattice sites. The exciton dispersion is given by:
\[ E_X(k) = E_g - E_b + \frac{\hbar^2 k^2}{2m_X} \]  

(2.1)

where \( \hbar k \) is the momentum of the exciton center of mass. The exciton effective mass \( m_X \) is given by the combination of electron and hole effective masses \( m_e \) and \( m_h \) as:

\[
\frac{1}{m_X} = \frac{1}{m_e} + \frac{1}{m_h}
\]

(2.2)

### 2.1.2 Excitons in Quantum Wells

Semiconductor quantum wells (QWs) consist of a thin layer of semiconductor material inserted between another semiconductor material of larger band gap, e.g. 10nm of In\(_{0.1}\)Ga\(_{0.9}\)As inserted in bulk GaAs. Excitons are then confined in one direction (that we call \( z \)), forming a two-dimensional system in the QW plane. Localization of electrons and holes in the QW leads to an enhanced exciton binding energy [33,34]. Another consequence of the QW confinement is that a QW exciton, which has a given momentum in the QW plane, is coupled to a continuum of photons which have the same in-plane component of momentum. It is therefore possible to compute an intrinsic exciton lifetime \( \tau_X \) using the Fermi’s golden rule [35] and the corresponding homogeneous linewidth \( \gamma_X = \hbar / \tau_X \). The latter is proportional to the exciton oscillator strength \( f_{osc} \), which characterizes the strength of the excitonic transition (or the probability for a photon of

![Fig.2.1. Schematics of a quantum well. A layer of InGaAs (5%) is inserted between two GaAs layers with larger band gap, resulting in a quantum well for excitons. The dashed purple lines represent the first electron and heavy-hole energy levels in the conduction and valence bands respectively.](image)
2.1.3 Excitons in Quantum Dots

Quantum dots are dimensionless semiconductor nanocrystals whose excitons are confined in all three spatial dimensions. When the size of the material is smaller than the critical characteristic length, called the “exciton Bohr radius”, the crowding of electrons leads to the splitting of the original energy levels into smaller ones with smaller gaps between each successive level. The exciton Bohr radius is larger than the bulk Bohr radius due to the effect of dielectric screening and the influence of periodic lattice structure of the crystal. The exciton confined in a QW (or QD) can be modeled as a canonical particle in a box, while the constituent e and h can be likened to a hydrogen atom in the Bohr model, where the hole of positive charge takes the place of the atomic nucleus. Thus, the energy levels of the exciton can be represented as the solution to the particle in a box at the ground level \( n = 1 \) using the reduced mass of the Bohr model. Using this model, the quantum dot exciton binding energy and the Bohr radius are:

![Fig.2.2. (Left) InAs Quantum dots on GaAs (001), 100nm x 100nm. (Right) Measured electroluminescence intensity from the a single QD as a function of emission photon energy (vertical axis) and drive current (horizontal axis). Sharp line emission (marked \( X \) and \( X_2 \)) is seen arising from a single quantum dot in the structure [36].](image)
2.1.4 Excitons in GaN/ZnO Nanowires

Since a majority of this work involves GaN and ZnO nanowires-based light emitters, it will be useful to comment about the exciton structure in these nanowires. The nanowires have diameters ranging from 50 nm to 200 nm, and hence the exciton localization length is much longer than the thermal de Broglie wavelength and its eigenstates can be approximated by a 1D continuum of states. Thus the material can be treated as a bulk semiconductor and the exciton

\[ E_X = -\frac{1}{\epsilon_r^2} \frac{m_X}{m_e} R_y \]  

(2.3)

\[ a_B^* = \epsilon_r \frac{m_X}{m_e} a_B \]  

(2.4)

**Fig. 2.3.** (Left) Room temperature luminescence from GaN nanowires shows bulk optical characteristics. Due to high surface-to-volume ratios, nanowires exhibit higher extraction efficiencies. (Right) Photoluminescence spectrum at 25K, revealing free and donor bound excitons [37].
luminescence coincides with the band-edge of the material (see Fig. 3.3). The wurtzite crystalline structure gives rise to three free exciton transitions, \( X_A \), \( X_B \) and \( X_C \) (see Fig. 3.3), of which the \( X_A \) and \( X_B \) excitons are purely polarized perpendicular to the c-axis whereas the \( X_C \) exciton is strongly polarized parallel to the c-axis, as has been obtained from interband momentum-matrix calculations. One inherent advantage of these nanowires, which can be considered one-dimensional systems, over planar systems is the ability to relieve strain energy by means of lateral relaxation. The absence of dislocations leads to drastically reduced nonradiative exciton recombination rates and hence reduced exciton linewidth.

2.2 Cavities: Light Confinement

2.2.1. Distributed Bragg Reflectors

A distributed Bragg reflector (DBR) is a periodic succession of layers of two materials with different refractive indices. Reflection by the structure is based on an interferometric principle: the optical thickness of every layer is matched to be a quarter of the desired wavelength \( \lambda_0 \), so that reflections from every interface constructively interfere. The result is the opening of a

![DBR reflectivity spectrum](image)

Fig 2.4. DBR reflectivity spectrum computed using the transfer matrix method for \( \lambda_0 = 900 \text{ nm} \) with alternating layers of AlAs/GaAs. The layer thickness corresponds to \( \lambda_0/4n \), where the refractive indexes have been taken to be \( n_{\text{GaAs}} = 3.5 \) and \( n_{\text{AlAs}} = 3 \).
photonic band gap, centered at $\lambda_0$, within which propagation is forbidden in the direction perpendicular to the layers (called the z direction). It can be seen as a one-dimensional photonic crystal, and results in a very high quality reflector. Figure 1.4 shows typical reflectivity spectra for alternating layers of refractive indexes corresponding to those of GaAs and AlAs. This example shows that increasing the number of GaAs/AlAs pairs increases the reflectivity in the photonic band gap.

2.2.2. Microcavity

When two DBRs are placed face to face around a spacer, they form a Fabry-Pérot resonator. In this structure, electromagnetic waves are confined in the direction normal to the DBR faces (z-direction), but are still free to propagate in the in-plane directions (x and y-directions), forming thus a two-dimensional system. The only optical modes supported by the cavity are those satisfying the boundary conditions of the resonator, and only those modes will be transmitted through it. In this way, the microcavity spacer acts as an impurity in the one-dimensional photonic crystal [38]. A typical reflectivity spectrum is presented in Figure 2.5, for a
λ-cavity. The finite width of the cavity mode $\gamma_C$ is proportional to the cavity photon escape rate, or inversely proportional to the photon lifetime in the cavity $\Gamma_C$. The quality factor $Q$ of the cavity depends therefore on the quality of the mirrors, and is given by $Q = E/\gamma_C$, where $E$ is the emission energy. Depending on the area of measurement, the measured cavity linewidth $\gamma_C^*$ can be inhomogeneous due to thickness variations of the cavity spacer and DBRs [39].

2.2.3. Photon dispersion

There is a dependence of the cavity resonance on the angle of incidence of the field with respect to normal incidence. The cavity imposes a quantization of the wave vector in the growth direction, which we chose to be $z$, so let us write $\vec{k}$ in terms of its component $k_z$ along $z$ and $\vec{k}_\parallel = k_x \vec{x} + k_y \vec{y}$ in the plane parallel to the layers $(x, y)$: $\vec{k} = \vec{k}_\parallel + \vec{k}_z$ (see Fig.2.6). Thus one can write:

$$E = \frac{hc}{n_{cav} \lambda} = \frac{\hbar c k}{n_{cav}} = \hbar c \sqrt{k_z^2 + k_\parallel^2}$$ (2.5)

One can deduce a dispersion relation

$$E (k_\parallel) = \sqrt{E_z^2 + \frac{\hbar^2 c^2 k_\parallel^2}{n_{cav}^2}}$$ (2.6)

This equation is parabolic-like around $k_\parallel = 0$, and thus one can attribute an effective mass to the cavity photon, deduced from the curvature of equation 1.6 at $k_\parallel = 0$:

$$m_{cav} = \frac{n_{cav} \hbar}{\lambda c}$$ (2.7)
For a resonance wavelength of 800 nm, equation 1.7 yields a mass of $m_{\text{cav}} = 1.4 \times 10^{-36}$ kg. The photon dispersion curvature around $k_\parallel = 0$ is thus $10^4$ times bigger than the exciton’s curvature. This gives a mass of the order of $10^{-5}$ times the free electron’s mass, or $10^{-4}$ times the exciton’s mass. Finally, there is a one-to-one correspondence between the incidence angle $\theta$ and the in-plane wavenumber $k_\parallel$:

$$k_\parallel = n_{\text{cav}} \frac{2\pi}{\lambda} \tan\left[\sin^{-1}\left(\frac{\sin\theta}{n_{\text{cav}}}\right)\right] \approx \frac{2\pi}{\lambda} \theta, \text{ for } k_\parallel \ll k_z$$

(2.8)

2.3 Strong Light-Matter Coupling in Microcavities

2.3.1 Principles

The study of light-matter interactions with atoms in a cavity led Jaynes and Cummings to predict the existence of normal-mode splitting between the cavity mode and the atoms in the case of strong interactions between them [40]. As Hopfield did, but in a different picture, Jaynes and Cummings compared semi-classical and quantum mechanical approaches to study the coupling between an atom and a cavity mode [40]. They showed the existence of two interaction regimes - a weak-coupling regime, that could be treated by a perturbation theory with the application of the Fermi golden rule, and a strong-coupling regime, with the appearance of normal modes as atom-field quasiparticles. This work gave birth to the cavity electrodynamics research field.
The first clear evidence of normal-mode splitting was given at the end of the 1980’s by Raizen, in the group of J. Kimble [41]. In their experiment, they plotted a clear anticrossing behavior of the mode frequencies as a function of the cavity detuning. As we will see, this behavior is characteristic of normal-mode splitting.

A hamiltonian analog to the Jaynes-Cumming Hamiltonian can be written to describe exciton-photon interaction in a microcavity. Excitons are regarded as bosons and we assume a density low enough to avoid any contribution from their fermionic nature [42]. We define $b_{k||}^+ (b_{k||})$ as the creation (annihilation) operator of an exciton with in-plane wavevector $k||$. We also define $a_{k||}^+ (a_{k||})$ as the creation (annihilation) operator of a photon with in-plane wavevector $k||$. The interaction Hamiltonian is given by:

$$H_0 = \sum_{k||} E_C(k||) a_{k||}^+ a_{k||} + \sum_{k||} E_X(k||) b_{k||}^+ b_{k||} + \sum_{k||} \hbar \Omega (a_{k||}^+ b_{k||} + b_{k||}^+ a_{k||})$$

(2.9)

where $\hbar \Omega$ is the coupling energy. The coupling factor is given by [42]:

$$\Omega = \sqrt{\frac{2\pi}{\varepsilon_r} \frac{1}{4\pi \varepsilon_0 m \ell_{eff}} e^2 N f}$$

(2.10)

where $\varepsilon_r (\varepsilon_0)$ is the relative (vacuum) permittivity, $e(m)$ is the electron charge (mass), $N$ is the oscillator density coupled to the cavity mode, and $\ell_{eff}$ is the effective cavity length. In matrix form, the Hamiltonian reads:

$$H_0 = \begin{bmatrix} E_C(k||) & \hbar \Omega \\ \hbar \Omega & E_X(k||) \end{bmatrix}$$

(2.11)

The energies of the eigenstates of the Hamiltonian are:

$$E_{LP,UP}(k||) = \frac{E_C(k||) + E_X(k||)}{2} \pm \frac{1}{2} \sqrt{(\delta_{k||})^2 + 4|\hbar \Omega|^2}$$

(2.12)
where

\[ \delta_{k||} = E_C(k||) - E_X(k||) \]  

is the detuning between the cavity mode energy and the quantum well exciton energy. This quantity will often be used in the coming pages. When simply mentioning the detuning from now we will refer to its value at \( k|| = 0 \): \( \delta_{k||=0} \).

In the absence of coupling (\( \hbar \Omega = 0 \)), the eigen states of this system are just the uncoupled photon and exciton. Otherwise, the system is in a “strong coupling regime” and a splitting between \( E_X(k = 0) \) and \( E_C(k = 0) \) occurs. These are no longer the eigen states of the system, but rather the energies available to the system are new eigen states \( E_{LP} \) and \( E_{UP} \). The new eigen states of the system are the upper and lower polaritons. Fig.2.7 shows the polaritons’ dispersion curves, compared to the uncoupled exciton and photon dispersion curves.

Fig. 2.7. Polariton dispersions for \( \delta_{k||=0} = 0 \). Bare exciton and photon modes are indicated with dashed lines. The eigenmodes are the lower polariton (LP) and upper polariton (UP) branches, indicated with solid lines.
2.3.2 Hopfield Coefficients

The polaritons’ operators are written as follows in the original photon-exciton basis:

\[
\begin{pmatrix}
    p_k \\
    q_k
\end{pmatrix} = \begin{pmatrix}
    X_k & C_k \\
    -C_k & X_k
\end{pmatrix} \begin{pmatrix}
    b_k \\
    a_k
\end{pmatrix}
\] (2.14)

where \(p_k^\dagger, p_k\) and \(q_k^\dagger, q_k\) are the creation and annihilation operators for the lower and upper polariton, respectively, and \(X_k\) and \(C_k\) are the Hopfield coefficients, introduced by Hopfield in 1958 [31]. The exciton and photon fractions are defined by:

\[
|X_{k||}|^2 = \frac{1}{2} \left( 1 - \frac{\delta_{k||}}{\sqrt{(\delta_{k||})^2 + 4|\hbar\Omega|^2}} \right), \quad |C_{k||}|^2 = \frac{1}{2} \left( 1 + \frac{\delta_{k||}}{\sqrt{(\delta_{k||})^2 + 4|\hbar\Omega|^2}} \right)
\] (2.15)

with the unitary condition \(|X_{k||}|^2 + |C_{k||}|^2 = 1\). Finally the diagonalized Hamiltonian can be obviously written in the polariton basis:

\[
H_0 = \sum_{k||} E_{LP}(k||) p_{k||}^\dagger p_{k||} + \sum_{k||} E_{UP}(k||) q_{k||}^\dagger q_{k||}
\] (2.16)

2.3.3 Strong Coupling Conditions

Both excitons and photons have finite lifetimes: excitons are in a 1s state and recombine at the rate \(\gamma_X\); photons escape through the cavity mirrors at the rate \(\gamma_C\). These quantities are introduced in the hamiltonian as imaginary components of the respective exciton and photon energies, written as:

\[
E_X' = E_X - i\gamma_X, \quad E_C' = E_C - i\gamma_C
\] (2.17)

Inserted in Eqs. (1.12), this results in an effective correction of the coupling strength \(\Omega\) [43]. At \(k = 0\) and zero detuning, \(\Omega\), becomes
yielding the following condition for the strong-coupling regime

$$\Omega > \gamma_C, \gamma_X$$

This relation can be intuitively understood as follows. Brought close to resonance, excitons and photons exchange energy at a rate related to $\Omega$. During their lifetime, excitons and photons should experience several energy exchanges for strong coupling to occur. In other words, the energy exchange rate is larger than both decay rates in this regime. The strong-coupling condition can be formulated as $\Omega' > \gamma_{LP}, \gamma_{UP}$, showing that experimentally the linewidth of the polariton resonances should remain smaller than their energy separation at zero detuning to observe strong coupling.

### 2.3.4 Polariton Mass

For small in-plane wavevectors polariton dispersions can be approximated by parabolas, allowing us to define an effective mass for the polaritons. Neglecting the curvature of the excitonic dispersion, effective masses for the lower and upper polaritons are given for small $k_{\parallel}$ by

$$m_{LP} = \frac{m_{cav}}{|C|^2}, \quad m_{UP} = \frac{m_{cav}}{|X|^2}$$

We can therefore calculate the polariton effective mass for a GaAs $\lambda$-cavity, with $\lambda_0 = 835$nm, for zero exciton-photon detuning: $m_{LP,UP} = 2m_{cav} = 0.4\text{meV} \cdot \text{ps}^2 \cdot \mu\text{m}^{-2}$. This effective mass is four orders of magnitude smaller than the exciton effective mass and five orders of magnitude smaller than the free electron mass.
2.3.5 Experimental Signatures of Strong Coupling: Anti-crossing

In order to experimentally characterize the strong coupling regime, one needs to observe the splitting between the upper and lower polaritons at resonance between the exciton and the cavity photon. Several experiments are possible, based on reflection or transmission of an incident electromagnetic beam in order to probe the optical density of states, or based on photoluminescence experiments. We will develop these methods in chapter 3. In all the methods, spectra are taken at various positions of the cavity, and in case of strong coupling, two modes can be seen around the resonance energy instead of one degenerate mode. Nevertheless, this single observation could be attributed to many phenomena other than strong-coupling, such as the observation of the light-hole exciton for example. Certainty of the presence of the strong coupling regime can be gained only through observation of an anti-crossing behavior.

2.3.5.1 Momentum space anti-crossing

As we will see in chapter 4, it is possible to access the dispersion curve of the system through angle-resolved luminescence or Fourier-space imaging, thanks to its 2D character. An anticrossing behavior at a certain value of $k>0$ and at the symmetric value $-k$, under negative detuning [44], is characteristic of strong coupling.

2.3.5.2 Temperature tuning

It is possible to tune the exciton’s energy through temperature variation. This method is usually employed when it is not possible to vary the cavity mode’s energy, in order to demonstrate strong-coupling of a single quantum dot with a cavity or when the UP is pushed into the continuum states of excitons due to the large vacuum Rabi in wide bandgap semiconductors.
2.3.5.3 Anti-crossing through real-space cavity mode tuning

By suitable fabrication of the sample, one can introduce a linear variation of the cavity mode energy along the sample’s radius. This allows us to tune the detuning at $k_\parallel=0$ between the cavity and exciton energy. If, between $\delta << 0$ and $\delta >> 0$, one observes an anticrossing behavior of the resonances energies as illustrated on Fig.2.8, it is a signature of strong coupling. This method

![Graph showing temperature dependent dispersion characteristics](image)

Fig.2.8. Temperature dependent dispersion characteristics obtained from a single ZnO nanowire embedded in a dielectric microcavity. The anti-crossing happens at $T = 287K$ [45].

![Graph showing energy levels](image)

Fig.2.9. The energies of the upper ($E_{UP}$) and lower ($E_{LP}$) polaritons are calculated for a constant exciton energy $E_X = 1484.5$ meV and a varying cavity energy $E_C$, with a Rabi splitting $\Omega = 3.5$ meV. The anticrossing behavior is characteristic of the strong-coupling regime.
was used for the first experimental demonstration of strong-coupling in a semiconductor microcavity by Weisbuch et al. [1].

### 2.3.6 Strong to Weak Coupling Transition

One can now give some objective criteria for the recognition of the transition from strong to weak coupling regimes. Starting from a strong-coupling regime situation, the strong-coupling can be lost for two reasons:

**If \( \gamma_c \) and/or \( \gamma_X \) increase:** Concretely the most probable situation is an increase of the exciton’s linewidth \( \gamma_X \), due to an increase in temperature or because of screening due to increased injection.

**If \( \Omega \) decreases:** \( \Omega \) is the coupling strength between the excitons and the photons and depends on the exciton’s oscillator strength. The strong-coupling regime can therefore be lost by saturation of the excitonic oscillator strength, or in other words by a loss of the bosonic character of excitons, and in even simpler terms, by loss of the excitons in favor of electron-hole pairs.

### 2.4 Condensation of Microcavity Polaritons

#### 2.4.1 Validity of Bosonic Nature of Excitons

Excitons, like atoms, are bosons composed of bound fermions. A key difference between bosonic atoms and excitons, however, is the binding energy of the constituent fermions. For example, in hydrogen the binding energy is 13.6 eV, where it is only on the order of several meV for exciton in III-V semiconductors. Since an exciton state is spread over a high number of electronic states in a crystal, Pauli’s exclusion principle does not forbid a high occupancy of an exciton state, as long as the system is at relatively low exciton density. At high exciton densities, the occupation of the electronic states increases, and Pauli’s exclusion principle makes it more
and more difficult to create new excitons. The critical density over which the exciton cannot be considered as a boson anymore is $\sim 1/a_B^2$ [46], where $a_B$ is the exciton’s Bohr radius. In the case of an In$_{0.05}$Ga$_{0.95}$As in GaAs quantum well, the Bohr radius is approximately $a_B \sim 12\text{nm}$ [47]. This yields a critical density $n \sim 7 \times 10^{11} \text{cm}^{-2}$, consistent with experimental values of about $n \sim 10^{11} \text{cm}^{-2}$.

### 2.4.2 Kinetics of Polariton Condensation

As elementary excitations of the microcavity system, polaritons are most conveniently created by a laser pump pulse, after which they relax and, under appropriate conditions, accumulate at least partly in the ground state of the LP branch before completely decaying. In order to study a spontaneous phase transition, pumping should be incoherent, so that there are no phase relations between the pump light and the condensate. The main interactions which drive the relaxation and eventually the condensation stem from exciton-phonon interactions and Coulomb interactions between excitons. The exchange Coulomb interactions dominate over the direct Coulomb interactions between two excitons and give rise to a repulsive interaction between two polaritons with an identical spin. This interaction provides the fastest scattering mechanism which thermalizes the polariton gas, but the temperature of the gas can only be lowered by coupling to the cold acoustic phonon reservoir.

The phonon scattering rate is given by:

$$W_{k\rightarrow k'}^{\text{phon}} = \frac{2\pi}{\hbar} \left| \frac{\hbar |q|}{2 \rho\omega L_{QW}} \left( D_e f_e(|q|) - D_h f_h(|q|) \right) k_x k'_x \right|^2 \frac{m_P(k') A}{\pi \hbar^2} N_{\text{phn}}(E_{\text{phn}})$$ (2.21)

where $q = k' - k$ and the phonon number $N_{\text{phn}}(E_{\text{phn}}) = \frac{1}{\exp(|E_{\text{phn}}|/k_BT) - 1} + 1/2 \pm 1/2$, where the positive/negative term is used to describe the emission/absorption processes. The various
parameters used in the formula are as follows: $D_{e(h)}$ is the deformation potential for electrons (holes), $\rho$ is the density of the active region, $A$ is the cross-sectional area, $c_s$ is the speed of light in the medium, $L_{QW}$ is the thickness of the quantum well, $m_{LP}(k)$ is the lower polariton mass at wave vector $k$, $x_k$ is the exciton Hopfield coefficient at wave vector $k$, $q$ is the electron charge and $\hbar$ is the reduced Planck constant. The in-plane exciton-phonon wavefunction overlap is given by:

$$I_{e,(h)}^\parallel(q) = \left(1 + \left(\frac{m_{h,e}q a_b^{2D}}{2m_{exc}}\right)\right)^{-3/2}$$ (2.22)

where $a_b^{2D}$ is the exciton Bohr radius and $m_{exc}$ is the exciton mass. The polariton mass is given by the exciton and photon mass, weighted by their relative fraction:

$$\frac{1}{m_{LP}(k)} = \frac{|x_k|^2}{m_{exc}} + \frac{|c_k|^2}{m_{cav}}$$ (2.23)

where $c_k$ is the photon Hopfield coefficient and $m_{cav}$ is the cavity photon mass. The polariton-polariton scattering rate is given by:

$$W_{k\rightarrow k'}^{pol} = \frac{2\pi}{\hbar} |x_kx_{k'}x_qx_{q'}|^2 \frac{(a_b^{2D})^2}{A} E_b \left(\frac{m_{LP}(k')A}{\pi\hbar^2}\right)^2$$ (2.24)

### 2.4.3 Lifetime Variation in Momentum Space

The lifetime of excitons is on the order of nanoseconds. Polaritons, however, are a mix of excitons and photons, making their lifetime much shorter than the exciton lifetime. In the microcavity, the polariton lifetime is very much limited by the quality of the DBRs. The higher the reflector quality, or Q-factor, the longer the photon stays in the cavity, and consequently the longer the polariton lifetime. The value of the Q-factor, $Q = \lambda_c/\Delta \lambda_c$, where $\lambda_c$ is the cavity
resonance, is equivalent to the average number of round trips of a photon inside the cavity. For our single nanowire-dielectric microcavity sample, with cavity resonance $\lambda_c = 365$ nm and resonance width of $\Delta \lambda_c = 0.6$ nm, $Q \sim 600$ and the estimated photon lifetime, $\tau_{\text{cav}}$, is $\sim 0.6$ ps. Quantitatively, the polariton lifetime $\tau_{\text{pol}}$ depends on the fraction and lifetime of each individual photon and exciton fractions, according to

$$\tau_{\text{pol}} = \frac{|C|^2}{\tau_{\text{cav}}} + \frac{|X|^2}{\tau_{\text{exc}}}$$  \hspace{1cm} (2.25)

Hence, polaritons in the ground state have a much shorter lifetime compared to polaritons at higher energy states. This is detrimental to creating a condensate in thermal equilibrium. Fortunately, absolute time scales are irrelevant in the BEC transition. What matters is the thermalization time compared to the lifetime of the polaritons, and the thermalization time can be in the sub-picosecond range. Most importantly, in wide-bandgap semiconductor microcavities, the polariton-phonon rates can be very high since they operate at room temperature, making it easier to achieve condensation in these systems.

Knowledge of the lifetime variation in momentum space will be important later for converting from raw PL intensity to occupation number. For a constant number of polaritons, the shorter the lifetime of a polariton, the more intense the PL, since in a given detection time more particles decay from a particular state. The PL intensity is converted to occupation number by the lifetime correction.

**2.4.4 Bottleneck Effect**

The steepness of the polariton dispersion at low energies, which is responsible for the elevated transition temperatures, also means that the process of cooling via phonon emission is slow. Phonons have a shallow dispersion, so momentum and energy conservation require
emission of phonons with small energies. This means many phonons are required for such relaxation, leading to a ‘bottleneck effect’ [48] in which polaritons accumulate at the point where the dispersion switches from exciton-like to photon-like. This bottleneck effect prevented the investigation of coherence properties for a long time by hampering large accumulation of polaritons at low energies, and producing highly non-thermal distributions of polaritons. However, in wide-band gap systems operating at high temperatures, the bottleneck is not seen at all, due to enhanced relaxation originating from increased polariton-phonon scattering rates.

2.4.5 Bose-Einstein Condensation of Microcavity Polaritons

Bosons can show a spectacular transition to a peculiar condensed phase called a Bose-Einstein condensate. This condensation sees a macroscopic number of bosons accumulate in the ground state of a system. Much has been written regarding the Bose-Einstein condensate since its prediction by Bose and Einstein [49] till its discovery in 1995 in ultracold atomic gases [50]. In these systems temperatures of the order of nK need to be reached. In this sense, polaritons in the bosonic (low density) regime are of great interest as they have a very small effective mass in comparison to the exciton (thanks to their photonic component), which theoretically should allow them to show a temperature of condensation higher than 0.1K [51]. The three essential features of a BEC are:

\begin{itemize}
  \item \textit{a) A bimodal distribution with an accumulation of population in the ground state supported by an evaluation of its occupancy number, which has to be macroscopic (\textit{\textgreater\textless} 1).}
  
  \item \textit{b) Long-range order in correlation and spontaneous symmetry breaking.}
\end{itemize}
c) **Thermodynamic equilibrium**: The distribution of population along states having higher energies than the ground state should be a Boltzmann/Bose-Einstein distribution with a temperature close to the surrounding lattice temperature.

### 2.4.6 Polariton Laser

In conventional atomic BEC, the particle lifetime is much longer than the time it takes to establish equilibrium with itself and with its surroundings. This means that thermal equilibrium has to set in as the condensate is formed. The temperature is well defined and is equal to the surrounding (lattice) temperature. Conversely, there is the non-equilibrium condition where the polariton population decays, emitting photons that leak out of the cavity, before equilibration with itself and the lattice takes place. In that case, temperature of polaritons cannot be defined. The regime in between is called a quasi-equilibrium of polaritons, where, though the lifetime may not be long enough to establish thermal equilibrium with the lattice, the particles live long enough to equilibrate with each other. Here, the polariton temperature can be defined and is expected to be greater than the lattice temperature. Because of the short lifetime of the photonic component of microcavity polaritons, the polariton lifetime may not even be long enough for the polaritons to come to a complete quasi-equilibrium. However, the low-energy range polariton states may still have a definable temperature [52]. This process is often called non-equilibrium condensation or dynamic condensation of polaritons. Formation of the non-equilibrium polariton condensate is possible because of bosonic stimulated scattering [4,7,8,10,53-56]. Stimulated scattering is a basic property of Bose-Einstein statistics which implies that the scattering rate to a \( k \)-state is proportional to \( (1 + N_k) \), where \( N_k \) is the population of that state. The stimulated scattering effect is a sure sign that quantum degeneracy is achieved in the system, since the criterion for this to happen is that \( N_0 > 1 \) where \( N_0 \) is the lowest energy population. Once a
condensate is formed in the ground state, coherent light emission can be observed. This is the principle behind polariton lasers. Population inversion is not required. As a result, the polariton laser has a lower threshold than the photon laser by about two orders of magnitude. The key points needed to infer polariton lasing are:

a) Nonlinear increase of the emission intensity at \( k_\parallel = 0 \)

b) Increase in coherence of the emission indicated by a collapse in linewidth

c) Momentum-space buildup of polaritons around \( k_\parallel = 0 \).

2.4.7 Thermodynamic Picture: Critical density

The microcavity we study is not in a three-dimensional system. Confinement of photons in the microcavity makes it essentially a two-dimensional system. In two dimensions, the critical density of a Bose-Einstein condensate diverges at any temperature greater than 0 K. For a two-dimensional Bose gas the density of states \( g(\varepsilon) \) is a constant:

\[
g(\varepsilon) = \frac{m}{2\pi\hbar^2}
\]  

(2.26)

which gives the total density

\[
n = \int_0^\infty \frac{1}{e^{\beta(\varepsilon - \mu)} - 1} g(\varepsilon) d\varepsilon = \frac{m}{2\pi\hbar^2} \int_0^\infty \frac{1}{e^{\beta(\varepsilon - \mu)} - 1} d\varepsilon
\]  

(2.27)

Evaluating the integral gives

\[
n = -\frac{mk_B T}{2\pi\hbar^2} \ln(1 - e^{\beta\mu})
\]
Therefore, at finite temperatures, as we see from Eq. 1.28, the chemical potential \( \mu \) never goes to zero. In two dimensions, there is no upper bound in the density of excited states and hence there is no true Bose condensate. In fact, spontaneous symmetry breaking is prohibited in 2D. Nevertheless, a transition to a superfluid state can take place as predicted by Berezinskii[57] and independently by Kosterlitz and Thouless [58] or a finite size BEC can also happen.

2.4.8 Thermodynamic vs Kinetic Regimes: Role of Detuning

The exciton-cavity detuning \( \delta \) plays an important role in contributing to the kinetics and the thermodynamics of strongly coupled systems. In the negative detuning regime, or at low temperatures, the polariton-phonon scattering rate is small and it is difficult to attain thermodynamic equilibrium. The system is thus kinetically limited. In contrast, for positive detunings, the increased polariton-phonon scattering rates help to thermalize the lower polaritons (LPs) at \( k_\parallel \sim 0 \) at some effective temperature which can be close to the lattice temperature. However, the critical density for condensation increases with the increase of positive detuning and temperature. Therefore, there are optimum detunings for the observation of polariton lasing and for attaining a state close to thermal equilibrium. These effects are discussed in detail in Chapter 6.

2.5 Conclusion

In this chapter, the basic physics governing a strongly coupled exciton-polariton system and their uniqueness for BEC research has been reviewed. The conditions
required for strong coupling, definitions of several parameters, the difference between a quasi-equilibrium polariton lasing and a BEC in thermal equilibrium and the kinetic and thermodynamic criteria for condensation are discussed.
Chapter 3
Experimental Techniques

3.1 Materials Investigated

An optimal microcavity system for polariton BEC has the following: (1) high quality cavity factor, hence long cavity photon and polariton lifetime; (2) large polariton-phonon and polariton-polariton scattering rates, hence efficient polariton thermalization and no ‘relaxation bottleneck’; (3) a small exciton Bohr radius and large exciton binding energy, hence high exciton saturation density and hence the strong coupling regime can persist up to higher carrier densities; and (4) higher Rabi splitting, hence robust strong coupling. The best defect-free microcavity structures have been achieved with GaAs-based materials with a cavity quality factor $Q$ exceeding $10^5$[59]. Inhomogeneous broadening of exciton energy in this system is limited mainly by monolayer QW thickness fluctuation. Hence, many signatures of polariton condensation were first obtained in GaAs-based systems [60, 61]. However, in recent years, there has been intense interest in developing certain wide-band-gap material systems with small lattice constants. In these materials, the exciton binding energy and oscillator strength are large enough, so that a polariton laser may survive at $\sim 300$ K, operating at visible to ultraviolet wavelengths. More significantly, the Rabi splitting $\Omega$ in these materials is very large. In ZnO, $\Omega$ is almost an order of magnitude larger than in GaAs. The large value of $\Omega$ will allow the system to be operated under positive detuning ($E_C > E_X$, where $E_C$ is the cavity resonance and $E_X$ is the exciton transition energy),
providing a large trap depth $\geq kT$ at 300K while at the same time maintaining the nature of the polariton at the bottom of the lower polariton trap more exciton like. The shorter relaxation time of exciton-like polaritons prevents the relaxation bottleneck encountered in GaAs-based systems.

Two main drawbacks commonly encountered in the wide-band gap systems and the ways they have been overcome are the following:

<table>
<thead>
<tr>
<th></th>
<th>GaAs</th>
<th>GaN (bulk)</th>
<th>GaN (QWs)</th>
<th>ZnO (nanowires)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Exciton binding energy ($R_y^*$)</td>
<td>4.8 meV</td>
<td>27 meV</td>
<td>$&gt; 50$ meV</td>
<td>$\sim 60$ meV</td>
</tr>
<tr>
<td></td>
<td></td>
<td>$&gt; 45$ meV</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Bohr radius</td>
<td>11.8 nm</td>
<td>2.8 nm</td>
<td>1.4 nm</td>
<td></td>
</tr>
<tr>
<td>Valence band splitting</td>
<td>$\Delta_{LH-HH} = 0$ meV</td>
<td>$\Delta_{A_{1s}-B_{1s}} = 8$ meV</td>
<td>$\Delta_{A_{1s}-B_{1s}} = 6$ meV</td>
<td>$\Delta_{A_{1s}-C_{1s}} = 50$ meV</td>
</tr>
<tr>
<td></td>
<td></td>
<td>$\Delta_{A_{1s}-C_{1s}} = 26$ meV</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Refractive index</td>
<td>3.46</td>
<td>2.6</td>
<td>1.9</td>
<td></td>
</tr>
<tr>
<td>Band Gap</td>
<td>1.41 eV</td>
<td>3.4 eV</td>
<td>3.3 eV</td>
<td></td>
</tr>
<tr>
<td>Rabi Splitting</td>
<td>4 meV (single QW) ($&lt; R_y^*$)</td>
<td>30 meV (bulk)</td>
<td>56 meV (QWs)</td>
<td>120 meV ($&gt;&gt; R_y^*$)</td>
</tr>
<tr>
<td></td>
<td>48 meV (nanowires) ($\approx R_y^*$)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Temperature at which excitons dissociate (also a measure of critical temperature to observe BEC)</td>
<td>$\sim 50$ K</td>
<td>$\sim 320$ K</td>
<td>$\sim 560$ K</td>
<td></td>
</tr>
<tr>
<td>Mott density (density at which excitons are screened)</td>
<td>$2.8 \times 10^{16}$ cm$^{-3}$</td>
<td>$3 \times 10^{18}$ cm$^{-3}$</td>
<td>$2.4 \times 10^{18}$ cm$^{-3}$</td>
<td></td>
</tr>
</tbody>
</table>

Table I Comparison of material parameters in light of exciton-polariton emitters.

1. *The structures of wide-band-gap materials suffer a higher concentration of impurities and crystal defects* - We have used epitaxially grown catalyst-free nanowires as active materials. Remarkably, these nanowires are free of extended defects, have no alloy fluctuation to introduce inhomogeneous broadening, have a small surface recombination
velocity $\sim 10^3$ cm/s, which is two orders of magnitude smaller than that of GaAs, and very small surface depletion[62] and thus are easily reproducible [63-67].

2. Fabrication techniques for these materials are not as mature as for GaAs-based systems and integration of the QWs with microcavities is challenging due to the lack of lattice matched DBR layers – We have used dielectric DBRs to overcome this problem. Dielectric DBRs (alternating layers of SiO$_2$/SiN$_x$) deposited using plasma-enhanced chemical vapor deposition (PECVD) can span the entire UV range and can produce microcavities with quality factors similar to that obtained in GaAs-based microcavities. The use of a single GaN/ZnO nanowire in the active region of a dielectric microcavity is easy to fabricate and has a large Rabi splitting originating from large exciton oscillator strength and a modified cavity field that concentrates within the nanowire. In a single ZnO-nanowire dielectric microcavity $\Omega$ is $\sim 100$ meV and the use of QWs to enhance the oscillator strength is absolutely unnecessary.

In the context of this subject matter, the properties of GaN and ZnO are listed in Table 1. For demonstration of polariton lasing and Bose-Einstein condensation at room temperature, we have used single GaN/ZnO nanowires embedded in a dielectric microcavity whereas, for achieving electrically injected polariton lasing we have used GaAs-based microcavities with InGaAs/GaAs QWs since the fabrication techniques for vertical cavity surface emitting laser (VCSEL) are well established.

### 3.2 Fabrication Techniques

**3.2.1 Fabrication Procedure for a Single Nanowire Dielectric Microcavity**

GaN nanowires were grown on (001) Si substrates in the absence of a foreign metal catalyst in plasma-assisted molecular beam epitaxy (PA-MBE) system. First, the surface oxide on the
substrate was removed with a 900 °C anneal in the growth chamber. The substrate temperature was lowered to 800 °C and a few monolayers of Ga were deposited in the absence of N. GaN nanowire growth was initiated at the same temperature at a rate of 300 nm/hr under N-rich conditions. The N flow rate was held constant at 1 sccm. The aerial density of nanowires was varied in the range of $10^8–10^{11}$ cm$^{-2}$ by varying the duration and magnitude of the initial Ga flux to form the droplets.

ZnO nanowires (NW) were grown by pulsed laser deposition utilizing a KrF excimer laser ($\lambda=248$nm, 5-10Hz, 1.7-2 J/cm$^2$) at a substrate temperature of 600°C and a target to substrate distance of 6 cm. An initial ZnO epilayer was deposited onto a (111) n-type silicon substrate ($\rho=0.07$–0.13 Ω-cm) for 20 minutes at a partial oxygen pressure of 10mTorr. This polycrystalline film is estimated to be 100-200nm thick and provides an improved crystalline quality and preferred c-axis orientation for subsequent growth of nanorods [68]. Following the ZnO thin film, nanorods were grown at an oxygen partial pressure of 500mT for 120 minutes. The nanorod formation at high $P_{oxygen}$ is attributed to reduced kinetic energy of the ablated species [69], where nanorod/nanowire diameter will have a strong dependence on $P_{oxygen}$ and substrate temperature. The NW dimensions were determined to be 3-4μm long and 400-700nm wide based on scanning electron microscope measurements. Furthermore, closer inspection of the NW tips revealed hexagonal shaped steps indicating that each layer is epitaxial grown and not a vapor-liquid-solid growth mechanism. The NW growth is attributed to the vapor-solid mechanism [68,70-71] where the ZnO NWs are self-seeding [70]. The preferred columnar growth of the ZnO nanowires grown via the vapor-solid mechanism is attributed to surface instability induced by an Erlich-Schwoebel barrier [71]. The nanowires evolve with sequential layer-by-layer growth with an upper bound for nanowire diameter that is fixed according to
initial nucleation. The principle nanowire orientation was measured to be c-axis (0001) based on x-ray diffraction θ-2θ scans, and later confirmed by transmission electron microscopy. The nanowires exhibit sidewall facets corresponding to the (1100) m-plane. Faceting is also observed on the top of the nanowires, approximately along the (1102) r-plane.

The microcavity was fabricated by depositing the bottom DBR and half of the λ-sized cavity on a SiO₂ substrate by e-beam evaporation (alternating layers of SiO₂ and TiO₂) or plasma-enhanced chemical vapor deposition (alternating layers of SiO₂ and SiNx). Next, nanowires are dispersed by stripping them off the as-grown structure and drop-casting a low density mixture of iso-propyl alcohol and nanowires on the half cavity. Placing the active region at the anti-node of the cavity maximizes the coupling between the nanowire excitons and cavity mode. To isolate a

![Fig. 3.1 (a) Fabrication steps illustrating the fabrication procedure for placing a single nanowire at the antinode of a λ-sized dielectric cavity. (b) Schematic of the single ZnO nanowire-dielectric microcavity. The quality factor of the cavity is estimated to be 526 from the reflectivity spectrum of the entire cavity measured at normal incidence in a region devoid of the nanowires. The Q is mainly compromised by PMMA used for planarization.](image-url)
single nanowire, grid masks with alignment marks are prepared for the sample and a single nanowire is located by scanning electron microscopy with respect to the alignment marks. In case of the ZnO-based structure, since the nanowires are around 200 nm thick, the surface was planarized by spinning and baking PMMA. Finally, the top DBR is deposited and 5 μm mesas centered about the single nanowires are etched down to complete the microcavity. Detaching the nanowires from the Si substrate and dispersing them in the cavity releases any strain that may be present at the interface. The dispersed nanowires exhibited a negligible blueshift (~ 0.9 meV) of the $X_A$ free exciton peak confirming the nanowires are under negligible biaxial compressive stress.

3.2.2 Fabrication of GaAs-based microcavities

Molecular beam epitaxy is used to grow the device heterostructure on a semi-insulating GaAs(001) substrate. The microcavity, in general, consists of a $5\lambda/2$ GaAs cavity surrounded by 30 periods of Si-doped ($n = 2 \times 10^{18} \text{ cm}^{-3}$) GaAs / Al$_{0.85}$Ga$_{0.15}$As forming the bottom distributed Bragg reflector (DBR) and 26 periods of Be-doped ($p = 4 \times 10^{18} \text{ cm}^{-3}$) GaAs / Al$_{0.85}$Ga$_{0.15}$As forming the top DBR. 4 periods of the DBR adjoining the cavity on each side were left undoped to minimize doping related losses. 4 pairs of 10 nm In$_{0.1}$Ga$_{0.9}$As/10 nm GaAs quantum wells are grown at the antinodes of the cavity field distribution to maximize the coupling of the emitter region to the photon field in the cavity. In some cases (e.g. micropillar – see chapter 4) the top 26 pairs of DBR are replaced by only 4 pairs of undoped DBR followed by a p-doped $\lambda/4n$ layer of GaAs. The top DBR is completed by deposition of alternating layers of dielectric ZnSe/MgF$_2$. In the following, we will describe the fabrication procedure of a GaAs-based microcavity with a dielectric DBR at the top, since fabrication of this structure involves maximum complexity.
The process was initiated by the deposition of p-contact metal – Pd 10nm/ Zn 20 nm/ Pd 20 nm/ Au 300 nm and annealed using RTA thermal annealing under an inert N-atmosphere for 1 min at a temperature of 410° C. Next, the microcavity mesa was defined by dry etching using Plasmatherm with Ar/BCl$_3$ etchant gases. Alternately, a wet etching using H$_3$PO$_4$/H$_2$O$_2$/H$_2$O may also be employed if the mesa size is ≥ 20 μm. The wet etch rate is ~4.5 nm/sec although they are slightly different for AlGaAs, GaAs and the InGaAs/GaAs quantum wells. The dry etching process typically etches directionally or anisotropically (see Fig. 2.2(c)); however, it hardens the
organic photoresist which subsequently has to be removed by oxygen plasma. The mesa
definition is done in a GCA auto stepper photolithography tool. This is a I-line photolithography
tool which uses a mercury arc lamp for exposure having a wavelength of 385 nm. This limits the
minimum feature size realizable to ~ 0.5 μm. A high content aluminum layer that is grown
within the microcavity structure is oxidized to confine the current and reduce surface-related
losses. Next, the n-metal contact – Ni 25 nm/ Ge 32.5 nm/ Au 65 nm/ Ti 20 nm/ Au 300 nm is
deposited and annealed at a temperature of 300º C. The structure is now passivated by
conformally coating it with 300 nm of SiN. The deposition proceeds at 200º C in the presence of
silane and ammonia by the plasma enhanced chemical vapor deposition technique. This is
followed by interconnect metal (Ti/Al/Ti/Au) metal deposition. The fabrication is complete with
the selective deposition of 5 pairs of MgF2/ZnSe top mirror in a SJ-26 e-beam evaporator.

3.3 Measurement Techniques

3.3.1. Micro-photoluminescence

Single-QD spectroscopy is performed by micro-PL measurements in the far field in a
confocal geometry. The same microscope objective is used to focus the excitation beam on the
sample and to collect the PL signal. The experimental set-up is schematized in Fig. 2.3. The
sample is mounted on the cold finger of a continuous flow helium cryostat. The excitation beam,
provided by a tunable pulsed Ti:sapphire laser , is focused on the sample with a spot size of
approximately 1μm. The sample is accurately positioned using X–Y piezoelectric stages with a
precision of ±0.05 μm. Moreover, a pinhole located in an image plane allows for spatial
resolution and increases the signal-to-noise ratio. The size of the pinhole is determined by the
magnification of the collection optics (microscope + lens). For the measurement of exciton/bi-
exciton luminescence from a single QD our setup the magnification is about 100 so that a 100µm pinhole selects a 1µm area on the sample surface.

### 3.3.2 Angle-resolved photoluminescence

In this experiment, the excitation is performed by non-resonant excitation with the linearly polarized emission of a frequency tripled ($\lambda = 267$ nm, $f_{\text{rep}} = 80$ MHz and pulse width of 150 fs) Ti:sapphire laser. at normal incidence at high energy. A doublet lens (with a focal length of 10 cm) was used to focus the incident pump beam from a slant angle of $20^\circ$ to a spot size of 100 μm on the sample. The emission is detected at different angles by an optical fiber mounted on the extended rails of a goniometer centered at the sample with an angular resolution of $1^\circ$ and transmitted to a spectrometer. The spectrometer is a SP 2750 monochromator with a focal length of 750 mm and gratings upto 1200 g/mm, allowing a resolution of .023 nm at 435.8 nm. The detector is a Princeton Instruments photo-multiplier tube with phase-sensitive lock-in amplification and is capable of detecting weak signal from a single nanowire.
3.3.3 Time-resolved photoluminescence

Since the time-scale of polariton dynamics is of the order of few ps, it is impossible to use a single photon detector, which has a resolution of ~ 50 ps, for measuring the transient behavior of exciton-polaritons. We use a Hamamatsu Streak Camera with a time resolution < 2 ps. The streak camera is an ultra-high speed detector which captures light emission phenomena occurring in extremely short time periods. In addition to superb temporal resolution, the streak camera can capture spatial (or spectral) data simultaneously. A 50 cm monochromator is attached to the front...
of the streak camera which may degrade the time resolution. When using the lowest grating of the spectrometer with 100-grooves per millimeter and a spectral resolution of \( \gg 1 \) nm, the time resolution increases to \( \gg 3 \) ps. The streak camera is triggered by the same mode-locked Ti-Sapphire laser used for pumping. Instability of the mode-locked laser operating in the picosecond mode causes time jitters of the streak signal, hence in practice, with an average integration time of about one minute per spectrum, the time resolution is 4 to 5 ps.

### 3.3.4 Output polarization measurements

The polarization of the output in the strong coupling regime was investigated by placing a Glan-Thompson linear polarizer at the entrance of the monochromator. In order to remove any correlation with the pump signal, during the linear polarization measurements, the pump was circularly polarized by passing it through a quarter wave plate. The measured intensities as a function of the angle between the polarizer and the reference x-axis, for polariton emission from a single ZnO nanolaser normal to the sample (from \( k_{\parallel} \sim 0 \)) are displayed in the polar plot in Fig. 3.6 for pump powers below, at an above threshold. The degree of linear polarization is defined to

![Image of linear polarization measurement setup and polar plot](image)

**Fig. 3.6** (Left) Schematic of the linear polarization measurement setup. (Right) Dependence of the PL intensity on the angle of polarization for a ZnO polariton nanolaser.
be \( \frac{I_{\text{max}} - I_{\text{min}}}{I_{\text{max}} + I_{\text{min}}} \), where \( I_{\text{max}} \) and \( I_{\text{min}} \) are the maximum and minimum intensities.

### 3.3.5 Measurement of circular polarization in a magneto-optic cryostat

The degree of circular polarization for the spin polarized single photon source was measured using a photoelastic modulator (PEM) and a Glan Thompson linear polarizer. The schematic of the setup is shown in Fig. 3.7. Stoke polarimetry with a PEM has the advantage that no mechanical changes of the optics is required for the measurement. In fact two PEMs can be combined to measure all four Stoke parameters simultaneously. The principle of operation for a PEM is based upon the photoelastic effect, in which a mechanically stressed quartz (or fused silica) exhibits birefringence proportional to the resulting strain. The light intensity reaching the silicon photodetector is given by:

\[
I = \frac{1}{2} S_0 + S_2 J_2(2.405) \sin(2\omega t) + \frac{1}{2} S_3 J_1(2.405) \sin(\omega t)
\]

(2.1)

where \( \omega \) is the resonant angular frequency of the PEM and \( J_n \) is the nth-order Bessel function of the first kind. The circular polarization component will produce a signal at the modular frequency 1f, and this signal is proportional to Stokes parameter \( S_3 \). The linear polarization component will give a signal at twice the modulator frequency 2f. A signal conditioner is required to separate the DC and AC components and sufficiently amplify these signals. Using a lock-in amplifier locked to the 1f modulation frequency of the PEM, we are able to measure the ratio of AC to DC signals. This ratio is proportional to the degree of circular polarization. Very careful calibration experiments should be conducted to eliminate electrical offsets in the measurement equipment and to determine the calibration coefficient. The calibration coefficient is required to account for the differential gain for the DC and AC components present in the
amplifier electronics and for the fact that the lock-in measures an RMS value of the AC component.

3.3.6 First-order temporal coherence

First order temporal coherence of the emission was measured using a Michelson interferometer. The light emission is divided by a beam splitter oriented at 45° to the beam and reflected back by two mirrors that interfere to produce fingers in the screen. One of the mirrors is mounted on a piezo-stage so as to change the path difference and thus a dark fringe changes to a bright one. The interference fringe contrast is given by $C = (I_{\text{max}} - I_{\text{min}}) / (I_{\text{max}} + I_{\text{min}})$, where $I_{\text{max}}$ and $I_{\text{min}}$ denote the maximum and the minimum of the interference pattern). The coherence length is the length for which the measured fringe contrast decays to 1/e times the maximum value. Coherence time (length) is an important parameter for polariton lasers. This time should be significantly higher than the relaxation time $\tau_{\text{rel}}$ and the polariton lifetime $\tau_{\text{pol}}$ (see chapter 6).

Fig. 3.7 Experimental setup for measurement of the degree of circular polarization using a photoelastic modulator.
3.3.7 First-order spatial coherence

We measured the first order spatial coherence using a Michelson interferometer setup as shown in Fig. 2.8. The beam splitter creates identical images of the condensate in the two arms of the interferometer and they are overlapped spatially by the translational movement of a mirror (mirror I) mounted diagonally on a piezo-stage. The arrangement creates a double image of the condensate with a spatial resolution of 0.3 μm. A pinhole placed at the overlap image plane serves as a spatial filter for selecting the dark and bright fringes, the intensities of which are measured by a PMT. The imaging of the interference pattern using a CCD camera was limited by the gain of the camera since the intensity of light emitted from a single nanowire is extremely low. In contrary, the PMT, with its huge gain, allows the measurement of the intensity of a fringe, after it has been spatially selected by the pinhole. Spatial coherence is often times described in terms of visibility or the degree of contrast of the interference pattern. Visibility is defined as: \( V = \frac{(I_{\text{max}} - I_{\text{min}})}{(I_{\text{max}} + I_{\text{min}})} \), where \( I_{\text{max}} \) and \( I_{\text{min}} \) are intensities of the bright and dark fringes and is related to the first order correlation function as

\[
V = \frac{2\sqrt{I_1 I_2}}{I_1 + I_2} g_{12}^1
\]  (2.2)
Thus, we see that if the intensities of the two fields from the mirrors are equal, the visibility of the interference pattern determines the first-order coherence. For infinitely coherent fields, the visibility $V = 1$, and $V = 0$ for a totally incoherent source.

![Misaligned Michelson interferometer setup](image)

Fig. 3.9 Misaligned Michelson interferometer setup for measuring spatial coherence.

### 3.3.8 Second-order temporal coherence

The second order correlation function $g^2(\tau) = \langle I(t)I(t+\tau)\rangle/\langle I(t)\rangle\langle I(t+\tau)\rangle$, where $I(t)$ is the luminescence intensity at time $t$ was measured using a Hanbury-Brown and Twiss like setup which consists of a 50:50 non-polarizing beam splitter and two identical single photon counters. The photon counters are avalanche photodiodes that have a time resolution as short as 50 ps and a dark count rate of 30 Hz. The device under test is housed and cooled in a cryostat and the emitted light is first transmitted through a monochromator to select the exciton or the bi-exciton or the polariton emission wavelength and is then equally split by the beamsplitter. Because of the faster dynamics of polaritons, a correction as suggested by Kasprzak et al [72] needs to be undertaken. For cw measurements, the actual zero delay peak $g^2(0)$ can be estimated from the
measured zero delay peak $g_m^2(0)$ by: $g_m^2(0) \approx 1 + (g^2(0)-1)\tau_C/\tau_{AC}$, where $\tau_C$ is the polariton coherence time and $\tau_{AC}$ is the resolution of the setup. The use of pulsed excitation has some benefits when $g^2(\tau)$ is to be measured. First, the time resolution of the detectors is no longer an issue, unless it is longer than the excitation pulse period (12.5 ns in our case). Second, photons are emitted within a finite time interval after each excitation pulse. As a result, for a given counting rate, the number of coincidences is significantly higher than for cw measurements, making data acquisition times shorter and thus permitting to obtain better signal-to-noise ratio. However, a correction involving the polariton relaxation dynamics has to be undertaken $g_m^2(0) \approx 1 + (g^2(0)-1)\tau_C/\tau_{rel}$, where $\tau_{rel}$ is the polariton relaxation rate and is a function of the incident power.

### 3.3.9 Momentum space mapping - Calculation of occupancy

The polariton occupancy was estimated from the integrated luminescence intensity ($I_{LP}(k_{||})$) measured for different in-plane wave-vector $k_{||}$. In the angle-resolved measurements, the emission was collected by a fiber-coupled lens with an angular resolution of $\sim 1^\circ$ which is in turn connected to a spectrometer. The collection optics are on extended rails of a goniometer centered at the sample. The occupation number is given by the formula, $I_{LP}(k_{||}) = \eta N_{LP}(k_{||})|C(k_{||})|^2M/
\[ \tau_C, \text{ where } \eta \text{ is the collection efficiency, } \tau_C/|C(k||)|^2 \text{ is the radiative lifetime of the LPs and } M \text{ is the number of transverse states included in the detection cone. } |C(k||)|^2 \text{ is the photon fraction at a wave-vector } k||. \text{ The number of states within the detection cone is given by } M = \frac{D^2}{16} (k_0 \Delta \theta)^2, \text{ where } D \text{ is the diameter of the excitation spot, } k_0 = 2\pi/\lambda \text{ and } \Delta \theta \text{ is the detection half angle. The value of } \eta \text{ is estimated by replacing the sample with a source of known power (the frequency tripled Ti:sapphire laser with suitable attenuation).} 

3.4 Conclusion

In this chapter the different materials used as emitters for the strongly coupled microcavities in the dissertation, the fabrication techniques of these microcavities and the measurement set-ups have been discussed.
Chapter 4
Optically Excited Single Nanowire Polariton Lasers

In large bandgap semiconductors such as GaN or ZnO, it is possible to observe polariton lasing at room temperature because of the large exciton binding energy and small exciton radius, providing a very large critical temperature for polariton condensation, a necessity for high-temperature polariton lasing. Furthermore, there is evidence of the absence of a polariton relaxation bottleneck in bulk GaN microcavities at high temperatures. Ga(In)N and ZnO nanowires can be grown defect-free and very low background carrier concentration on silicon and other substrates. There is also little or no residual strain in these nanowires and hence problems associated with strain-induced piezoelectric fields and decreasing electron-hole overlap are absent. Also absent is any inhomogeneous broadening due to interface roughness in multi-quantum well heterostructures. Therefore a single pristine GaN/ZnO nanowire of appropriate diameter embedded in a high-Q microcavity is an excellent candidate for realizing a high temperature polariton laser.

4.1 Room Temperature Ultra-Low Threshold GaN Nanowire-Polariton Laser

In this work, we developed a new structure — GaN nanowires — as the active media, and produced a room-temperature polariton laser operating at an energy threshold orders of magnitude lower than those reported earlier. Remarkably, the nanowires are free of extended defects, have no alloy fluctuation to introduce inhomogeneous broadening, have a surface
recombination velocity $\sim 10^3$ cm/s, which is two orders of magnitude smaller than that of GaAs, and very small surface depletion and thus are easily [63-67]. It has been seen experimentally that the polarization field in the nanowires is negligible [73] and the internal quantum efficiency is $\sim$ 50-60%. We enclosed a single nanowire in a dielectric microcavity. The characteristic polariton dispersions were measured by angle-resolved photoluminescence. We also studied the linewidth narrowing and energy shift of the lasing mode, population redistribution in the momentum space, and coherence functions of the polariton laser. A second threshold, which we believe corresponds to photon lasing with an inverted carrier population, was observed at 2700 times the polariton lasing threshold.

Fig. 4.1 (a) Schematic representation of the dielectric microcavity with a single GaN nanowire of diameter 60 nm (inset) buried in the center of a $\lambda$-sized cavity; (b) photoluminescence spectrum from a single GaN nanowire measured at 25K showing free and bound exciton transitions.

### 4.1.1 Device Structure

The GaN nanowire-microcavity used in this study is shown schematically in Fig. 4.1(a). It consists of a SiO$_2$ $\lambda$-cavity sandwiched by a top and a bottom distributed Bragg reflector (DBR)
made of 7 pairs of SiO$_2$/TiO$_2$. A single nanowire is placed at the central antinode of the cavity field to maximize the interaction between the GaN nanowire excitons and the cavity mode. The inset to Fig. 4.1(a) shows the scanning electron microscope image of the nanowire lying on the half cavity, before the top cavity was deposited. The wire has a diameter of $\sim$60 nm and a length of $\sim$750 nm. The nanowire diameter corresponds to the total thickness of 50 quantum wells used in similar devices [23]. A low background carrier concentration of $8.5 \times 10^{16}$ cm$^{-3}$ was derived from measurements made on Pt-GaN nanowire Schottky diodes [64]. A photoluminescence (PL) spectrum of the nanowires at 25 K is shown in Fig 4.1(b). The three free exciton transitions, $X_{A,B,C}$, corresponding to the wurtzite crystalline structure are indicated. The peaks labeled DBX$_{A}$ and DBX$_{B}$ are bound exciton transitions. The broad and weak transition at $\sim$ 3.41 eV is related to the surface [64]. The lineshape of the exciton transitions resembles a Lorentzian function instead of a Gaussian function, suggesting negligible inhomogeneous broadening of the exciton resonances in the nanowires. After the complete cavity is fabricated, enclosing the nanowire, the cavity is etched into a mesa of 5 $\mu$m diameter. We have studied two devices, fabricated from different samples of GaN nanowires with different cavity-exciton detunings. The measured results are qualitatively similar and in the following we describe the sample which is negatively detuned at 300K.

4.1.2 Dispersion Characteristics

We measured the dispersion of the resonant modes at 300K by angle-resolved photoluminescence and the spectra are shown in Fig. 3.2(a). A peak below the exciton resonances is clearly observed in the spectra, which asymptotically approaches the $X_{A}$ exciton energy at larger angles. A weaker peak above the exciton resonances can also be identified, and the two peaks anti-cross with increasing angle. Using the one-to-one correspondence between the
angle of the out-coupled photon and the in-plane wavenumber of the polaritons, we obtained the energy-momentum dispersion of the resonances (Fig. 4.2(b)). The results are well described by the polariton dispersions calculated by the coupled oscillator model (as shown by the solid curves). Here we consider only coupling of X$_A$ exciton and the cavity mode, because the X$_B$ and X$_C$ excitons have much weaker oscillator strengths according to PL measurements. The cavity to exciton detuning $\delta$ and photon-exciton interaction potential $V_A$ are obtained by fitting the data and are found to be $\delta = -20$ meV and $V_A = 48$ meV for the X$_A$ exciton. The exciton linewidth without thermal broadening used in the Hamiltonian is $\Gamma_{X_A} = 4.5$ meV and is obtained from independent photoluminescence measurement on a single GaN nanowire at 25 K. The relatively large value of $V_A$ for a single nanowire is due to both the large oscillator strength of excitons resulting from a small internal electric field of ~0.19 MV/cm [67,73], and a modified cavity field that concentrates within the nanowire. Since the refractive
Fig. 4.3 The calculated electric field intensity distribution of the fundamental resonance mode around the nanowire of length 400 nm and diameter 60 nm, embedded in a 5 µm diameter pillar cavity. The GaN nanowire is embedded horizontally at the center of the cavity spacer. The figures show the cross-sectional profiles of the electrical field intensity along the x, y and z directions. The y direction is defined to be along the length of the nanowire. The z direction is in the cavity growth direction. The origin is at the center of the nanowire. (courtesy: Lei Zhang)

index of the nanowire is higher than that of the cavity, the cavity field is mainly within the nanowire region even without additional transverse confinement of the cavity mode. This is

Fig. 4.4 (a) Photoluminescence spectra of GaN nanowire-microcavity measured as a function of angle from 0 to 21° at 200K. The peak energy positions corresponding to the ‘data’ points in Figure (b) have been marked in the spectra. The energy position of the excitons is also shown. (b) Experimental polariton emission energy resonances obtained from the spectra of Figure (a) as a function of angle and in-plane wave vector together with the polariton dispersion curves obtained from solving the coupled harmonic oscillator Hamiltonian. The error bars denote uncertainty in energy of the peak resonances.
confirmed by finite difference time domain (FDTD) simulations (see Fig 4.3). The polarization field in a bulk GaN microcavity is calculated to be ~ 1 MV/cm for a reported compressive stress of 30 kBar [74]. The oscillator strength in the single nanowire is therefore approximately twice than in the bulk microcavity [75] and the interaction potential $V_A$ is expected to be ~ 44 meV compared to the bulk value of 31 meV [76,77]. Consistent polariton dispersions similar to the data of Fig. 2 were also obtained at 200K and is shown in Fig. 4.4.

### 4.1.3 Polariton Lasing Characteristics

To investigate the non-linear optical properties of the nanowire microcavities, we pump the device non-resonantly with a frequency-tripled Ti-Sapphire laser focused to a spot size of ~100μm in diameter. Photoluminescence from the normal direction was analyzed with a high resolution monochromator (with a spectral resolution of ~ 0.03 nm), and detected with a photomultiplier tube using phase-sensitive lock-in amplification. With increasing pumping power, a sharp super-linear increase of the PL intensity from lower polaritons (LPs) at $k_\parallel \sim 0$ was observed at both 200 and 300K (Fig. 4.5), accompanied by a sharp decrease in the emission linewidth and a small blueshift of the LP energy. Both features indicate the onset of stimulated scattering into the LP states near $k_\parallel = 0$. The incident excitation energy at threshold was $E_{th1} = 92.5$ nJ/cm$^2$ at 300 K and 63 nJ/cm$^2$ at 200 K. We estimate an upper bound of the LP density at the threshold to be $N_{LP}(E_{th1}) = 2 \times 10^{16}$ cm$^{-3}$, using the relation $N_{3D} < E_{th1}/(E_{pump}D)$. The LP density at the threshold, $N_{LP}(E_{th1})$, is two orders of magnitude lower than any previously reported GaN polariton lasers, and three orders of magnitude less than the exciton Mott density of $3 \times 10^{19}$ cm$^{-3}$, [78] the latter corresponding to the required carrier density for conventional photon lasing. Remarkably, the gain media in our device is a single nanowire with the lateral size of
∼0.045 μm², which is orders of magnitude smaller than the optical excitation spot size used in these measurements. Therefore the nanowire-microcavity device can allow further reduction in the threshold energy with electrical injection.
Above threshold, the emission spectra showed multiple peaks with greatly reduced linewidths, corresponding to different transverse modes of the LPs due to localization of the LP modes by the nanowires. Similar multi-mode lasing has been observed in bulk [11] and multiple-QW [23] GaN microcavities, where the localization was attributed to photonic disorders in the structures, and the modes varied with spatial alignment of the pump with the sample. In contrast, in our device, the modes are intrinsic to the nanowire-microcavity. They are not due to crystal defects and do not depend on the spatial alignment of the pump. Below threshold, the modes are indistinguishable due to a thermally broadened linewidth of >10 meV. At threshold, the linewidths are greatly reduced to a mere 0.67 meV at 200K and 1.1 meV at 300K, and thus revealing the discrete modes in the spectra. The reduction in linewidth indicates the increase in the coherence time of the LPs in the lasing mode to 6.2 ps at 200K and 3.8 ps at 300K.

A small blueshift $\delta E$ of the LP emission was observed with increasing pump power up to $\delta E \sim 1.78$ meV (200K) and 1.92 meV (300K) around the threshold. Further increasing excitation density above threshold led to only negligible change in $\delta E$ until exciton saturation sets in at much higher pump powers. The behavior is typical in polariton lasers. The blueshift is caused by repulsive self-interaction of the exciton-polaritons and the resultant renormalization of the exciton self-energy [79]. From the blueshift, the exciton-polariton population $N_{3D}$ may be estimated using $\delta E \approx 3.3\pi E_X^B a_B^3 N_{3D}$, [11] where $E_X^B = 30$ meV is the $X_A$ exciton binding energy and $a_B = 3.5$ nm is the exciton Bohr radius. We found $N_{3D} = 1.43 \times 10^{17}$ cm$^{-3}$ at threshold. This value is a factor of 7 larger than that estimated from the incident excitation energy, which could be due to a couple of reasons. The first is the spread of values of $a_B$ for GaN quoted in the literature. Second, the exciton population introduces both phase space filling and a saturation of the oscillator strength, both resulting in a blueshift of the lower polariton energy. The formula
quoted above is based on phase space filling only. Hence the exciton population calculated based on the total measured blueshift is higher than the actual exciton population.

4.1.4 Transition to Weak Coupling and Photon Lasing

With further increase of excitation density (by using an objective lens and spot size of ~ 10 µm) to \((10^2-10^3)E_{th1}\), the emission becomes strongly blueshifted and broadened (see Fig. 4.7), indicating exciton saturation and transition to the weak coupling regime [80]. A second distinct threshold was observed at \(E_{th2} \approx 2700E_{th1} = 250 \mu J/cm^2\), which correspond to a population slightly higher than the Mott density and represents the onset of photon lasing. The integrated emission intensity over the entire range of excitation power density is shown in Fig. 4.5(c). The two distinct thresholds, three orders of magnitude apart in excitation density, are strong evidence of the very different origin of the nonlinear emissions. Polariton lasing results from stimulated scattering of polaritons into the quantum degenerate polariton states at \(k_{||} \sim 0\), without requiring population inversion. Photon lasing results from stimulated emission into the cavity modes upon population inversion in the electronic media.

Fig. 4.6 Variation of emission linewidth (ΔE) and blueshift (δE) with incident pump power density measured at 300K from the strong coupling (polariton lasing) to the weak coupling (photon lasing) regime.
4.1.5 Occupancy

To confirm the physical mechanism for the first nonlinear threshold, we measured the LP population distribution in k-space by angle-resolved photoluminescence. Figure 4.7 shows the spectra of angle-resolved PL between $0^\circ$ and $27^\circ$ at 300 K, both below (a) and above (b) threshold. There is no obvious energy-relaxation bottleneck at all excitation densities. Below threshold, emission intensities are comparable at different angles. Above threshold, the emission becomes sharply peaked at small angles, showing condensation in k-space. The number of polaritons per $k_{\parallel}$ state is estimated from the PL intensity ($I_{LP}(k_{\parallel})$) by using the formula,

$$I_{LP}(k_{\parallel}) = \eta \frac{N_{LP}(k_{\parallel}) |C(k_{\parallel})|^2 M}{\tau_{C}}$$

where $\eta$ is the collection efficiency, $\tau_{C}$/ $|C(k_{\parallel})|^2$ is the
radiative lifetime of the LPs and M is the number of transverse states subtended by the detection cone and is shown in Fig. 4.7(c). As can be seen again that below threshold the distribution is random whereas above threshold there is a sharp increase in the occupancy near \( k_\parallel \sim 0 \) states. The occupancy above threshold fits with a Maxwell-Boltzmann distribution with a temperature of the lower polariton gas \( T_{LP} = 392 \) K.

### 4.1.6 Coherence properties

![Graphs of Fringe Contrast and g(2)(0)](image)

**Fig. 4.8** Interference fringe contrast \( C = (I_{\text{max}} - I_{\text{min}}) / (I_{\text{max}} + I_{\text{min}}) \) (where \( I_{\text{max}} \) and \( I_{\text{min}} \) denote the maximum and the minimum of the interference pattern) measured in a Michelson interferometer as a function of pump power density. (b) The measured second order coherence function \( g_m^{(2)}(0) \) versus incident pump energy. The insets show the number of coincidences as a function of delay \( \tau \) for the polariton luminescence at \( k_\parallel = 0 \), at excitation energies of 100 nJ/cm\(^2\) (around threshold) and 162.5 nJ/cm\(^2\).

Since observation of a threshold is not sufficient to prove lasing from a coherent state, we investigated the coherence properties of the emission by measuring the first and second order correlations. The narrowing of the linewidth did give us some insight into the coherence of the emission. The change in linewidth from 14 meV to 1.14 meV corresponds to an increase in coherence time \( \tau_c = \lambda^2/c\Delta\lambda \) (where \( \lambda \) is the emission peak wavelength, \( c \) is the phase velocity of light in the medium and \( \Delta\lambda \) is the emission linewidth) from 0.318 ps to 3.62 ps. Further evidence
was provided by measuring the first order coherence of the emission in a Michelson interferometer below and above threshold. Fig 4.8(a) plots the interference fringe contrast \( C = (I_{\text{max}} - I_{\text{min}}) / (I_{\text{max}} + I_{\text{min}}) \) (where \( I_{\text{max}} \) and \( I_{\text{min}} \) denote the maximum and the minimum of the interference pattern) measured as a function of pump power density. Below threshold, the contrast has a noise limited value ~2-5% and it increases to a value of ~ 32% above threshold indicating a significant increase of the coherence length.

The second order coherence function in time domain was measured as a function of incident excitation energy by a Hanbury-Brown and Twiss type measurement system. Similar results as reported by Kasprzak et al \[72\] were obtained, and are shown in Fig. 4.8(b). Below threshold, we measure a \( g_m^{(2)}(0) \sim 1.00 - 1.0036 \) at zero time delay, while a \( g^{(2)}(0) \sim 2 \) is expected. The discrepancy stems from the fact that photon bunching occurs only during the coherence time \( \tau_c \) of polaritons, which is much shorter than both their relaxation time \( \tau_{\text{rel}} \) and the time resolution of the photodetector. Just above threshold, \( \tau_c \) increases to 4 ps while \( \tau_{\text{rel}} \) decreases sharply due to stimulated scattering. Therefore \( g_m^{(2)}(0) \) may give a good estimate of the actual \( g^{(2)}(0) \), and \( g_m^{(2)}(0) \sim 1 \) may suggest the suppression of photon bunching. Higher above threshold, \( g_m^{(2)}(0) \) increases with increasing pump power, consistent with the observed linewidth broadening, suggesting decoherence induced by increased polariton-polariton interactions.

**4.1.7 Conclusion**

In conclusion, room temperature polariton lasing at record low threshold polariton densities was achieved in a vertical microcavity containing a single GaN nanowire. The threshold density was two orders of magnitude lower than reported values for GaN-based polariton lasers. Dynamic and coherence properties of the device all suggest that the low-threshold lasing results
from stimulated scattering and accumulation of a quantum degenerate polariton population near the band minimum. In comparison, a second threshold corresponding to conventional photon lasing was also observed at a density 2700 times the polariton lasing threshold density. Reducing the lateral size of the microcavity would increase the photon-exciton coupling and further improve the optical qualities of the polariton laser. Our work represents a first step in developing a new generation of ultra-low power and ultra-compact, room-temperature photonic devices based on GaN nanowire polaritons.

4.2 Room Temperature Strong Coupling Effects from Single ZnO Nanowire-Microcavity

To observe strong coupling effects [11,23,39,61,62,80,81] at temperatures close to or equal to room temperature, materials which provide large coupling strength, and hence large exciton oscillator strength and binding energy, are desired. Therefore, attention has shifted from GaAs-based microcavities to GaN-based [11,23,37], and more recently to ZnO-based ones [24,82-86]. ZnO is a wide bandgap semiconductor with an exciton binding energy $E_B \sim 60$ meV and a Bohr radius $a_B \sim 1.4$ nm. The critical temperature for Bose condensation, $T_C$ is $\sim 560$K [87]. In comparison, $E_B$ is $\sim 7$-9 meV and $T_C$ is $\sim 100$K for GaAs quantum wells. The characteristics of exciton-polaritons and their strong coupling in bulk [24,82,83], microwire and nanowire [84,85]. ZnO-based microcavities at room temperature and polariton lasing in a bulk ZnO microavity at 120K have been reported [86]. However, room temperature polariton lasing with ZnO, in any form, has not yet been realized.

In the present study we have investigated strong coupling effects in a single nanowire-dielectric microcavity at room temperature. The single ZnO nanowire is embedded in a dielectric and surrounded by distributed Bragg reflectors (DBRs) in the top and bottom. Thus the excitons
strongly couple to normal cavity modes in this configuration and not to whispering gallery modes [85] or to cavity modes along the length [84], as in the case of a free-standing nanowire, which itself also serves as the resonant cavity. The embedded nanowire modifies the cavity field and reduces the mode volume, thereby increasing the coupling constant. The polariton dispersion characteristics at room temperature have been calculated and measured by angle resolved photoluminescence from which a Rabi splitting of 103 meV is obtained. Non-linear emission characteristics are observed at room temperature with a distinct threshold at a very low optical excitation density of 1.63 μJ/cm², accompanied by linewidth narrowing. The measured population distribution in momentum space and the polariton relaxation and recombination times confirm the absence of a relaxation bottleneck and the attainment of quantum degeneracy at \( k_\parallel \sim 0 \).

4.2.1 Device Structure
ZnO nanowires (NW) were grown on (111) silicon substrate by the pulsed laser deposition technique [68]. The nanowires are typically 1-2μm long and 150-700nm in diameter based on scanning electron microscope (SEM) imaging. An SEM image of the nanowires is shown in Fig. 4.9(a). The nanowire density is estimated to be ~1×10⁹ cm⁻². Figure 4.9(b) shows a high resolution transmission electron microscope (TEM) image of a single nanowire, which has good crystalline structure and no dislocations or stacking faults are observed. It is worth mentioning

Fig. 4.10 (a) Schematic of the nanowire-microcavity device with the SEM image of a single nanowire placed on the partial cavity; (b) The calculated electric field intensity distribution of the fundamental x-polarized resonance mode around the nanowire of length 1 μm and diameter 200 nm, embedded in a dielectric cavity. The figures show the cross-sectional profiles of the electrical field intensity along the x, y and z directions.(courtesy: Junseok Heo)
that the contrast present in this image is due to the relatively large diameter (D) of the nanowire, ~200-300 nm. A schematic representation of the mesa-shaped single nanowire-dielectric microcavity is shown in Fig. 4.10(a). The microcavity was fabricated by depositing the bottom DBR (15 pairs of alternating SiO\textsubscript{2} and Si\textsubscript{3}N\textsubscript{4}) and 44 nm of the 3\lambda/2 SiO\textsubscript{2} cavity on a Si substrate by plasma enhanced chemical vapor deposition (PECVD). Next, nanowires are dispersed by drop-casting a low density mixture of iso-propyl alcohol and nanowires on the surface. The ZnO nanowire is 200 nm in diameter and hence it extends over 2 antinodes of the 3\lambda/2 cavity. To isolate a single nanowire, grid masks with alignment marks are prepared for the sample and a single nanowire is located by scanning electron microscopy with respect to the alignment marks. Because of the relatively large NW diameter, the surface was planarized by spinning and baking PMMA. Finally, the rest of the SiO\textsubscript{2} cavity and the top DBR are deposited and 10 \mu m mesas, centered around the single nanowires, are etched down to complete the microcavity. The mesas are at least 1 mm apart and hence a 100 \mu m excitation spot only excites a single mesa. Finite difference time domain (FDTD) simulations were performed with the polarization of the excitation source set perpendicular to the c-axis of the nanowire because X\textsubscript{A} and X\textsubscript{B} transitions
are more dominant over $X_C$ as discussed later. For simplicity, the ordinary refractive index of ZnO was only taken into account, instead of considering the anisotropy of ZnO. The calculated profile of the dominant cavity mode (shown in Fig. 4.10(b)) confirms that $E_x$ and $H_y$ field components are the dominant ones. The field along the z-direction is similar to that of a planar microcavity. In addition to the ZnO-dielectric index step, the top and bottom DBRs of the microcavity provide better confinement along the z-direction, so that the light is more strongly confined within the nanowire. The polarization field in the nanowire is small and assumed to be of the same order, ~0.1 MV/cm, as in comparable GaN nanowires. Both of these factors contribute to a large oscillator strength in the ZnO nanowire [45]. The estimated quality factor is ~300, which corresponds to a cavity Q factor of a 10 μm long isolated nanowire without any DBR. Thus the dielectric microcavity provides a relatively high Q for a very short length of the nanowire. It is worthwhile to mention that polariton lasing at 120K in a planar bulk ZnO based microcavity with similar Q-factor has been reported recently [86].

### 4.2.2 Excitonic Spectra and Dispersion Characteristics

The optical properties of the nanowires were first studied by photoluminescence (PL) measurements on a nanowire sample performed with excitation from a frequency-tripled mode locked Ti:Sapphire laser (pulse width 130 fs; repetition rate 80 MHz) at 267 nm. The wurtzite crystalline structure gives rise to three free exciton transitions, $X_A$, $X_B$ and $X_C$, of which the $X_A$ and $X_B$ excitons are polarized perpendicular to the c-axis whereas the $X_C$ exciton is strongly polarized parallel to the c-axis, as confirmed by interband momentum-matrix calculations [88,89]. The PL spectrum at 10K, shown in Fig. 4.11(a), is characterized by free ($X_A$, $X_B$) and donor-bound exciton transitions and their LO-phonon replicas. The temperature dependence of the free exciton ($FX_A$) peak can be fitted well with the Manoogian and Wooley equation and is
shown in Fig. 4.11(b). At 300K the FXA peak is observed at ~3.312 eV. The transmission characteristics of the nanowires were also measured at room temperature and are shown in Fig. 4.11(c). The energy position of the absorption edge measured from the transmission characteristics coincides with that of the PL peak, indicating negligible Stokes shift and a low density of defects in the nanowires.
The anti-crossing behavior of the resonant polariton modes were determined by both temperature dependent and angle resolved PL measurements. Excitation for these measurements is again provided by the frequency tripled Ti:sapphire laser focused to a spot size of 100 μm on the sample. Temperature-dependent PL spectra measured in the normal direction are shown in Fig. 4.12(a). The spectra are characterized by a strong lower polariton (LP) peak below the FX_A transition energy while the resonant energies in the UPB are not observed at all, similar to previous observations with ZnO-based microcavities [24,83,90,91]. The explanation offered by these authors is the large vacuum Rabi splitting that pushes UPB into the absorption states of the ZnO active region. The emission feature is also not from a Bragg mode which is observed at 3.139 eV in the 300K reflectivity spectrum (not shown here) measured in the normal direction. The LP peak energy is plotted in Fig. 4.12(b), together with the measured temperature-dependent FX_A energies, E_X, as described above and cavity resonance E_C. Data obtained from angle resolved PL measurements are shown in the color contour plot of Fig. 4.12(c). The orientation of the angle is orthogonal to the wire since the excitons are coupled to a 2D-cavity mode in a direction perpendicular to the wire. A clear signature of the LPB is again seen at all angles of the out coupled photons and the LPB energy asymptotically approaches the X_A energy at high angles. This proves that the emission is not from a cavity mode. The experimental results have been analyzed with the exciton-photon coupled oscillator model, considering only the coupling of X_A exciton and cavity mode, since the X_B exciton exhibits a weak oscillator strength in PL spectra. The exciton linewidth used in the Hamiltonian is obtained from independent temperature dependent PL measurements from ZnO nanowires. The polariton dispersions calculated by the coupled oscillator model are shown by solid lines in Fig. 4.12(b) and by dotted lines in Fig. 4.12(c). The analysis yields a cavity-to-exciton detuning δ = +1±2 meV and Ω =
100±3 meV. The large value of Ω is attributed to the excellent crystalline quality of the ZnO nanowires and a concentration of the cavity field within the single nanowire in the microcavity. A Rabi splitting ~ 100 meV creates a LP trap depth of ~ 50 meV even with a positive detuning. The squared modulus of the exciton and photon Hopfield coefficients derived from the analysis reveals that the LPB is mostly exciton-like with an exciton fraction of ~ 0.51 at k∥ = 0. These characteristics are very desirable for achieving condensation and quantum degeneracy of polaritons at room temperature in a microcavity with moderate Q, since with increase of exciton fraction the polariton relaxation times are reduced and the radiative lifetimes are enhanced [92]. However, it is realized that a positive detuning is not optimal for achieving a low threshold density of excitation for the observed non-linearity described below.

4.2.3 Lasing Characteristics

To investigate non-linearity and coherence properties of polariton emission from k∥ ~ 0 states, the microcavity was excited at an angle and the luminescence at zero detection angle was analyzed as a function of pump power. The integrated emission intensity is plotted in Fig. 4.13(a) as a function of the incident energy density (E_{exc}) and the corresponding LP density N_{3D} = E_{exc}/(E_{pump}D). Here D = 200 nm, E_{pump} = 4.64 eV; it is assumed that 100% of the pump photons are absorbed and all injected hot electrons relax down to the ground state of the lower polariton branch without losses. The estimation gives an upper bound of the LP density [37]. A distinct non-linearity of the output power is observed at an incident energy density of 1.63 μJ/cm², where the characteristics change from a sub-linear (with slope 0.7) to a more super-linear increase (with slope of 1.75), which corresponds to a threshold polariton density n_{th} = 1.1×10^{17} cm⁻³. The latter is an order of magnitude smaller than the Mott density at which the transition from excitons to e-h plasma takes place in ZnO [93]. The onset of non-linearity is accompanied by a significant
A decrease in the emission linewidth (shown in Fig. 4.13(b)) from 17 meV to ~3.7 meV at the non-linear threshold, which is well below the cavity photon linewidth of ~11 meV estimated from FDTD simulations. It may be noted that we do not observe multimode emission with very small individual linewidths below and above thresholds. Such multimode emission has been attributed to photonic defects leading to localization [11,86] or simply to the different transverse modes in the nanowire [37]. It is possible that such modes are present, but are not detected due to lack of resolution and thermal broadening and the measured linewidth of 3.7 meV is an aggregate of several peaks. For larger excitation intensity, the linewidth increases again. This behavior is commonly observed and is believed to be due to decoherence induced by polariton-polariton interactions. The onset of non-linearity in the LP emission is also accompanied by a very small blueshift (<1 meV) in the emission peak energy with increasing excitation intensity. A small blueshift is desirable and confirms that the coherent emission is from the ground state and not from the cavity mode which is at a higher energy by ~50 meV.
4.2.4 Occupancy

To learn more about condensation in k-space as a function of excitation, we have determined polariton occupancy and scattering rates as a function of \( k_\parallel \). In Fig. 4.14(a), the LP number density per k-state is plotted as a function of the energy difference with \( E(k_\parallel = 0) \) for different excitation levels. The plots are analyzed by using the Maxwell-Boltzmann (MB) distribution, \( N_{MB}(k) = N_0 \exp(-E/k_B T_{LP}) \) or the Bose-Einstein distribution: \( N_{BE}(k) = 1/[\exp(E/k_B T_{LP})(1 + N_0^{-1}) - 1] \), where \( T_{LP} \) is the effective polariton temperature, \( N_0 = N_{LP}(k_\parallel = 0) \). Below threshold, neither distribution fits the data well; at threshold, the data can be fitted with the MB distribution using \( T_{LP} = 323 \text{K} \); and above threshold, a good fit to the data is obtained with a BE distribution, using \( T_{LP} = 380 \) and \( 415 \text{K} \), respectively, for \( P = 1.3P_{th} \) and \( 1.8P_{th} \). The results indicate that a non-equilibrium degenerate LP condensate is produced in the system above threshold. From the LP occupation shown in Fig. 4.14(a), it is possible to deduce the relative scattering rates into the
ground state from an in-plane wave vector \( k_{\parallel} \), for small values of \( k_{\parallel} \) and low excitation densities, by using a steady state solution of the Boltzmann equation for LPs as

\[
W_{in}^{rel}(k_{\parallel}) = \frac{I_{PL}(k_{\parallel})}{I_{PL}(k_{\parallel}=0)} = \frac{W_{in,k_{\parallel}}}{W_{in,k_{\parallel}=0}}
\]

4.1

where \( I_{PL}(k_{\parallel}) \) is the integrated PL intensity at \( k_{\parallel} \) and \( W_{in,k_{\parallel}} \) is the scattering rate into a state with in-plane wave-vector \( k_{\parallel} \). Values of \( W_{in}^{rel} \) as a function of emission angles are plotted for different excitation powers in Fig. 4.14(b). There is no evidence of a relaxation bottleneck and the scattering time decreases at lower \( k_{\parallel} \) values. Again, these are favorable conditions for the formation of a degenerate condensate.

4.2.5 Temporal Measurements

We have performed time-resolved PL (TRPL) measurements with a streak camera to determine the LP relaxation time and recombination lifetime. The system has an overall temporal resolution of \( \sim 5 \) ps. The transient data for excitation powers below, equal to, and above threshold power are depicted in Fig. 4.15(a). The rise time, which principally reflects the filling of the exciton reservoir, in all instances is limited by the system resolution. On the other hand, with increase in excitation power the decay times decrease rapidly due to enhanced polariton relaxation from the exciton reservoir to the \( k_{\parallel} \sim 0 \) states. The data can be analyzed by a simple two-level rate equation model to determine the polariton relaxation dynamics. The exciton reservoir at large \( k_{\parallel} \) values is characterized by a population \( n_{R}(t) \) and a polariton recombination lifetime \( \tau_{R} \), while the ground state at \( k_{\parallel} = 0 \) has a population \( n_{0}(t) \) and a polariton lifetime \( \tau_{0} \). Then

\[
\frac{dn_{R}}{dt} = P(t) - \frac{n_{R}}{\tau_{R}} \frac{n_{R}}{\tau_{relax}}
\]

4.2(a)
where $\tau_{\text{relax}}$ is the relaxation or scattering time of polaritons from the reservoir to $k_\parallel \sim 0$. The value of $\tau_R$ is assumed to be 300 ps since the polariton in the reservoir is mostly exciton-like.

Fig. 4.15 (a) Time resolved photoluminescence measured normal to the sample (from $k_\parallel = 0$) below, at, and above threshold with a streak camera having an overall resolution of 5 ps; theoretical fits obtained from a solution to a two-rate equation model to the measured transients (b) below and (c) at threshold; (d) normalized relaxation time obtained from the two-rate equation model plotted as a function of normalized pump power. The error bars indicate the spread in the normalized relaxation time values that fit the transients.
\[ \tau_0 = \frac{|c|^2}{\tau_c} \] where C is the photon Hopfield coefficient at \( k_\parallel = 0 \) and \( \tau_c \) is the cavity photon lifetime equal to 2 ps. The pump \( P(t) \) is modeled as a Gaussian pulse with a pulse width of 3 ps. Two such theoretical fits are shown as solid lines in Figs. (b) and (c). Values of \( \tau_{\text{relax}}/\tau_0 \) are plotted against excitation power in Fig. 4.15(d). At low powers, polariton cooling occurs primarily by polariton-phonon scattering and \( \tau_{\text{relax}} \) is large. However, with increase in excitation power \( \tau_{\text{relax}} \) decreases rapidly due to the onset of stimulated polariton-polariton scattering, leading to quantum degeneracy in the ground state. The two experiments confirm that a degenerate population of polaritons are formed due to stimulated polariton-polariton scattering in the strong coupling regime, leading to the observed polariton lasing at room temperature.

### 4.2.6 Polarization

![Emission intensities plotted as a function of the angle of the Glan-Thompson linear polarizer for 3 different normalized pump powers.](image)

Fig. 4.16 Emission intensities plotted as a function of the angle of the Glan-Thompson linear polarizer for 3 different normalized pump powers.
The polarization of the output in the strong coupling regime was investigated by placing a Glan-Thompson linear polarizer at the entrance of the monochromator. The measured intensities as a function of the angle between the polarizer and the reference x-axis, for emission normal to the sample (from $k_{\parallel} \sim 0$) are displayed in the polar plot for pump powers below, at an above threshold in Fig. 4.16. The linear polarization is horizontal in all the cases, roughly aligned at $60^\circ$ to the reference x-axis of the microcavity. The dependence of the PL intensity on the angle of polarization can be fitted by a cosine-squared law $I \sim I_{\text{max}} \cos^2(\pi/2 - \theta)$ (shown by a solid line), where $\theta$ is the angle between the polarizer and the NW axis determined from the SEM analysis. The PL intensity peaks when $\theta = 90^\circ$, indicating that the E-field is perpendicular to the c-axis. We observe almost no variation in the degree of linear polarization at these 3 pump powers; this indicates that the polarization is not dependent on the cavity modes (which would favor emission polarized along the c-axis), rather it is intrinsic to the nanowire due to its high asymmetry in emission polarization.

**4.2.7 Conclusion**

In conclusion, a giant Rabi splitting ($\sim 103$ meV) and room temperature polariton lasing with a threshold of $1.6 \ \mu J/cm^2$ has been demonstrated by using a single ZnO nanowire grown by pulsed laser deposition as the active medium embedded in a dielectric microcavity. While occupancy and time-resolved photoluminescence indicate condensation into the ground state, the polariton emission is always linearly polarized due to the geometry.
Chapter 5

Electrically Injected Polariton Laser Diodes

Inversionless ultra-low threshold lasing, obtained by stimulated scattering of exciton-polaritons into the ground state of a strongly coupled semiconductor microcavity, will be useful in applications currently limited by energy consumption. Low threshold polariton lasers with optical excitation are being realized; however, for device applications, it is necessary to demonstrate polariton condensation and coherent emission with non-resonant electrical injection, which still remains a challenge. The difficulty in achieving a polariton condensate with non-resonant excitation lies in overcoming the relaxation bottleneck, which is the result of a decrease in the density of states of the lower polaritons (LPs) when their characteristics on the LP dispersion curve transitions from exciton-like to photon-like. Consequently, polariton-phonon scattering becomes less efficient due to a reduced scattering rate at the bottleneck and the LP lifetime also decreases. Polariton relaxation to the ground state is thereby inhibited. In this chapter, we report the polariton emission characteristics of GaAs-based quantum well microcavity diodes with modulation doping and with the application of an external magnetic field. The relaxation bottleneck is suppressed in both the cases while a distinct non-linear coherent emission appears in presence of an applied magnetic field.
5.1 Multi-QW Microcavity Diode with Modulation Doping

One approach to solving the 'relaxation bottleneck' problem is by introducing polariton-electron scattering, which has been shown both theoretically [94] and experimentally [95, 96] to be more efficient than polariton-phonon scattering. Experiments involving polariton-electron scattering typically involve the introduction of a suitable 2-dimensional electron gas (2DEG) within a wide QW through photo-excitation in an adjacent narrow QW [95, 96]. In an electrically injected device with a QW active region, direct doping of the wells would result in broadening of the emission linewidth due to ionized impurity scattering. A more elegant approach would be to introduce the electrons into the QWs through modulation doping in the barrier regions adjacent to the QWs.

5.1.1 Device Design

The microcavity device used in this study, shown in Fig. 5.1, was grown by molecular beam epitaxy on a n-doped GaAs (001) substrate and consists of an undoped (n ≈ 10^{15} cm^{-3}) 5λ/2 GaAs cavity surrounded by 32 periods of Si-doped (n = 2 x 10^{18} cm^{-3}) GaAs / Al_{0.85}Ga_{0.15}As forming the bottom distributed Bragg reflector (DBR) and 25 periods of Be-doped (p = 4 x 10^{18} cm^{-3}) GaAs / Al_{0.85}Ga_{0.15}As forming the top DBR. Four periods of the DBR adjoining the cavity on both sides were left undoped to minimize doping related losses. Four pairs of undoped 10 nm In_{0.1}Ga_{0.9}As/10 nm GaAs QWs are grown at the antinodes of the cavity photon field to maximize the interaction between the QW excitons and the cavity photons. A 1 nm thick region at a distance of 10 nm below each pair of QWs was Si-doped n-type. Three samples were investigated with a Si-doped concentration of n = 0, 1x10^{17} cm^{-3} and 2x10^{17} cm^{-3} respectively. Since at these levels the modulation doping is highly degenerate, the impurity band overlaps with
the conduction band and the dopants require no energy for ionization and thus the corresponding thermally ionized electron sheet densities are $n_e = 0, 1 \times 10^{10}$ and $2 \times 10^{10}$ cm$^{-2}$ for the 3 samples. Standard photolithography and wet chemical etching techniques were employed to form a mesa of 50 μm diameter. Ring-shaped annular Pd/Zn/Pd/Au p-contact and Ni/Ge/Au/Ti/Au n-contact were formed, for hole and electron injection, respectively, to complete device fabrication. The contacts were deposited on the first doped Bragg pair to minimize Joule heating. The top p-contact has an annular width of 10 μm and therefore covers more than half of the device mesa area and ensures a nearly uniform injection.

5.1.2 Dispersion Characteristics

The dispersion of the resonant modes was measured at 30K by angle resolved electroluminescence, with an angular resolution of 1°, at an injection current density of 0.8
A/cm$^2$ with a high resolution monochromator (spectral resolution ~ 0.03 nm) and detected with a photomultiplier using phase sensitive lock-in amplification. The measured spectra for the sample with a modulation doped sheet electron density of $n_e = 1 \times 10^{10}$ cm$^{-2}$ are shown in Fig 5.2(a). The result shows two distinct polariton peaks, with the LP peak asymptotically approaching the exciton energy at larger angles. Signature of a weak middle polariton (MP) branch is also observed, which has been attributed to the effect of charged excitons or trions ($X^-$) [95,97]. The resonances obtained from the electroluminescence spectra are plotted in Fig. 5.2(b) together with
the calculated dispersion curves (solid lines) of the upper polariton (UP), MP and lower polariton (LP) branches by using the one-to-one correspondence between the emission angle of the out-coupled photons and the in-plane wave number of the polaritons. The interaction between the cavity and exciton modes was modeled with a coupled harmonic oscillator Hamiltonian:

$$\begin{pmatrix}
E_{cav} - j\Gamma_{cav} & V_{X_{hh}} & V_{X^-} \\
V_{X_{hh}} & E_{X_{hh}} - j\Gamma_{X_{hh}} & 0 \\
V_{X^-} & 0 & E_{X^-}
\end{pmatrix}$$

(5.1)

where $E_{cav}$ is the bare cavity photon resonance and $V_{X_{hh}}$ and $V_{X^-}$ are the interaction potentials of the heavy-hole exciton and trion with the cavity mode. The measured values of heavy-hole exciton energy ($E_{X_{hh}} = 1.3779$ eV), exciton linewidth ($\Gamma_{X_{hh}} = 1.2$ meV) and cavity photon linewidth ($\Gamma_{cav} = 0.21$ nm) have been used in the analysis. Only the interaction between the cavity photon and the heavy-hole exciton is considered because the light-hole exciton transition is energetically situated 15 meV above the hh-exciton transition due to biaxial strain in the quantum wells. The measured results exhibit excellent agreement with the values obtained from the calculations for interaction potentials $V_{X_{hh}} = 5.1$ meV and $V_{X^-} = 1.8$ meV and a cavity to heavy-hole exciton detuning $\delta$ of -3.5 meV. To determine the exciton and photon fraction of each polariton branch, the square modulus of the Hopfield coefficients is plotted in Fig 5.2(c) as a function of in-plane momentum. The upper and lower polariton branches are almost exclusively the result of the mixing of the cavity mode and $X_{hh}$. The MP branch is made up of the trion state with contribution from the cavity mode at higher $k_{||}$ values.

5.1.3 Occupancy and Suppression of 'Bottleneck'

In order to determine the effect of the polariton-electron scattering in suppressing the bottleneck and enhancing the rate of polariton relaxation to the $k_{||} \sim 0$ states, the polariton
occupancy in k-space as a function of $n_e$ was calculated from the measured angle-resolved electroluminescence data at an injection current density of 4A/cm$^2$ by using the formula,

$$I_{LP}(k_{||}) = \eta N_{LP}(k_{||}) |C(k_{||})|^2 M/\tau_C$$

where $\eta$ is the collection efficiency, $\tau_C/|C(k_{||})|^2$ is the radiative lifetime of the LPs, $M$ is the number of transverse states included in the detection cone and $|C(k_{||})|^2$ is the photon fraction at a wave-vector $k_{||}$. The number of states within the detection cone is given by

$$M = \frac{D^2}{16} (k_0 \Delta \theta)^2,$$

where $D$ is the diameter of the emission spot, $k_0 = 2\pi/\lambda$ and $\Delta \theta$ is the detection half angle. $\tau_C$ is estimated from the cavity Q, and the value of $\eta$ is estimated by replacing the sample with a source of known power (Ti:sapphire laser with suitable attenuation). The LP number density per k-state is plotted in Fig. 5.3(a). A pronounced relaxation bottleneck at $k_{||} \sim 1.17 \times 10^4$ cm$^{-1}$ is observed for the sample with $n_e = 0$. However for samples with $n_e = 1 \times 10^{10}$ and $2 \times 10^{10}$ cm$^{-2}$, the population of the ground state increases, while the reservoir population remains almost unchanged, and the bottleneck is removed.
To investigate the dependence of occupation of different k-states on injected current density and the possibility of polariton condensation, we measured the angle-resolved electroluminescence at 30K for the sample with \( n_e = 1 \times 10^{10} \, \text{cm}^{-2} \). The data are shown in Fig. 5.3(b). At low injection densities, as mentioned earlier, a distribution without any bottleneck is observed with comparable occupancies between \( k_\parallel = 0 \) and \( 1.23 \times 10^4 \, \text{cm}^{-1} \). At higher injection currents, the occupancy of the ground state shows a super-linear increase, while that of the reservoir shows very little increase. Although a bimodal distribution (with a massively occupied ground state) associated with polariton lasing is not observed, the occupancy profile can be characterized by the Maxwell-Boltzmann distribution: \( N = N_0 \exp(-\frac{E(k)-E(0)}{kT}) \), (where \( E(k) \) is the LP energy at \( k_\parallel \) and \( N_0 \) is the occupancy at \( k_\parallel = 0 \)). It is evident that polariton-electron scattering plays a role similar to polariton-phonon scattering and thermalizes the LP distribution [94].

**5.1.4 Excitation Dependent Luminescence Characteristics**

Finally, the dependence of light output on the injection current density was investigated by measuring the electroluminescence at \( k_\parallel \sim 0 \) as a function of injection current (pulsed to avoid device heating). With increasing current density the UP and LP polariton peaks progressively come closer in energy (Fig 5.4(a)) and merge into a single emission line, signaling the transition to the weak coupling regime. The two peak energies merge at the cavity photon energy of 1.374 eV at \( \theta = 0^\circ \) (see dispersion curve in Fig.5.2 (b)) and for an injection current density of 22 A/cm². At this point the normal modes of the system become cavity photons and QW excitons and the corresponding carrier density is \( 4.2 \times 10^{10} \, \text{cm}^{-2} \), in agreement with previous reports [99]. As the current density increases, linewidth narrowing of the LP emission is not observed. The integrated electroluminescence from the lower polaritons was measured as a function of injection current...
density and the data are shown in Fig.5.4 (b). In the absence of polariton-electron scattering ($n_e = 0$), the intensity grows linearly with injection, which is a signature of polariton-phonon scattering. In contrast, in the presence of such scattering, the measured intensity grows super-linearly with increasing current density (in agreement with the increase in polariton occupation at
A quadratic increase in the LP emission intensity is observed in a small range of current density \((15 \text{ A/cm}^2 \leq J \leq 22 \text{ A/cm}^2)\). The observed super-linear dependence on current is attributed to enhanced relaxation of LPs due to polariton-electron scattering, in agreement with the data of Fig. 5.3(a). The quadratic dependence at the highest injection current densities suggests that the relaxation in this regime is dominated by polariton-polariton scattering [96].

### 5.1.5 Conclusion

In conclusion, the effect of polariton-electron scattering on the polariton relaxation dynamics and output characteristics of an electrically injected GaAs-based microcavity p-i(MQW)-n diode in the strong coupling regime was investigated. Excess electrons were provided in the quantum wells by modulation doping with Si. It is observed that the relaxation bottleneck is suppressed and the emission develops a superlinear dependence on injection current density with increasing density of electrons available for polariton-electron scattering. Polariton lasing is not observed, which suggests that polariton-electron scattering is not adequate for attaining ground state quantum degeneracy.

### 5.2 Effect of Magnetic Field on Polariton Emission Characteristics of a Quantum-Well Microcavity Diode

It is known that in semiconductors a magnetic field shrinks the exciton wave function and enhances the electron-hole overlap, resulting in an increase in oscillator strength \(f\). In a system with strong coupling, since \(\Omega \propto \sqrt{f}\), the vacuum Rabi splitting also increases with field. This behavior has been demonstrated in magneto-photoluminescence and magneto-absorption measurements [100-101]. Here, the effect of an external magnetic field applied to a In_{0.1}Ga_{0.9}As/GaAs quantum well microcavity diode operating in the strong coupling regime is reported. In particular, the effect of the magnetic field on the polariton relaxation has been
investigated. It is seen from angle-resolved electroluminescence measurements that the relaxation bottleneck, commonly observed in this system with non-resonant pumping, is suppressed due to enhanced polariton-phonon scattering under a magnetic field.

### 5.2.1 Device Design

The microcavity device used in this study was very similar to the one used for studying modulation doping effects. It was grown by molecular beam epitaxy on a n-doped GaAs (001) substrate and consists of an undoped \((n \sim 10^{15} \text{ cm}^{-3})\) \(5\lambda/2\) GaAs cavity surrounded by 32 periods of Si-doped \((n = 2 \times 10^{18} \text{ cm}^{-3})\) GaAs / Al\(_{0.85}\)Ga\(_{0.15}\)As forming the bottom distributed Bragg reflector (DBR) and 26 periods of Be-doped \((p = 4 \times 10^{18} \text{ cm}^{-3})\) GaAs / Al\(_{0.85}\)Ga\(_{0.15}\)As forming the top DBR. 4 periods of the DBR adjoining the cavity on each side were left undoped to minimize doping related losses. 4 pairs of undoped 10 nm In\(_{0.1}\)Ga\(_{0.9}\)As/10 nm GaAs quantum wells are grown at the antinodes of the cavity field distribution to maximize the coupling of the emitter region to the photon field in the cavity. Standard photolithography and dry etching techniques were employed to form a mesa of 400 μm diameter. Ring-shaped annular Pd/Zn/Pd/Au p-contact and Ni/Ge/Au/Ti/Au n-contacts were formed, for hole and electron injection, respectively, to complete device fabrication. The contacts were deposited on the first doped Bragg pair to minimize Joule heating. The top p-contact has an annular width of 100 μm and therefore covers more than half of the device mesa area and ensures a nearly uniform injection. A micro-photoluminescence (PL) emission peak at 891 nm with a linewidth of 0.16 nm was measured, which gives a cavity Q of 5568. The experimental Q factor is limited by the numerical aperture of the objective lens which collects emission from zero and non-zero in-plane wave vectors within the collection angle. Finite difference time domain (FDTD) simulations yield a cavity Q of \(\sim 12,000\) and such high values are important for polariton condensation in
planar GaAs microcavities [102]. Photoluminescence spectra measured from the bare QWs at 4.2K for different values of applied magnetic field are shown in Fig. 5.5. The zero-field spectrum shows the e$_1$-hh$_1$ exciton peak at 1.387 eV and the e$_1$-lh$_1$ exciton at 1.402 eV. With increasing magnetic-field strength, there is a clearly observable diamagnetic shift of the exciton to higher energies.

### 5.2.2 Dispersion Characteristics in presence of Magnetic Field

The dispersion of the resonant modes was measured at 4.2K under zero magnetic field by angle resolved electroluminescence (with an angular resolution of 1°) at an injection current of 0.1 mA with a high resolution monochromator (spectral resolution ~ 0.03 nm) and detected with a photomultiplier using phase sensitive lock-in amplification and the spectra are shown in Fig 5.6(a). The two distinct polariton peaks are clearly seen, with the lower polariton (LP) peak asymptotically reaching the exciton energy at larger angles. Similar results at 4.2 K are also obtained in the photocurrent spectra (also shown in Fig. 5.6(a)) of the device biased negatively (-
1.5 V) as a photodiode. The sample was mounted in a cryostat with facilities for sample rotation.
and was excited by a tunable narrow bandwidth cw Ti:sapphire laser. The resonances obtained from electroluminescence and photocurrent measurements are plotted in Fig. 5.6(b) together with the dispersion curves of the upper polariton (UP) and LP branches, by using a one-to-one correspondence between the angle of the out-coupled photon and the in-plane wave number of the polaritons. The measured data are analyzed by the coupled oscillator model using a measured exciton linewidth of 1.2 meV at 4.2K and a cavity photon linewidth of 0.16 nm and the calculated dispersions are shown by the solid curves. From these a Rabi splitting of 6.1 meV and a negative detuning $\delta$ of -1.7 meV are derived.

Electroluminescence (EL) measurements were next made in the presence of a magnetic field in the Faraday geometry by placing the device in a magneto-optical cryostat. Angle-resolved measurements similar to that shown in Fig. 5.6(a) were made at fields varying from 1-7 Tesla at a constant injection current of 0.1 mA with an Ocean Optics HR 4000 spectrometer having a spectral resolution of 0.84 nm. To study the effect of magnetic field on the on-resonance polariton, a detuning of -1.7 meV is maintained at each field by varying the temperature. The temperature is increased with increasing field to compensate for the measured diamagnetic shift of the exciton energy to higher energies; an increase of temperature decreases the exciton transition energy while the oscillator strength remains fairly constant in this temperature range [103]. The measured on-resonance polariton electroluminescence data at a detection angle of 9.5° are shown in Fig. 5.6(c). Figure 5.6(d) shows the increase of the Rabi splitting with magnetic field, obtained from the relevant dispersion curves at different fields, assuming a constant exciton linewidth. The exciton fraction also increases from 0.355 to 0.395 in this range of fields.

**5.2.3 Occupancy as a function of Magnetic Field**
The polariton occupancy in k-space as a function of injection current and magnetic field was determined from the measured angle-resolved electroluminescence data. The LP number density per k-state is plotted in Figs. 5.7(a) and (b). A pronounced relaxation bottleneck is observed at low injection current densities for zero magnetic field, with the bottleneck persisting at higher injection levels for low magnetic fields. On the other hand, a distribution with an increase in occupancy near $k_\parallel \sim 0$ with complete suppression of the bottleneck evolves for a field $\geq 5$ Tesla.

Fig. 5.7 (a) Occupancy of LPB as a function of in-plane wave vector for different magnetic fields and injection currents obtained from angle-resolved electroluminescence; (b) occupancy of LPB for various injection currents at $B = 7$ Tesla; (c) variation of the lower and upper polariton peak energy positions with injection. Inset shows the light current characteristics of a VCSEL made with the same heterostructure; (d) integrated emission intensity measured in the normal direction versus injection current density at 0 and 7 Tesla.
Figure 5.7(b) illustrates the occupancy as a function of current injection for $B = 7$ Tesla. At low injection, the occupancy is comparable at all in-plane wave vectors with no observable relaxation bottleneck. At high injection levels, the occupancy of the ground state shows a non-linear increase, while that of the reservoir remains fairly constant. Figure 5.7(c) shows the energy of the two polariton peaks as a function of injection and their merging at a current density of $\sim 15$ A/cm$^2$, signaling the transition to the weak coupling regime and a Mott transition density of $\sim 1.1\times10^{11}$ cm$^{-2}$. This data confirms that strong coupling conditions exist even at the highest injection levels shown in Figs. 5.7(a) and 5.7(b). A similar device with a mesa diameter of 20 $\mu$m was fabricated with the heterostructure of Fig. 1(a). The measured photon lasing characteristics of this vertical cavity surface emitting laser (VCSEL) is shown in the inset of Fig. 5.7(c), with a threshold of 9.1 kA/cm$^2$.

### 5.2.4 Analysis of the Non-linear Characteristics

In seeking an explanation for the occupancy data of Figs. 5.7(a) and (b) and the suppression of the relaxation bottleneck with the magnetic field, the relative polariton-acoustic phonon and polariton-polariton scattering rates [104] into the $k_{//} = 0$ state was calculated. Application of a magnetic field parallel to the growth direction increases the exciton binding energy and decreases the 2D Bohr radius ($\alpha 1/\sqrt{B}$ in the high field limit above 3.5 Tesla). This causes an increase in the exciton oscillator strength which in turn increases the Rabi splitting, as confirmed by our measurements. Furthermore, for a fixed negative detuning, an increase in the Rabi splitting increases the exciton fraction and the density of states of the LP modes. Since the polariton-phonon scattering rates are proportional to the density of states of the polariton modes at the scattering energy, the exciton fraction of the initial and the final states, and overlap integrals between the excitons in the quantum wells and the phonons in the bulk, an increase in
the Rabi splitting with increasing magnetic field should enhance the rates (for the same detuning). It is seen that the polariton-phonon scattering rates from the bottleneck angle to the polariton ground state as a function of in-plane wave vector and applied magnetic field. Inset to (a) shows the increase in the relative polariton-acoustic phonon scattering rates in the bottleneck region.

Fig. 5.8 Calculated relative (a) polariton-acoustic phonon and (b) polariton-polariton scattering rates from $k_\parallel$ to the polariton ground state as a function of in-plane wave vector and applied magnetic field.
ground state increases approximately by an order of magnitude in the magnetic field range of 0-7 Tesla (Fig. 5.8(a)). The polariton radiative lifetime at $k_\parallel = 0$ also increases by a factor of 1.2 in the same range of fields. It is suggested that these factors may be instrumental in suppressing the bottleneck. The slight decrease in the polariton-polariton scattering rates (Fig. 5.8(b)) will not affect the overall picture. We also rule out any role played by electron-polariton scattering in the suppression of the relaxation bottleneck [94-97] since the active region in the devices is undoped. Finally, the integrated electroluminescence from the lower polaritons at $k_\parallel \sim 0$ was measured as a function of injection current (pulsed to avoid device heating) and applied magnetic field. The data are shown in Fig. 5.7(d). A distinct non-linearity appears in the light-current characteristics with application of magnetic field. At 7 Tesla, the threshold of the non-linearity is at 0.32 A/cm$^2$, well within the strong coupling regime. We believe that the observed non-linearity is related to the enhanced polariton-phonon scattering rates and a suppression of the relaxation bottleneck in the presence of a magnetic field, leading to a faster relaxation of the LPs down to $k_\parallel \sim 0$ and a subsequent enhancement of the electroluminescence in the normal direction [28,105].

5.2.5 Conclusion

In conclusion, suppression of bottleneck and enhanced relaxation to the ground state was demonstrated with the application of a magnetic field under constant negative detuning.

5.3 Exciton-polariton Laser Diode

In this work, evidence of polariton lasing from a planar modulation doped microcavity under electrical injection in the presence of a magnetic field is provided. Under a sufficiently strong magnetic field, the current saturation density at which the exciton-microcavity system crosses
over from the strong coupling regime to the weak coupling regime is increased due to a reduction of the exciton Bohr radius. As a result, the system is allowed to remain in the strong coupling regime at higher injection current densities, which increases the possibility of observing polariton lasing before the saturation density is reached. We observe linewidth narrowing of the lower polariton emission at the onset of the first nonlinear threshold at a current density that is more than three orders of magnitude lower than the photon lasing threshold on the same device. The LP population distribution in the momentum space shows condensation of LP at $k_\parallel \sim 0$ and confirms polariton lasing from the ground state. We also show evidence of a local quasi-BEC formed in the microcavity at the polariton lasing threshold from first-order spatial coherence measurements. This is the first report of polariton lasing in a solid state microcavity.

5.3.1 Device Design and Dispersion Characteristics

The $p-i-n$ multi-QW microcavity diode used in this experiment is described in section 5.1.1. To introduce electrons into the QWs, a 1 nm thin layer at a distance of 10 nm above each pair of QWs were Si(n)-doped with a concentration of $n = 1 \times 10^{17} \text{ cm}^{-3}$. This corresponds to a thermally ionized electron sheet density of $n_s = 1 \times 10^{10} \text{ cm}^{-2}$ in the QWs. The quality factor (Q) of the cavity is estimated to be \( \sim 6000 \) based on a low temperature (20 K) micro-photoluminescence measurement of the cavity mode at 1.392 eV with a linewidth $\gamma_c = 0.23 \text{ meV}$ corresponding to a cavity mode lifetime of $\tau_c = 3 \text{ ps}$. EL spectra as a function of detection angle and the corresponding dispersion curve in the absence of applied magnetic field has already been shown in Fig. 5.1. Angle-resolved EL from the device was then measured in the presence of a magnetic field of 7 Tesla with $J = 2 \text{ A/cm}^2$ is shown in Fig. 5.9. The sample was placed inside a magneto-optical cryostat with the magnetic field being parallel to the growth axis of the microcavity structure (Faraday geometry). In this experimental configuration, it is well known
that in the high field limit (> 3 Tesla), the applied magnetic field can reduce the exciton Bohr radius (~1/√B) to ~ 6 nm and increase the QW oscillator strength and hence the Rabi splitting. In Figure 5.9, it can be seen from the angle-resolved spectra that the exciton energy has increased to 1.3803 eV as a result of the diamagnetic shift caused by the magnetic field. The spectra reveal anti-crossing between a LP branch, a weak MP branch originating from the trion (X')-photon interaction and a UP branch. The error bars denote uncertainty in the peak energy position. (c) The calculated square modulus of the Hopfield coefficients for each of the polariton branches showing the contributions from the exciton and cavity modes.

Fig. 5.9 (a) Measured angle-resolved electroluminescence spectra and (b) the corresponding dispersion characteristics obtained from the solutions to a 3x3 coupled harmonic Hamiltonian at B = 7 Tesla for a current density of 2 A/cm² injected in the continuous mode (CW). The spectra reveal anti-crossing between a LP branch, a weak MP branch originating from the trion (X')-photon interaction and a UP branch. The error bars denote uncertainty in the peak energy position. (c) The calculated square modulus of the Hopfield coefficients for each of the polariton branches showing the contributions from the exciton and cavity modes.
more negatively detuned ($\delta = -5.9$ meV). The increase of Rabi splitting improves the robustness of the strongly coupled system and allows strong coupling to persist up to higher injection currents.

5.3.2 Polariton Lasing Characteristics

Electroluminescence from the device as a function of injection current density was then measured in the normal direction under a magnetic field of 7 Tesla. In Figure 5.10(a), a clear nonlinear threshold is observed at a current density of $\sim 11$ A/cm$^2$, accompanied by a reduction of LP emission linewidth from 1.2 to 0.68 meV. The linewidth narrowing at the threshold indicates an increase in the first-order temporal coherence of the ground state LPs. As the current density increases above threshold the LP emission linewidth increases again due to the exciton-exciton interaction. We have calculated the LP density from the injection current density by using the relation $n_{LP} = \eta_i J \tau / q$, where $\eta_i$ is the internal quantum efficiency, $J$ is the current density and $\tau$ is the exciton lifetime. By assuming $\eta_i = 1$ and using an estimated exciton lifetime of 500 ps $\sim$ 1 ns, the polariton density at the nonlinear threshold is found to be in the range of $(3.3 \sim 6.5) \times 10^{10}$ cm$^{-2}$, which is slightly above the calculated critical density ($\sim 2 \times 10^{10}$ cm$^{-2}$) for Kosterlitz-Thouless (KT) transition in In$_{0.1}$Ga$_{0.9}$As/GaAs QWs at 30 K. This means that it is possible for the KT phase transition to occur in our system, and both the normal and superfluid phases can coexist. As can be seen in Figures 5.10(b) and 5.10(c), with further increase in injection current, the polariton linewidth broadens, the Rabi splitting decreases and eventually the system enters the weak coupling regime with the emission taking place from the bare cavity mode. The transition to weak coupling regime takes place at $J = 70$ A/cm$^2$ or a Mott density of $1.5 \times 10^{11}$ cm$^{-2}$. The blueshift of LP energy due to exciton-exciton self-interaction cannot be identified here,
because the change of Rabi splitting with the injection current is much more significant. With
increase in current injection (pulsed to avoid device heating), photon lasing is observed with a threshold \( J = 3 \times 10^4 \text{ A/cm}^2 \) that is \( \sim 3 \times 10^3 \) times the polariton lasing threshold. The two-threshold behavior is shown in Fig. 5.10(d) at \( B = 7 \) Tesla with the photon lasing spectrum at \( J = 4 \times 10^4 \text{ A/cm}^2 \) as an inset.

It is important to note that the combined effects of modulation doping and magnetic field contribute to polariton lasing. It has been shown both experimentally and theoretically that polariton-electron scattering is more efficient than polariton-phonon scattering and helps reduce the relaxation bottleneck that occurs at the region where the LP characteristic transitions from more exciton-like (higher \( k_\parallel \)) to more photon like (lower \( k_\parallel \)). This additional mechanism is added to the already existing polariton-acoustic phonon and polariton-polariton scattering to provide a more efficient means for LPs to overcome the bottleneck, reach the lower energy states and trigger bosonic final-state stimulation. It is not sufficient, however, to keep the system in the strong coupling regime by only having a higher scattering rate. The system may still not reach the quantum degeneracy threshold in the ground state before the saturation density is reached. On the same device, we have measured the total integrated EL intensity as a function of injection current density in the absence of magnetic field at 30 K (see section 5.1). The system entered the weak coupling regime just before the onset of the nonlinear threshold, which confirms that the saturation density was reached before polariton lasing could take place. The application of magnetic field solves this problem by reducing the 2D Bohr radius of the excitons and thereby increasing the exciton saturation density (\( \sim 1/a_B^2 \)), which allows the system to remain in the strong coupling regime up to higher current densities. The reduction of Bohr radius and the increase of Rabi splitting also increase the polariton-phonon scattering rates [106] and enhance the relaxation of polaritons.
5.3.3 Occupancy and Condensation

In order to investigate the condensation of polaritons and their relationship with the observed non-linearity with a distinct threshold injection current, we measured the LP population distribution in k-space below and above threshold with an applied magnetic field of 7 Tesla by angle-resolved electroluminescence. In Figure 5.11(a-c), we have shown the polariton emission intensity in the momentum space for different injection current densities as false color plots. The plots reveal that below the threshold current density, the LP emission has a broad distribution in both energy and momentum, whereas above threshold, the emission originates from a condensate formed at $k_\parallel \sim 0$ with a linewidth of 0.8 meV and $\Delta k \sim 4.3 \times 10^3$ cm$^{-1}$ (note that the experimental angular resolution is 1° which corresponds to a $\Delta k \sim 1.2 \times 10^3$ cm$^{-1}$). We calculated the LP occupancy from the angle-resolved EL measurements. The result of occupancy for different injection current densities is shown in Figure 5.11(d). Below the polariton lasing threshold at $J = 2$ A/cm$^2$, the distribution of LPs can be fitted by a thermal Maxwell-Boltzmann (MB) distribution with an effective temperature $T_{LP} = 160$ K. This suggests that even below the condensation threshold electron-polariton scattering effectively thermalizes the LP distribution [94]. At the lasing threshold $J = 12$ A/cm$^2$, the occupancy follows the MB distribution with $T_{LP} = 70$ K. Finally, at $J = 25$ A/cm$^2$ above the threshold, the polariton density reaches $\sim 1 \times 10^{11}$ cm$^{-2}$ and the LP distribution can be characterized by the Bose-Einstein (BE) distribution with $T_{LP} = 280$ K. It is known that polariton lasing does not require thermal equilibrium within the polariton Bose gas. However, as observed, with the fast relaxation kinetics in the presence of modulation doping and the applied magnetic field, the distribution of polaritons can be described by some thermal distribution with an effective temperature $T_{LP}$ that is larger than the temperature of the crystal lattice $T_{latt}$. The polariton gas is therefore in self thermal equilibrium.
5.3.4 First-order Coherence and Polarization

The long-range spatial coherence properties of the polariton emission were measured with a slightly misaligned Michelson interferometer. Figure 5.12A plots the contrast of the fringes,
Fig. 5.12 First-order spatial coherence and degree of polarization of light output. (A) Interference fringe contrast measured as a function of the displacement between a double image of the polariton emission for two injected current densities. The FWHM of the contrast is ~10 μm above threshold and is a measure of the size of the local quasi-condensates formed throughout the 200 μm mesa. (B) Degree of linear $\rho_{\text{lin}}$ and circular $\rho_{\text{cir}}$ polarization measured as a function of injected current density. Both values increase from a noise-limited value of 10% below threshold (~12 A/cm$^2$) to ~51% for $\rho_{\text{lin}}$ and to ~41% for $\rho_{\text{cir}}$ above threshold.

$$\frac{(I_{\text{max}} - I_{\text{min}})}{(I_{\text{max}} + I_{\text{min}})}$$

(where $I_{\text{max}}$ and $I_{\text{min}}$ are intensities of the bright and dark fringes) as a
function of displacement (x) between the two images of the polariton emission. The measurement gives an estimate of the spatial coherent length of the condensate. Below threshold, the contrast remains within the noise margin for any overlap between the images, indicating that there is negligible coherence across the emission. At threshold, with $J = 12 \text{ A/cm}^2$ the contrast increases to 15%, whereas above threshold, at $J = 25 \text{ A/cm}^2$, a maximum value of 21% is observed for complete overlap between the two images. The full width half maximum (FWHM) of the contrast data is $\sim 10 \text{ μm}$, indicating the finite size of the condensate formed above threshold. The presence of finite-sized condensates in a 2D microcavity has been predicted by theoretical calculations [107] and has been experimentally observed in a CdTe system (3, 4). For such a finite-sized quasi-condensate the critical density at $J = 25 \text{ A/cm}^2$ is estimated to be $n_c = 3.3 \times 10^{10} \text{ cm}^{-2}$ for $T = 30 \text{ K}$. Finally, the polarization of the LP emission was measured as a function of the injected current density. The emission was found to be almost depolarized below threshold (12 A/cm²) and elliptically polarized above threshold with the degree of linear polarization $\rho_{\text{lin}}$ increasing to 0.51 and the circular polarization $\rho_{\text{cir}}$ to 0.41 from noise limited values. The linear polarization is along a [110] crystallographic axis [13,23,61]. The circular polarization originating from the same condensate builds up due to the Zeeman splitting, induced by the magnetic field, which favors LP condensation in the $\sigma^+$-state [108,109].

5.3.5 Conclusion

In conclusion, polariton lasing from a GaAs-based modulation-doped quantum well-microcavity diode in the strong coupling regime with the application of an external magnetic field is demonstrated. The combined effects of modulation doping and magnetic field contribute to polariton lasing. A distinct non-linearity is observed in the light-current output characteristics with a threshold of 12 A/cm² at 30K under a magnetic field of 7 Tesla. Conventional photon
lasing is observed in the same device at higher injection levels. A local quasi-condensate of polaritons is observed at the polariton lasing threshold, which is also confirmed by the polarization measurements of polariton emission. The results presented here demonstrate the first electrically injected polariton laser in a solid state system. Although our device operates at cryogenic temperatures under a magnetic field, it elucidates the issues involved in obtaining such lasing and opens the door to future research on such devices, which have the potential to become the next generation low-threshold highly efficient coherent light sources.
Chapter 6

Bose-Einstein Condensation in a Single Nanowire Microcavity

Research in the field of microcavity exciton-polaritons with semiconductors has been driven with two interrelated goals. The first is the achievement of a coherent light source, also termed a *polariton laser* (see chapters 4 and 5), wherein a degenerate and coherent state of exciton-polaritons is generated by a combination of polariton-phonon and stimulated polariton-polariton scattering. The coherent polariton states generate coherent light by spontaneous radiative recombination. While the polariton lifetime has to be comparable to the relaxation times for such lasing to occur, the system is in a metastable condensed state in which the bosonic polaritons in the degenerate condensate are only in equilibrium amongst themselves, and not with the lattice. At the other extreme is the case of a degenerate Bose-Einstein condensate in perfect thermal equilibrium with the lattice and the polariton lifetime is extremely long, leading to long-term temporal coherence and long-range spatial coherence [60,61,110]. The attainment of Bose condensation of polaritons in a single GaN nanowire strongly coupled to a dielectric microcavity and in a graded Al(Ga)N spatial trap in a similar microcavity with evaporative cooling are described. A BEC in equilibrium at 300K is demonstrated for the latter case.
6.1 Effect of Detuning on Dynamic Polariton Condensation in Single GaN Nanowire-Dielectric Microcavity

In Chapter 3, the strong coupling characteristics of a single GaN nanowire embedded in a dielectric microcavity[37] was reported and it is evident that the single nanowire-dielectric microcavity is an ideal system to study the effects of exciton-cavity photon detuning on strong coupling and dynamic Bose condensation. In this work, the time-integrated momentum distribution and dynamics of polaritons in a single GaN nanowire-dielectric microcavity by angle-resolved and time-resolved spectroscopy have been investigated. Measurements have been made over the temperature range of 25-100K (a corresponding detuning range of -3.0 to +4.2 meV). Polariton lasing is observed at all temperatures, showing a progression from the kinetic to the thermodynamically controlled regime, with a minimum threshold excitation power at 50K. In terms of dynamic Bose condensation, the best results are observed at 85K with a positive detuning of 2.3 meV. The temperature of the lower polaritons $T_{LP}$ becomes almost equal to the temperature of the lattice $T_{latt}$ and the polariton relaxation time is smaller than the polariton lifetime at $k_\parallel \sim 0$ by a factor of 2.

The sample being characterized is a single GaN nanowire of diameter 60 nm and length 750 nm placed at the central antinode of a dielectric microcavity. The latter consists of a SiO$_2$ $\lambda$-cavity sandwiched by distributed Bragg reflectors (DBRs) on either side consisting of seven pairs of SiO$_2$/TiO$_2$ layers. The microcavities that have been investigated are mesa-shaped of 10 $\mu$m diameter. A detailed description of the sample and the experimental techniques used for its fabrication have been described elsewhere[37]. A typical micro-photoluminescence spectra from the nanowire before being inserted into the cavity exhibit the three free exciton and the corresponding donor bound (DB) transitions. Of these the exciton $X_A$ and DB$X_A$ are the dominant transitions.
6.1.1 Integrated Light Output Characteristics

The time integrated momentum distribution of the lower polaritons were determined from angle resolved photoluminescence measurements by non-resonant excitation with the linearly polarized emission of a frequency tripled (\(\lambda = 267\) nm) Ti:sapphire laser. These measurements were made at temperatures ranging from 25-100K. The spectra at all angles of the out-coupled photon are characterized by a strong LP resonance and a weaker peak corresponding to the upper polariton (UP) (see Chapter 4; section 4.1.2). Using the one-to-one correspondence between the angle of the out-coupled photon and the in-plane wave vector of polaritons, we obtain the energy-momentum distribution of the polaritons. These are shown as false color plots in Fig. 6.1
at different excitation levels for measurements made at 85K. The dashed lines represent the dispersion curves obtained from a solution of the coupled harmonic oscillator model considering only the coupling of the $X_A$ exciton and the cavity mode. Values of Rabi splitting $\Omega_R = 48$ meV and detuning $\delta = +2.3$ meV have been used in the analysis. The plots in Fig. 6.1 reveal that below threshold excitation, the LP emission has a broad distribution both in energy and momentum, whereas above threshold, the emission originates from a condensate formed at $k || ~ 0$ with a linewidth of 0.78 meV and $\Delta k \sim 7 \times 10^3$ cm$^{-1}$ (note that the experimental angular resolution is $1^\circ$ which corresponds to a $\Delta k \sim 3 \times 10^3$ cm$^{-1}$). The non-linear optical properties of the nanowire microcavities were also determined from the measured photoluminescence in the normal output direction. A very distinct superlinear increase of the luminescence is observed at the onset of stimulated LP-LP scattering, accompanied by a large decrease in the emission linewidth and a small blueshift of the LP emission peak. For example, at 70K the linewidth decreases from a maximum of 11 meV to a minimum of 0.58 meV, corresponding to a coherence time of 2.27 ps. The blueshift is 1.83 meV. The threshold polariton density, $n_{th}$, for the onset of non-linearity derived from the relation $n_{th} = E_{exc}/(E_{pump}D)$ (where $D = 60$ nm, $E_{pump} = 4.64$ eV and it is assumed that 100% of the pump photons is absorbed) is plotted in Fig. 2 as a function of detuning. A minimum threshold excitation energy of 57 nJ/cm$^2$ and a corresponding $n_{th} = 1.27 \times 10^{16}$ cm$^{-3}$ is observed for a negative detuning of $\delta = -1.3$ meV at a temperature of 50K. At large negative detunings, the polariton-phonon scattering rate is small, the relaxation kinetics is insufficient and the threshold is therefore kinetically limited. At the other extreme, in the positive detuning regime, the large polariton effective mass increases the threshold for stimulated scattering and the measured threshold is again high. At the minimum, both kinetics and thermodynamics play an equal limiting role. The calculated critical densities for the formation of
a condensed phase [111-113] in this system at different detunings (temperatures) are shown by the solid curves in Fig. 6.2. The equation used is:

\[
\begin{align*}
n_c &= \frac{1}{S} \sum_{\mu \to 0} \frac{1}{\exp\left[\frac{E(k) - E(0) - \mu}{k_B T}\right]} \\
&= \frac{1}{(2\pi)^2} \int_{\mu \to 0}^{2\pi/D_S} \frac{k dk}{\exp\left[\frac{E(k) - E(0) - \mu}{k_B T}\right]}
\end{align*}
\]  

Fig. 6.2 Variation of the measured polariton density at threshold and the calculated critical density for the formation of a degenerate condensed phase, plotted as a function of detuning (temperature).

where \( E(k) \) is the LP energy at wave vector \( k \) and \( E(0) \) is the energy of the ground state. The chemical potential \( \mu \) tends to zero. \( D_S \) is the size of the system (in this case the microcavity diameter) and \( 2\pi/D_S \) is the lowest \( k \)-state. To convert the critical density from 2D to 3D, we have divided by the nanowire diameter.
6.1.2 Dependence of Scattering Rates and Critical Density for Condensation on Detuning and Temperature

It is known that the rate of polariton-phonon scattering is dependent on both detuning and temperature [114-116]. The effects of the two parameters on polariton-phonon interaction can be separately examined by varying them independently in calculating the scattering rates. For a fixed detuning and a variable temperature, the probability of scattering into the ground state increases with temperature due to an increase in phonon occupation number. Similarly, given a fixed temperature and a variable detuning, the probability of scattering into the ground state is enhanced with increasing detuning by an increase in the exciton fraction of the ground state and the density of states of the LPB. The relative probability of scattering into the ground state is calculated for the two cases. For variable detunings (δ ranging from -3 meV to +4.2 meV and the temperature fixed at 25K), the probability of scattering from a state k into the ground state is calculated relative to the same state at a detuning of -3 meV and is shown in Fig. 6.3(a). Every k-state is seen to experience an increase in the scattering rate down to the ground state as the detuning increases, with the state adjacent to the bottom of the LPB experiencing an increase by a factor of ~2 for a positive detuning of +4.2. For variable temperatures (temperature varying from 25K to 100K and δ fixed at -3 meV) the probability of state k scattering to the ground state is calculated relative to the same state at a temperature of 25K and is shown in Fig. 6.3(b). Here we again see a monotonic increase in the probability of scattering from every k-state to the ground state as the temperature increases. The enhancement is greatest near the bottom of the LPB, with the state adjacent to the ground state experiencing an increase by a factor of ~ 4 in its scattering rate as the temperature increases from 25K to 100K. The effect of temperature on the kinetics is more pronounced than the effect of detuning, however both parameters are instrumental in the realization of a degenerate condensate in thermal equilibrium. The effects of
detuning and temperature on the critical density for condensation were also studied. Figure 6.3(c) plots the critical density as a function of both detuning ($\delta$ ranging from -3 meV to +4.2 meV) and temperature (temperature varying from 25K to 100K). It is evident that the effect of temperature on the critical density in the thermodynamic regime is more pronounced compared to the effect of detuning.

Fig. 6.3 (a) Polariton-phonon scattering rate to the ground state as a function of in-plane wave vectors for (a) variable detunings at a fixed temperature of 25K calculated relative to the same state at a detuning of -3 meV and (b) variable temperature at a fixed detuning of -3 meV calculated relative to the same state at a temperature of 25K; (c) critical density for condensation calculated in the thermodynamic regime as a function of temperature and detuning.
6.1.3 Momentum Space Distribution

To investigate dynamic condensation in more detail the polariton occupancy in k-space as a function of excitation was studied. For this the time-integrated intensity of the angle-resolved LP emission into the number density of LPs by taking into account the \( k_{||} \)-dependent density of states and the LP radiative lifetime weighted by the relative photon fraction of the polaritons (see chapter 3). It may be noted that in the pulsed excitation scheme used here to avoid sample heating, the polariton temperature and density will change with delay after the excitation. However, in the regime of excitation in which polariton relaxation time is larger than or comparable to the polariton emission time \( (P \leq 1.7P_{th}) \), the distribution of LP density in \( k_{||} \)-space remains almost invariant. A time-integrated emission intensity from the pulsed measurement is a good approximation for the LP density under these conditions. Far above threshold, the approximation is no longer valid due to the faster polariton dynamics. In Fig. 6.4(a), the LP number density per k-state at 85K is plotted against energy difference \( E - E(k_{||}=0) \) for different excitation levels. Below threshold, the scattering mechanisms do not allow for LPs to have enough time to scatter to the ground state and the distribution is non-thermal. At threshold, the kinetics is fast enough and a metastable state is reached wherein the polaritons thermalize via polariton-phonon scattering and which is well described by a Maxwell-Boltzmann (MB) distribution: 

\[
N_{MB}(k) = N_0 \exp(-E/k_B T_{LP})
\]

The solid line is a fit to the data with \( N_0 = 1 \) and \( T_{LP} = 88.8K \). Above threshold, a Bose-Einstein distribution: 

\[
N_{BE}(k) = 1/\left[\exp(E/k_B T_{LP})(1+ N_0^{-1}) - 1\right]
\]

describes the data very well except for an occupancy of the condensate at \( k_{||} = 0 \). The variable \( T_{LP} \) and \( N_0 \) which are, respectively, the LP temperature and the LP population at \( k_{||} = 0 \), are used as fitting parameters. The fitting with the BE distribution at \( P = 1.3P_{th} \) was achieved with \( T_{LP} = 90.2 \) K and \( N_0 = 3.87 \). As the excitation density is increased, \( T_{LP} \) increases and this
dependence is plotted in the inset of Fig. 6.4(a). \( T_{LP} \) remains constant at \( \sim 89K \) up to \( 1.2P_{th} \), and then increases rapidly. It can be said, with some caution, that with \( T_{latt} = 85K \) and \( T_{LP} = 88.8K \) the degenerate polariton condensate is in equilibrium with the phonon bath via dynamic Bose condensation and an optimal balance of kinetics and thermodynamics. However, as the pump rate is increased the LP-phonon scattering is not adequate to cool the polaritons and thermal equilibrium with the phonon bath is not reached any more, though the system can still exhibit polariton lasing. Similar measurements and analysis performed in the temperature range of 25-
100K yields the data of Fig. 6.4(b) and those shown in Table 1. The values of $T_{LP}$ shown in the figure and table reflect excitation levels of $P/P_{th} \approx 1$ where the best fit of the occupancy data is obtained with a MB distribution. The normalized chemical potential defined as $\alpha = -\mu/k_B T_{LP} = \ln(1+N_0^{-1})$ obtained from the fit of the data of Fig. 5.4(a) with a MB/BE distribution is plotted in Fig. 6.4(c) as a function of excitation energy for different detunings. The normalized chemical potential gives insight into the establishment of quantum degeneracy in the system since a quantum degenerate polariton gas in thermal equilibrium is described by a Bose-Einstein distribution with the same temperature as the phonon bath, and with a chemical potential close to zero. Quantum degeneracy is achieved when $N_0 = 1$ and consequently $\alpha = \ln2 \approx 0.7$. It is seen that the threshold power increases as the detuning increases from negative to positive, in accordance with the data of Fig. 2. It should be noted that all the data points (detunings) of Fig. 2 are not represented in the plots of $\alpha$ in Fig. 6.4(c). It is interesting to note that the plots of $\alpha$ approximately represent the non-linear polariton lasing behavior as a function of pump power. The quantum degeneracy threshold occurs for $P = P_{th}$ and $N_0 = 1$ and a saturation takes place at higher powers due to phase space filling. The plots of measured integrated light intensity versus pump power are shown in the supplemental material.

6.1.4 Transient Characteristics

The dynamic condensation behavior was also studied by performing time-resolved PL (TRPL) measurements using the same pulsed excitation described earlier and detected with a streak camera having an overall system temporal resolution $\sim 5$ ps. The transient data recorded at 85K for different excitation powers are shown in Fig. 6.5(a). The data have been analyzed (as shown in the inset) by a simple two-level rate equation model[117] describing the dynamics between an exciton-polariton reservoir and the LP ground state at $k_\parallel \sim 0$. The thermalization time
from the hot exciton reservoir down to the ground state is modeled as a single time constant $\tau_{\text{relax}}$. Since our objective is to investigate the effect of the interaction of the polaritons with the thermal phonon bath, we neglect any non-linear time constants and population terms describing the effects of polariton-polariton scattering and stimulated scattering. Accordingly,

$$\frac{dn_R}{dt} = P(t) - \frac{n_R}{\tau_R} - \frac{n_R}{\tau_{\text{relax}}} \tag{6.2}$$

$$\frac{dn_0}{dt} = -\frac{n_0}{\tau_0} + \frac{n_R}{\tau_{\text{relax}}} \tag{6.3}$$

where $n_R$ and $n_0$ are the reservoir and ground state population, respectively, $P(t)$ is the excitation modeled as a Gaussian pulse and $\tau_{\text{relax}}$ and $\tau_0$ are scattering time of polaritons from the reservoir to the ground state and the polariton lifetime in the ground state, respectively. The value of $\tau_0$ is fixed and is determined from the cavity Q ($\sim 600$ that corresponds to a photon lifetime $\tau_{\text{ph}} \sim 0.26$ ps) and the corresponding photon fraction in the LP branch at the ground state ($\tau_0 = \tau_{\text{ph}}/|C_{k||0}|^2$ and $|C_{k||0}|^2$ is determined from the dispersion curves at each temperature). The coupled equations were solved using the non-linear Runge-Kutta technique. Despite the simplicity of the model, the analysis leads to a good fit of the measured data and provides us with a simple insight into the evolution of the thermalization time as a function of pump power. Values of the normalized relaxation time $\tau_{\text{relax}}/\tau_0$ obtained from the analysis of the data of Fig. 6.5(a) with the coupled rate equations (1) and (2) are plotted against $P/P_{\text{th}}$ for various detunings in Fig. 6.5(b). While the model of relaxation invoked here is very simplistic, the data exhibited in Fig. 6.5(b) give some realistic insight to the dynamic condensation process. At low excitation powers polariton relaxation occurs primarily by polariton-phonon scattering and $\tau_{\text{relax}}$ is large. For all detunings there is a sharp decrease of $\tau_{\text{relax}}/\tau_0$ at $P/P_{\text{th}} \approx 1$ which signals the onset of stimulated polariton-polariton scattering and quantum degeneracy at $k_{||} \sim 0$. For the two cases of positive detuning (T
Fig. 6.5 (a) Time resolved LP luminescence measured normal to the sample at 85K ($\delta = 2.3$ meV) and at different excitation powers with a resolution of 5 ps. Inset shows the measured transient (normalized) for $P = 2P_{th}$ with the calculated values in accordance with the rate equations; (b) normalized relaxation time vs pump power derived from data similar to that of (a) at different temperatures (detunings).

Fig. 6.5 (a) Time resolved LP luminescence measured normal to the sample at 85K ($\delta = 2.3$ meV) and at different excitation powers with a resolution of 5 ps. Inset shows the measured transient (normalized) for $P = 2P_{th}$ with the calculated values in accordance with the rate equations; (b) normalized relaxation time vs pump power derived from data similar to that of (a) at different temperatures (detunings).

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TABLE II. Variation of the lower polariton temperature obtained from analysis of occupancy data, with lattice temperature (detuning).

<table>
<thead>
<tr>
<th>$T_{\text{latt}}$ (K)</th>
<th>$\delta$ (meV)</th>
<th>$T_{\text{LP}}$ (K)</th>
<th>Thermal Equilibrium</th>
</tr>
</thead>
<tbody>
<tr>
<td>25</td>
<td>-3</td>
<td>138.46</td>
<td>No</td>
</tr>
<tr>
<td>70</td>
<td>1</td>
<td>92.3</td>
<td>No</td>
</tr>
<tr>
<td>85</td>
<td>2.3</td>
<td>88.84</td>
<td>Yes</td>
</tr>
<tr>
<td>100</td>
<td>4.2</td>
<td>107</td>
<td>Reasonable</td>
</tr>
</tbody>
</table>

6.1.5 Conclusion

In conclusion, we have investigated strong coupling characteristics in a single GaN nanowire-dielectric microcavity over the temperature range of 25 - 100K, which corresponds to an exciton-cavity photon detuning range of -3 to +4.2 meV. Time integrated and time-resolved measurements have been made in this temperature range and the results have been analyzed with appropriate models. Polariton lasing is observed in the entire range of temperature (detuning). The threshold power for the onset of quantum degeneracy, indicated by the onset of non-linear behavior in the lower polariton luminescence goes through a minimum for a detuning $\delta = -1.3$ meV at $T = 50$K resulting from the interplay of kinetic and thermodynamic limitations to dynamic Bose condensation. It is observed that the polaritons reach equilibrium with the phonon bath, and $T_{\text{LP}} \approx T_{\text{latt}}$ at $T_{\text{latt}} = 85$K.
6.2 Polariton Bose-Einstein condensate at room temperature in a Al(Ga)N nanowire-dielectric microcavity with a spatial potential trap

The design of the graded nanowire microcavity with a spatial potential trap is illustrated in Fig. 6.6. The nanowire is grown on silicon and consists of graded Al(Ga)N along the length. Thus a spatial potential trap is created. If the nanowire is selectively excited at the Al$_{0.05}$Ga$_{0.95}$N end, the exciton-polaritons which are more photon-like will relax to lower energies by scattering as they drift and diffuse to the central GaN region where they are less photon-like. The polariton-phonon scattering rate will gradually increase due to the increasing exciton-photon detuning in the microcavity. At the same time the higher energy polaritons (at higher $k_\parallel$ values) will be lost as photons, thereby providing evaporative cooling. The coldest polaritons near $k_\parallel \sim 0$ will reach the bottom of the potential trap in the GaN region. In the following the results of strong coupling experiments performed with such a nitride-based compositionally graded nanowire microcavity are reported. The formation of a polariton BEC in thermodynamic equilibrium at room temperature is demonstrated.

6.2.1 Device Structure and Experimental Methods

The experimental sample consists of a single graded bandgap Al(Ga)N nanowire of length equal to 6 μm and diameter ~ 50 nm embedded in a SiN$_x$ $\lambda$-cavity surrounded by SiO$_2$/SiN$_x$ distributed Bragg reflectors (DBRs) on top and bottom. The nanowire sample is grown by molecular beam epitaxy on (001) silicon substrate and has wurtzite crystalline form with the c-axis parallel to the growth direction. The nanowires are dispersed on half of the SiN$_x$ cavity and
Fig. 6.6 (a) Schematic of the Al(Ga)N nanowire showing the variation of the exciton energy as a function of position. The exciton energies at the AlGaN end and the GaN region are estimated from the measured photoluminescence peaks as shown. Photon and exciton fraction of the lower polariton branch at $k_\parallel \sim 0$ are also shown as a function of position in the trap; (b) schematic representation of the dielectric microcavity with a single Al(Ga)N nanowire of diameter 50 nm and length 6 μm buried in the center of a $\lambda$-sized cavity. The microcavity is etched into mesas of size ~ 10 μm with 1 μm of the Al$_{0.05}$Ga$_{0.95}$N end of the nanowire exposed; (c) high-resolution transmission electron microscope image of a GaN nanowire reveals that the nanowires are relatively free of extended defects and stacking faults. Inset shows the selected area diffraction pattern confirming that the nanowires have a wurtzite structure and grow along the c-axis.
single nanowires are selected by e-beam lithography. Square shaped mesa microcavities of 10 μm side are formed by photolithography and etching on a Si substrate. The nanowire consists of a 2.5 μm Al$_{0.05}$Ga$_{0.95}$N region, followed by 2 μm graded Al(Ga)N, 1 μm GaN and 0.5 μm graded Al(Ga)N terminating in Al$_{0.05}$Ga$_{0.95}$N (Fig. 6.6(a)). The spatial potential profile that is formed is therefore not symmetric. The Al composition at the end of the nanowire was estimated to be 5% from the measured exciton luminescence peak [118] of 356 nm (3.483 eV) (Fig. 1(a)) and from independent X-ray diffraction measurements. The average polariton diffusion length for this range of alloy composition is estimated to be 50 μm (see Appendix). The energy difference between Al$_{0.05}$Ga$_{0.95}$N and GaN, or the height of the spatial trap, is 85.7 meV over a distance of ~4.5 μm. The single nanowire is positioned in the microcavity such that ~1μm of the Al$_{0.05}$Ga$_{0.95}$N region is exposed from the mesa and optical excitation is provided at this end (Fig. 6.6(b)). We will henceforth refer to this sample with the spatial trap as the Al(Ga)N nanowire microcavity (sample 1). A sample consisting of a single GaN nanowire-microcavity (sample 2) having the same cavity photon-exciton detuning $\delta$ was also fabricated for comparison of experimental results. Micro-photoluminescence spectra from the GaN nanowire exhibits the three free excitons $X_A$, $X_B$ and $X_C$ and the corresponding donor bound (DB) transitions. Of these the $X_A$ and DB$X_A$ are the most dominant in the spectra, and hence coupling of the $X_A$ exciton to the cavity mode is only considered for analyzing measured polariton dispersion characteristics with the coupled oscillator model [37].

Time-integrated photoluminescence (PL) measurements were performed on the microcavity samples by non-resonant excitation with the linearly polarized output of a frequency tripled ($\lambda = 267$ nm, $f_{\text{rep}} = 80$ MHz and pulse width of 150 fs) Ti:sapphire laser. A doublet lens was used to focus the incident pump beam at an incident angle of 20° on to the exposed
\[ \text{Al}_{0.05}\text{Ga}_{0.95}\text{N} \] end of the nanowire. The excitons created by optical absorption enter the microcavity and spontaneously form exciton-polaritons which then drift and diffuse to the center GaN region of the nanowire. However, this arrangement does not completely prevent light backscattered by the lower DBR from exciting the entire nanowire. Hence, a finite difference time domain (FDTD) calculation was carried out to investigate the extent of such direct excitation. In the simulation, the source with a wavelength of 267 nm was placed near the edge of the mesa and absorption in the distributed Bragg reflectors was taken into account. The relative energy density inside the mesa with respect to that just below the source was recorded at various incidence angles. It is observed that the direct excitation of the nanowire inside the mesa is at a minimum when the pump laser excitation is at 20° with respect to the normal. The calculated light intensity inside the mesa is only \(~1\%\) of the direct excitation (as shown in Fig. 6.7). Hence the number of exciton-polaritons directly generated in the GaN region of the nanowire is negligible. The luminescence was collected from the center GaN region of the nanowire by a 1.5 μm core fiber patch cord and transmitted to a spectrometer. The collection optics is located on extended rails of a goniometer centered at the sample and has an angular
resolution of 1°. In the case of samples with a GaN nanowire, the entire mesa-shaped microcavity is uniformly illuminated by the same laser.

### 6.2.2 Dispersion and Polariton Lasing Characteristics

The polariton dispersion characteristics of the Al(Ga)N nanowire microcavity was determined from measured angle resolved PL (0°-30°) data recorded for low excitation densities, which show strong polariton luminescence corresponding to the GaN (center) region of the nanowire. The results were analyzed with a coupled oscillator model taking into account the coupling of the X_\text{A} exciton and the cavity mode and values of Rabi splitting Ω = 48 meV and cavity-to-exciton detuning δ = +2 meV are derived. The same value of δ is derived for sample 2 with the single GaN nanowire.

The integrated intensity of polariton luminescence recorded at room temperature in the normal direction (k_∥ \sim 0) as a function of pump power exhibits a non-linear behavior, accompanied by a sharp decrease of the emission linewidth. Figures 6.8(a) -(c) illustrate these characteristics for the Al(Ga)N nanowire microcavity (sample 1). Similar data recorded for the GaN nanowire sample (sample 2) is illustrated in Fig. 5.8(d). The incident excitation energies at the threshold of the non-linearity are 102 nJ/cm$^2$ and 125 nJ/cm$^2$ for sample 1 and 2, respectively, and the minimum emission linewidths are 0.43 meV and 1.1 meV respectively. It may be noted that the reduction in threshold power and emission linewidth agrees with the observation made by Balili et al [119] where similar trends were recorded for an increase in the locally applied stress (trap depth) for the GaAs/AlGaAs QW microcavity. The emission spectra above threshold (Fig. 6.8(a)) exhibits multiple peaks with narrow linewidth corresponding to different discrete LP transverse modes localized in the nanowire. Figure 6.8(c) shows the variation of the LP and UP peak energies. The extremely small shift in energy of the LP mode (~
1.5 meV, compared to $\Omega = 48$ meV) indicates that the system remains in the strong coupling regime over the entire range of excitation energy. In addition, as seen in Fig. 5.8(b) for sample 1, the linewidth of 0.43 meV that persists for a range of excitation is at the resolution limit of the spectrometer. Therefore, the emission linewidth could be smaller than 0.43 meV. The coherence times corresponding to the minimum measured emission linewidths are 9.6 ps (sample 1) and 3.75 ps (sample 2). The upper bounds of the LP density at threshold are $2.2 \times 10^{16}$ cm$^{-3}$ (sample 1)
and $2.7 \times 10^{16}$ cm$^{-3}$ (sample 2), using the relation $N_{3D} < E_{th1}/(E_{pump}D)$. These polariton densities are 3 orders of magnitude smaller than the exciton Mott density$^{33}$ of $3 \times 10^{19}$ cm$^{-3}$. Furthermore, in a single GaN nanowire microcavity device identical to sample 2, we have measured a value of $E_{th} = 92$ nJ/cm$^2$ at 300K. We also measured conventional photon lasing in the same device with an energy threshold 2700 times larger in value$^{37}$. While these characteristics indicate coherent emission from a degenerate and coherent LP condensate, or polariton lasing, they do not confirm that the condensate is in equilibrium with the lattice. It is evident, however, that the incorporation of the spatial potential trap produces noticeable differences in the characteristics of polariton emission.

6.2.3 Momentum Space Mapping

In order to gain a better understanding of the polariton dynamics and the occupation of the LPs in $k_{||}$ space, we have performed angle-resolved PL measurements as a function of excitation density for samples 1 and 2. It may be noted that pulsed excitation has been used and hence the polariton density and temperature may change with delay after the excitation pulse. However, for excitation powers $P \leq 1.7P_{th}$, the polariton relaxation time remains larger than or comparable to the polariton emission time constant and the distribution of LP density in $k_{||}$-space can be considered invariant$^{14,23}$. Under these conditions a time-integrated emission intensity obtained from the pulsed excitation measurement is a good approximation for the LP density. At higher excitation powers, this approximation is not valid due to the higher polariton scattering rates. Below the threshold for non-linear emission (Fig. 6.9) the PL spectra remains broad and of comparable intensity for all angles. Above threshold the emission is characterized by a spectrally narrow and strong peak at $k_{||} \sim 0$. The relative occupancy, or the LP number density per $k_{||}$-state, at 300K is plotted against $E-E(k_{||} \sim 0)$ for different excitation powers in Figs. 6.10(a) and (b) for
the Al(Ga)N nanowire (sample 1) and GaN nanowire (sample 2) microcavities, respectively. Far below the non-linear threshold the polariton scattering rate is inadequate to relax the polaritons and the LP distribution remains non-thermal. Near and above threshold, the relaxation dynamics is fast enough and a dynamic condensation leading to a higher occupancy at or near ground state \((k|| \sim 0)\) is observed. At the higher excitation powers a bimodal distribution of the occupancy accompanied by a sharp increase in LP occupancy near \(k|| \sim 0\) is observed for sample 1 having the spatial potential trap. In the range \((0.9-1.0)P_{th}\), the occupation distribution in \(k||\)-space can be described by the Maxwell-Boltzmann (MB) distribution. At and above threshold, a Bose-Einstein (BE) distribution: \(N_{BE}(k) = 1/[\exp(E/k_B T_{LP})(1+ N_0-1) – 1]\) can be used to analyze the polariton occupation data. For example for \(P = 1.0P_{th}, 1.1P_{th}\), and \(1.3P_{th}\) in sample 1, values of \(T_{LP} = 298.1K, 300.7K\) and \(303.4K\) are obtained from a fit to the data. At higher excitation levels \((P=1.5P_{th})\), thermal equilibrium with the phonon bath is lost, as the hot polaritons generated by the excitation cannot be adequately cooled, and \(T_{LP}\) increases [120]. The significant bimodal polariton distribution of the occupancy \(N(E_{k||})\) at high excitation levels in sample 1 is predicted from steady-state quasi-equilibrium theory [121,122] and is a strong indication of the formation

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Fig. 6.9 Momentum distribution of polaritons at different excitation levels obtained from angle-resolved measurements and displayed as false color plots. The horizontal axis displays the in-plane momentum and the vertical axis displays the emission energy in a false-color scale. The false color scale is linear with red and blue representing high and low values, respectively. The exciton and cavity photon energies are also indicated.
of a Bose condensate. In contrast, a condensate in thermal equilibrium ($T_{\text{LP}} \equiv T_{\text{lattice}}$) and a distinct bimodal distribution are not observed for sample 2 without the spatial potential trap, although the detuning is the same ($\delta = +2$ meV). A value of $T_{\text{LP}} = 357$K is derived for $P = 0.9P_{\text{th}}$ for sample 2. For an ideal BEC, the chemical potential $\mu$ given by $-\mu/k_B T = \ln(1+N_0-1)$ tends to zero. The fit to the occupation data of sample 1 with a BE distribution for $P = 1.1P_{\text{th}}$ yields $\mu = -4.7$ meV. Furthermore, as the false color plots of Fig. 5.9 indicate, at this excitation, the condensate at $k_{||} \sim 0$ is described by extremely small values of $\Delta k \sim 1 \times 10^4$ cm$^{-1}$ and $\Delta E \sim 0.5$ meV.
6.2.4 Transient Characteristics

Time-resolved photoluminescence (TRPL) measurements have been made with the 267 nm pulsed excitation and a streak camera with an overall temporal resolution of 5 ps. The transient data recorded at room temperature for sample 1 are shown in Fig. 6.11(a). The rising part of the transient, which principally corresponds to the filling of the exciton reservoir in the Al$_{0.05}$Ga$_{0.95}$N region of the nanowire with excitation, is limited by system resolution. The decay time decreases with increasing excitation, reflecting enhanced polariton relaxation into the k$\parallel$ ~ 0 states in the GaN (center) region of the nanowire. The transport and scattering of the polaritons from the exciton reservoir in the Al$_{0.05}$Ga$_{0.95}$N region of the nanowire, across the spatial potential trap and into the bottom of the LPB (k$\parallel$ ~ 0) in the GaN region, was explained earlier and illustrated in Fig. 6.11(a). This complex dynamic process can be analyzed by detailed Monte Carlo simulation to determine the polariton scattering times at each point in the trap, which is beyond the scope of the present work. Instead, we describe the evaporative cooling due to emission of hot polaritons escaping as photons, as the polaritons are transported along the trap, and the population redistribution in momentum space due to polariton-phonon and polariton-polariton scattering, at each point in the trap, by a single time constant $\tau_{\text{therm}}$ in the coupled rate equations 6.2 and 6.3.

Here P(t) represents the excitation as a Gaussian pulse centered at t = 0, n$\text{R}$ and n$\text{0}$ represent the populations in the Al$_{0.05}$Ga$_{0.95}$N reservoir and the condensate in the k$\parallel$ ~ 0 in the GaN region, respectively, and $\tau_{\text{0}}$ is the polariton lifetime in this condensate. The exciton lifetime in the reservoir, $\tau_{\text{R}}$, is usually much larger (~ 1 ns) and $\tau_{\text{0}}$ and therefore the corresponding term in the rate equation is neglected. The values of $\tau_{\text{0}}$ (= 0.55 ps) is fixed at k$\parallel$ ~ 0, given by $\tau_{\text{0}} = \tau_{\text{ph}}|C_{k\parallel=0}|^2$ where $\tau_{\text{ph}}$ ~ 0.26 ps determined from the cavity Q ~ 600 and $|C_{k\parallel=0}|^2$ is the photon fraction in the LPB at k$\parallel$ ~ 0 (Fig. 6.6(a)). Plotted in Fig. 6.11(b) are the values of $\tau_{\text{therm}}/\tau_{\text{0}}$ against the relative
excitation power. It is evident that the value of \( \tau_{\text{therm}} \) decreases rapidly at \( P/P_{\text{th}} \sim 1 \) and reaches 0.09\( \tau_0 \) at the highest excitation power. Similar TRPL measurements were also made on the GaN nanowire microcavity (sample 2) and the transient data were again analyzed by the coupled rate equations (1) and (2). In this case \( \tau_{\text{therm}} \) represents the overall scattering (relaxation) time from the reservoir to the condensate at \( k_{||} \sim 0 \), neglecting the non-linear time constants and polariton densities characterizing the intermediate polariton scattering processes. The variation of \( \tau_{\text{therm}}/\tau_0 \) for sample 2 is also shown in Fig. 6.11(b) and it is evident that \( \tau_{\text{therm}} \) tends to saturate at a value comparable to that of \( \tau_0 \) (0.6\( \tau_0 \)). The steep decrease of \( \tau_{\text{therm}}/\tau_0 \) observed for sample 1 has not been reported for any other microcavity polariton emitter. From the measured linewidths of polariton emission in sample 1, the coherence time \( \tau_{\text{coherence}} \) increases from 0.34 ps below threshold to a value of \( \sim 10 \) ps just above threshold. This is significantly larger than polariton lifetime (\( \tau_0 \)) of 0.55 ps at \( k_{||} \sim 0 \). In turn, \( \tau_0 \) is larger than \( \tau_{\text{relax}} \) by an order of magnitude above
threshold. We therefore believe that the incorporation of graded Al(Ga)N in the nanowire, forming a potential trap and enabling evaporative cooling, assists the dynamic condensation process and singularly creates an equilibrium phase transition in the Bose condensate at 300K. In contrast, in the GaN nanowire sample, polariton-phonon scattering is not sufficient enough to cool the polaritons to form a BEC at 300K.

6.2.5 First-order Spatial Coherence

Additional characteristics of a BEC include long-range coherence and spontaneous symmetry breaking of the polarization of the polaritons. We have measured the first-order correlation of the polariton luminescence with a Michelson interferometer. The cw excitation source is a 325 nm HeCd laser and the incident power is varied from 0.3 mW to 1.5 mW. The experimental arrangement, schematically shown in Chapter 3, creates a double image of the condensate with a spatial resolution of 0.3 μm. Figure 6.12 shows the measured contrast \((I_{\text{max}} - I_{\text{min}})/(I_{\text{max}} + I_{\text{min}})\) of the fringes as a function of displacement between the images in the two arms. The measurement therefore provides direct evidence of coherence in the first-order correlation of the polariton emission and an estimate of the spatial coherence length of the condensate along the length (or c-axis) of the nanowire. Below threshold, the contrast remains within the noise margin for any overlap between the images, indicating that there is negligible coherence across the condensate. Above threshold, a maximum contrast of 21% is observed at \(P = 1.1\) mW for complete overlap between the two images and the contrast decreases as the images are moved apart. The linewidth (FWHM) of the contrast profile is ~ 1.2 μm, which is a measure of the size of the condensate formed in the trap above threshold and it is approximately the same as the length of the GaN region in the nanowire.
6.2.6 Polarization

The polarization of the polariton emission was also measured as a function of incident power and the output was linearly polarized below and above threshold, unlike the case of bulk/quantum well planar microcavities, where spontaneous symmetry breaking is responsible for a linearly polarized emission above threshold [123]. The measured polarization is shown in Fig. 6.13 and the results can be understood in the context of the geometry of our sample shown in the inset to Fig. 6.13. The nanowire is embedded in-plane in the microcavity. The $\text{X}_\text{A}$ exciton-polariton emission is collected in the direction vertical to the $c$-axis of the nanowire. The emission must therefore be linearly polarized in a direction perpendicular to the $c$-axis and the direction of emission, independent of the excitation intensity. Polariton superfluidity and associated quantized vortices observed in exciton-polariton condensates [124,125] and dilute
atomic gas BECs [126,127] could not be investigated because of the intrinsically weak emission from a 1 μm length of GaN in the nanowire microcavity which limits any imaging of the interference pattern and subsequent extraction of phase information.

6.2.7 Conclusion

In conclusion, a spatial polariton trap is created along the length of a Al(Ga)N nanowire and strong coupling effects have been investigated in the polariton emission of a planar nanowire-dielectric microcavity sample. It is evident that the trap assists in evaporative cooling of the optically excited exciton-polaritons as they are transported along the nanowire, to form an equilibrium BEC in the lowest bandgap GaN region of the nanowire at 300K.

Fig. 6.13 Polar plot indicating the polariton emission intensities measured as a function of angle of the polarizer for 3 different pump powers. Inset shows the measurement geometry.
Chapter 7
Conclusions and Suggestions for Future Work

7.1 Summary of Present Work

This thesis has presented several polariton light sources based on III-V semiconducting materials with emphasis on increasing the operating temperature for observing strong coupling effects and polariton condensation. While most studies on microcavity exciton-polaritons are carried out at cryogenic temperatures, the work described in this thesis is mostly at room temperature, moving towards practical applications opposed to a study of physical phenomena. Also, an electrically injected polariton laser was realized, which had so far remained elusive. Key results and conclusions are highlighted in the following sections.

7.1.1 Room temperature polariton lasing with a single GaN nanowire

We developed a new structure — GaN nanowires — as the active media, and produced a room-temperature polariton laser. Remarkably, the nanowires are free of extended defects, and have negligibly small with an internal quantum efficiency of ~ 50-60%. We enclosed a single nanowire in a dielectric microcavity. The characteristic polariton dispersions were measured by angle-resolved photoluminescence and yielded a Rabi splitting $\Omega$ of 48 meV. The relatively large value of $\Omega$ for a single nanowire is due to both the large oscillator strength of excitons resulting from a small internal electric field and a modified cavity field that concentrates within the
nanowire. The LP density at the threshold was found to be $\sim 2 \times 10^{16} \text{ cm}^{-3}$ which is two orders of magnitude lower than any previously reported GaN polariton lasers, and three orders of magnitude less than the exciton Mott density. A second threshold, which we believe corresponds to photon lasing with an inverted carrier population, was observed at 2700 times the polariton lasing threshold.

### 7.1.2 Dynamic polariton condensation in a single nanowire-microcavity

We have experimentally investigated the dynamic polariton condensation behavior in a single GaN nanowire strongly coupled to a dielectric microcavity under non-resonant optical excitation. Both time-integrated and time-resolved polariton luminescence measurements have been made in the temperature range of 25 to 100K (corresponding to exciton-cavity photon detuning of -3.0 to +4.2 meV). Polariton lasing is observed in the entire temperature range, with the lowest threshold energy of 57 nJ/cm² measured at 50K ($\delta = -1.3$ meV). All the measurements indicate that at the lower temperatures, the degenerate polariton condensate is not in thermal equilibrium with the phonon bath. At 85 and 100K ($\delta = +2.3$ and 4.2 meV) the system attains a state close to thermal equilibrium via dynamic Bose condensation. The best results are obtained at $T_{\text{lat}} = 85\text{K}$, for which $T_{\text{LP}} = 88.8\text{K}$, and this is the highest temperature recorded for an equilibrium phase transition in exciton-polariton condensates.

We have also investigated strong coupling effects in a single nanowire-dielectric microcavity at room temperature. The single ZnO nanowire is embedded in a dielectric and surrounded by distributed Bragg reflectors (DBRs) in the top and bottom. The polariton dispersion characteristics at room temperature have been calculated and measured by angle resolved photoluminescence from which a Rabi splitting of 103 meV is obtained. Non-linear emission
characteristics are observed at room temperature with a distinct threshold at a very low optical excitation density of 1.63 μJ/cm², accompanied by linewidth narrowing. The measured population distribution in momentum space and the polariton relaxation and recombination times confirm the absence of a relaxation bottleneck and the attainment of quantum degeneracy at $k_|| \sim 0$.

7.1.3 Room temperature dynamic Bose-Einstein condensation in a graded Al(Ga)N nanowire-microcavity with a potential trap

A spatial potential trap is formed in a 6.0 μm Al(Ga)N nanowire by varying the Al composition along its length during epitaxial growth. The polariton emission characteristics of a dielectric microcavity with the single nanowire embedded in-plane has been studied at room temperature. Excitation is provided at the Al(Ga)N end of the nanowire and polariton emission is observed from the lowest bandgap GaN region of the nanowire. Comparison of the results with those measured in an identical microcavity with an uniform GaN nanowire and having an identical exciton-photon detuning suggests evaporative cooling of the polaritons as they are transported across the trap in the Al(Ga)N nanowire. Measurement of the spectral characteristics of the polariton emission, their momentum distribution, first-order spatial coherence and time-resolved measurements of polariton cooling provide strong evidence of the formation of an equilibrium Bose-Einstein condensate, a unique state of matter in solid state systems, in the GaN region of the nanowire, at room temperature. An equilibrium condensate is not formed in the GaN nanowire dielectric microcavity without the spatial potential trap.

7.1.5 Exciton-polariton lasing in a modulation doped MQW exciton-polariton microcavity diode with a magnetic field

Polariton relaxation can be enhanced by deploying polariton-electron scattering, which is more efficient in cooling the LPs compared to polariton-phonon scattering and should facilitate
the elimination or reduction of the relaxation bottleneck. In an optically excited system, it has been demonstrated that photo-excitation of a secondary quantum well produces a two-dimensional electron gas, which is transformed to the primary quantum well and interacts with LPs contained therein. In an electrically injected microcavity device, modulation doping can be achieved more easily by introducing a sheet of (Si) dopants in the barrier regions at a suitable distance from the quantum wells. However, even with an enhancement of the scattering rate, the system may transition to the weak coupling regime before polariton lasing can be observed. It is known that in semiconductors a magnetic field ($B$) shrinks the exciton wave function and enhances the electron-hole overlap, resulting in an increase in oscillator strength $f$. In a system with strong coupling, since, the Rabi splitting also increases with field. This behavior has been demonstrated in magneto-photoluminescence and magneto-absorption measurements (18-20). The reduction in Bohr radius helps to increase the exciton-polariton saturation density. Therefore, in the presence of a magnetic field and polariton electron scattering, it should be possible to observe coherent emission in the appropriate temperature range. In this work we have investigated strong coupling effects in a quantum well microcavity diode with modulation doping of the wells in the presence of an applied magnetic field of 7 Tesla at $T = 30$ K. Polariton lasing from the microcavity diode is observed under these conditions. The polariton dispersion characteristics were measured by angle-resolved electroluminescence (EL). The Rabi splitting increases from 5.1 meV at 0 Tesla to 6.98 meV at 7 Tesla. The measured light-current characteristics exhibit a distinct non-linearity with a threshold at a current density of $\sim$12 A/cm$^2$, which corresponds to a polariton density of $\sim$5x10$^{10}$ cm$^{-2}$. Measurement of LP occupation in $k_\parallel$-space, first-order spatial coherence and output polarization characteristics confirm polariton condensation and lasing. Finally, a second threshold, corresponding to photon lasing with
population inversion, is observed from the same diode at a current density ~3 orders of magnitude higher than the polariton lasing threshold.

### 7.2 Suggestions for Future Work

#### 7.2.1 1.3 and 1.55 µm Electrically Injected Quantum Dot and Quantum Dash Microcavity Polariton Emitters

A large oscillator strength $f$ can be attained with a quantum dot enclosed in a high-Q cavity. A single quantum dot will provide the narrow linewidth required and the g-factor and Rabi splitting $\Omega$ will be enhanced with a large $f (g \propto \sqrt{f})$ and a small mode volume. Low density InAs quantum dots emitting at 1.3µm can be grown on GaAs and nanopillar microcavities with top and bottom DBRs can be designed, grown and fabricated. Electrical contact to the top p-contact will be made after planarization. The device is schematically shown in Fig. 7.1. Since the value of $g$ and $\Omega$ in this system is not expected to be very large, operation of this device will be restricted to low temperatures. However, one can strive to measure device performance in the temperature range of 30-77K.

![Schematic](image)

**Fig. 7.1** Schematic of an electrically injected polariton laser with a single InAs QD operating at 1.3 µm.
Self-organized quantum dashes can be grown on InP-based heterostructures (Fig. 7.2) and these nanostructures exhibit peak emission in the 1.5-1.65 μm range [128]. We will fabricate microcavity device heterostructures as shown in Fig. 8.2 such that a single quantum dash forms the radiator. These devices have an internal quantum efficiency ~ 100%, are ultrafast (with a theoretical frequency ~ 10-100 GHz) and can operate in the fiber-optic wavelengths of 1.3 and 1.55 μm. Also, 0D micropillars are useful for quantum cryptographic applications.

7.2.2 1.3μm Single Nanowire-Quantum Dot Microcavity Device

The approach to be used is schematically shown in Fig. 7.3(a), where a p-i-n GaAs nanowire [129] with a InAs quantum dot is buried in a dielectric microcavity. The nanowires can be grown on (111)B GaAs substrates and dispersed selectively on the dielectric half-cavity. P- and n- contacts, as shown in Fig. 7.3(b) will be made outside the microcavity. Luminescence measurements can be made to optimize growth on the dot-in-nanowires. I-V characteristics measured for similar single GaN p-i-n nanowires are shown in Fig. 7.3(c).

The single dot device described here and in the last section can also be used to study fundamental quantum strong coupling phenomena at the single QD level. One can characterize the enhancement in the coupling constant, the polariton relaxation rates and the thermal
properties of the degenerate Bose-Einstein condensate. In addition to the electrical injection mode of excitation described here, one can also perform strong coupling experiments on the same microcavities with optical excitation.

7.2.3 Room Temperature Nitride-Based Nanowire Microcavity Device

As stated earlier in Chapter 2, Ga(in)N nanowires present a unique opportunity in this field of research. They can be grown defect free on (001) silicon, can be doped n- and p-type, can form heterojunctions, have very small density of surface defects and small polarization field and the emission wavelength can be tuned over a wide (visible) range. Most importantly, the strong coupling constant $g$ is very large in these materials, promising the observation of strong coupling effects and polariton lasing at room temperature. From practical application point of view, one can work with clusters such that a large light output can be obtained.

The device is schematically shown in Fig. 7.4. A cluster of (GaN )dot-in- (GaAlN) nanowires (DNW) will be grown on (100) Si. The nanowires should be doped to form p-n junctions. Planarization of the nanowires should be done with parylene deposition, followed by mesa etching, top DBR (SiO$_2$/SiN$_X$) deposition and removal of the substrate. This will be
followed by the deposition of a semitransparent Au film, n-ITO and another SiO$_2$/SiN$_x$ deposition. Since the exciton binding energy and $\Omega$ are large in GaN, this device is expected to operate at room temperature.
APPENDICES
APPENDIX A
MATLAB CODES

A.1 2x2 Coupled Harmonic Oscillator Hamiltonian

```matlab
% Parameters
% -> Physical Constants
c = 3*10^10;
e = 1.6*10^-19;
ephys0 = 8.858*10^-12;
m0 = 9.1*10^-31;
hbar = 6.582*10^-16;
% -> Energies
Ephys0 = 3.399;  % Exciton Energy in eV
gamma0 = 0.005;  % Exciton linewidth in eV
gammacav = 1.41/(600/.37);  % Photon linewidth from Q = 600
% -> Index of Refraction
nc = 1.75904;
nl = nc;
nh = 3.052;
% Calculations
% -> Calculate the Cavity Parameters
lambdac = 390*10^-7;
mcav = Ephphys0/(c^2/nc^2);
Lcav = 251*10^-7;
Leff = Lcav + lambdac/4*nh/(nh-nl);
% -> Calculate the interaction potentials
V1 = 0.048;
theta = -30*pi/180:pi/360:30*pi/180;
% Calculate Momentum at this angle
k = 2*pi/lambdac*theta;
% Cavity mode Energy at this angle
Ephys = Ephphys0 + hbar^2*k.^2/(2*mcav);
Elphys = 0.5*(Ephys+Ephys0+i*(gammacav+gamma0)-sqrt(V1^2+(Ephys0-Ephys+i*(gammacav-gamma0)).^2));
Elphys = 0.5*(Ephys+Ephys0-sqrt(V1^2+(Ephys0-Ephys).^2));
Euph = 0.5*(Ephys+Ephys0+i*(gammacav+gamma0)+sqrt(V1^2+(Ephys0-Ephys+i*(gammacav-gamma0)).^2));
Euph = 0.5*(Ephys+Ephys0+sqrt(V1^2+(Ephys0-Ephys).^2));
X_k = 0.5*(1+(Ephys0-Ephys)/sqrt((Ephys0-Ephys).^2+V1^2));
C_k = 0.5*(1-(Ephys0-Ephys)/sqrt((Ephys0-Ephys).^2+V1^2));
Eexp = Ephys0*theta/theta;
%plot(theta*180/pi, sqrt(Elphys.* conj(Elphys)),'b');
plot(k, sqrt(Elphys.* conj(Elphys)),'b');
hold on;
%plot(theta*180/pi, sqrt(Euph.* conj(Euph)),'r');
plot(k, sqrt(Euph.* conj(Euph)),'r');
hold on;
%LPB_PL_data = [3.442, 3.44, 3.446, 3.448, 3.45, 3.454, 3.456, 3.46];
```
%UPB_PL_data = [3.49,3.496,3.498,3.506,3.518,3.524,3.542];
%LPB_PL_angle = [0,3,6,9,12,15,18,21];
%UPB_PL_angle = [0,6,9,12,15,18,21];
%plot (LPB_PL_angle,LPB_PL_data,'bo','linewidth',2);
%plot (UPB_PL_angle,UPB_PL_data,'ro','linewidth',2);
hold on;
plot (k,Eph,'g');
plot (k,Eexp,'m');
plot (k,X_k,'y');
plot (k,C_k,'s');

A.2 3x3 Coupled Harmonic Oscillator Hamiltonian For Modulation-Doped Microcavity

%Parameters
%- Physical Constants
  c = 3*10^10;
  e = 1.6*10^-19;
  ephs0 = 8.858*10^-12;
  m0 = 9.1*10^-31;
  hbar = 6.582*10^-16;
%- Energies
  Eph0 = 1.3744;
  Eex0 = 1.3803;
  Eex1 = 1.378;
%- Index of Refraction
  nc = 3.43;
  nl = nc;
  nh = 3.052;
%- Calculations
%- Calculate the Cavity Parameters
  lambdac = 1240/Eph0*10^-7;
  mcav = Eph0/(c^2/nc^2);
  Lcav = 251*10^-7;
  Leff = Lcav + lambdac/4*nh/(nh-nl);
%- Calculate the interaction potentials
  V1 = 0.0057;
  V2 = 0.0015;
  \( g_0 = e\cdot hbar/nc\cdot sqrt(c\cdot fosc/2/m0/ephs0/Leff); \)
  theta = 0:pi/360:30*pi/180;
  k = 2*pi/lambdac.*theta;
  Eph = Eph0 + hbar^2.*k.*k/(2*mcav);
%- Solving the Hamiltonian
  \( H = [Eph, V1, V2; V1, Eex0, 0; V2, 0, Eex1]; \)
[P, SolvedH] = eig(H);

% Calculate the dispersion profile and plot

measure = size(theta);

plot(theta*180/pi, eval(Eph), 'r-', 'linewidth', 2);
hold on
plot(theta*180/pi, Eex0*ones(1, measure(2)), 'k--', 'linewidth', 2);
plot(theta*180/pi, Eex1*ones(1, measure(2)), 'k--', 'linewidth', 2);

colors = 'bgcy';

for j = 1:3;
    Pol = eval(SolvedH(j, j));
    plot(theta*180/pi, sqrt(Pol.*conj(Pol)), colors(j), 'linewidth', 2);
    hold on
end

xlabel('Angle (Degrees)');
ylabel('Energy (eV)');
title('Dispersion Relation of GaN Microcavity');
hold off

A.3 Critical Density for KT transition in a 2D Microcavity

%% Find the Densities associated with the KT transition
%% Initialize SI;
CriticalDensity = zeros(1, 4);
%% 100K
T = 100;  % K
Detuning = 4.2;  % meV
MicrocavityDispersion;
% CriticalDensity(1) = kB*T/pi/(hbar^2)*2*mLP(0).^2;
% LT = sqrt(2*pi*hbar^2/(mLP(0)*kB*T));
% CriticalDensity(1) = 2/LT^2*log(NW_D/LT);
CriticalDensity(1) = 1./(2*pi).*quadgk(@(k) k./(exp((E_LPB(k)-E_LPB(0))/(kB*T))-1), 2*pi/MC_D, inf);
%% 85K
T = 85;  % K
Detuning = 2.3;  % meV
MicrocavityDispersion;
% CriticalDensity(2) = kB*T/pi/(hbar^2)*2*mLP(0).^2;
% LT = sqrt(2*pi*hbar^2/(mLP(0)*kB*T));
% CriticalDensity(2) = 2/LT^2*log(NW_D/LT);
CriticalDensity(2) = 1./(2*pi).*quadgk(@(k) k./(exp((E_LPB(k)-E_LPB(0))/(kB*T))-1), 2*pi/MC_D, inf);
%% 70K
\[ T = 70; \quad \%K \]
\[ \text{Detuning} = 1; \quad \%\text{meV} \]
\[ \text{MicrocavityDispersion}; \]
\[
\begin{align*}
\text{CriticalDensity}(3) &= \frac{kB \cdot T}{\pi \cdot (hbar^2) \cdot 2 \cdot mLP(0)^2}; \\
\text{LT} &= \sqrt{\frac{2 \cdot \pi \cdot hbar^2}{(mLP(0) \cdot kB \cdot T)}}; \\
\text{CriticalDensity}(3) &= \frac{2}{LT^2 \cdot \log(NW_D/LT)}; \\
\text{CriticalDensity}(3) &= \frac{1}{(2 \cdot pi) \cdot \quad \text{quadgk}(\theta(k) \quad k/(exp((E_LPB(k)-E_LPB(0))/(kB \cdot T))-1), \quad 2 \cdot pi/MC_D, inf);}
\end{align*}
\]
\[ \% 25K \]
\[ T = 25; \quad \%K \]
\[ \text{Detuning} = -3; \quad \%\text{meV} \]
\[ \text{MicrocavityDispersion}; \]
\[
\begin{align*}
\text{CriticalDensity}(4) &= \frac{kB \cdot T}{\pi \cdot (hbar^2) \cdot 2 \cdot mLP(0)^2}; \\
\text{LT} &= \sqrt{\frac{2 \cdot \pi \cdot hbar^2}{(mLP(0) \cdot kB \cdot T)}}; \\
\text{CriticalDensity}(4) &= \frac{2}{LT^2 \cdot \log(NW_D/LT)}; \\
\text{CriticalDensity}(4) &= \frac{1}{(2 \cdot pi) \cdot \quad \text{quadgk}(\theta(k) \quad k/(exp((E_LPB(k)-E_LPB(0))/(kB \cdot T))-1), \quad 2 \cdot pi/MC_D, inf);}
\end{align*}
\]

**A.4 Scattering Rates in Presence of Magnetic Field**

\[
\% \text{ Plot the Polariton and Phonon Matrix Elements as a function of } B
\]
\[
\% \text{ The Magnetic field parameters as a function of } B
\]
\[ \text{Mag}_L = @(Mag_B) \sqrt{\frac{hbar}{(Mag_B)}}; \quad \text{Magnetic length in m} \]
\[ \omega_e = @(Mag_B) \frac{Mag_B}{me}; \quad \text{Cyclotron Frequencies} \]
\[ \omega_h = @(Mag_B) \frac{Mag_B}{mh}; \]
\[ le = 0; \]
\[ lh = 0; \]
\[ Bappl = [0,2:10]; \quad \%\text{Tesla} \]
\[ W_{\text{Phon Mag}} = \text{zeros}(\text{length}(Bappl),\text{Res}); \]
\[ W_{\text{Pol Mag}} = \text{zeros}(\text{length}(Bappl),\text{Res}); \]
\[
\begin{align*}
\% \text{ First, find the energy of the ground state of the well.}
\end{align*}
\]
\[
\begin{align*}
\text{k_E} &= @(E,m,V) \sqrt{2 \cdot m^* \cdot (E+V)/hbar^2}; \quad \%\text{Wave vector in well} \\
\text{K_E} &= @(E,m) \sqrt{2 \cdot m^* \cdot (-E)/hbar^2}; \quad \%\text{Wave vector outside}
\end{align*}
\]
\[ \text{BB} = @(m) \frac{2 \cdot m^* \cdot dEc}{hbar^2 \cdot QW_L^2}; \]
\[ \text{Boundstate} = @(y,m) \tan(y/2) - \sqrt{\text{BB}(m)/y^2 - 1}; \quad \%\text{Root gives energy of bound state} \\
\text{y_e} &= \text{fzero}(@(y) \text{Boundstate}(y,me), [pi/2, pi*0.99]); \quad \%\text{Ground State for electron} \\
\text{y_h} &= \text{fzero}(@(y) \text{Boundstate}(y,mh), [pi/2, pi*0.99]); \quad \%\text{Ground State for hole} \\
\text{E_e} &= y_e^2/QW_L^2 \cdot hbar^2/(2 \cdot me) \cdot dEc; \quad \%\text{Electron Energy} \\
\text{E_h} &= y_h^2/QW_L^2 \cdot hbar^2/(2 \cdot mh) \cdot dEv; \quad \%\text{Hole energy}
\]
\[
\begin{align*}
\% \text{ The resulting electron and hole wavefunctions.}
\end{align*}
\]
\% 1 | 2 | 3 - 2 is in the well, 1 and 3 are in the walls

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\[ A_{\psi} = @(E,m,V) \frac{1}{\sqrt{QW_L/2*(1+\sin(k_E(E,m,V).*QW_L)/(k_E(E,m,V).*QW_L))}}; \]

\[ B_{\psi} = @(E,m,V) A_{\psi}(E,m,V).*\cos(k_E(E,m,V).*QW_L/2).*\exp(K_E(E,m)*QW_L/2); \]

\[ \Psi_1 = @(x,E,m,V) B_{\psi}(E,m,V).*\exp(K_E(E,m)*x); \]

\[ \Psi_2 = @(x,E,m,V) A_{\psi}(E,m,V).*\cos(k_E(E,m,V)*x); \]

\[ \Psi_3 = @(x,E,m,V) B_{\psi}(E,m,V).*\exp(-K_E(E,m)*x); \]

% Piecewise solution of the three regions.
\[ \Psi = @(x,E,m,V) B_{\psi}(E,m,V).*\exp(K_E(E,m)*x).*(x<(-QW_L/2))+A_{\psi}(E,m,V).*\cos(k_E(E,m,V)*x).*(x>=(-QW_L/2)).*(x<=(QW_L/2))+B_{\psi}(E,m,V).*\exp(-K_E(E,m)*x).*(x>(QW_L/2)); \]

\[ \Psi_e = @(x) \Psi(x,E_e,me,dEc); \]

\[ \Psi_h = @(x) \Psi(x,E_h,mh,dEv); \]

%% Check out our wavefunctions
figure; fplot(@(x) \Psi_e(x).^2, [-10*QW_L 10*QW_L]);
figure; fplot(@(x) \Psi_h(x).^2, [-10*QW_L 10*QW_L]);

%% Test normalization: \quadgk(@(x) \Psi_e(x).^2,-1E1*QW_L,1E1*QW_L)
\quadgk(@(x) \Psi_h(x).^2,-1E1*QW_L,1E1*QW_L)

%% Now, solve for the reduced Bohr radius numerically.

B_cnt = 1; % Will be used later to iterate the magnetic field
Mag_B = Bappl(B_cnt); % Tesla

% The equation to be solved. This contains a nasty triple integral
% (see inplane)
\[ \text{RadiusEqn} = @(ab_p,B) hbar^2/(8*mexc)*(1-6*(ab_p/(2*Mag_L(B))).^4)-\]
qe.^2./(ephs0.*eps_r).*\quadgk(@(r) \text{InPlane}(r,QW_L,@(ze) \Psi_e(ze), @(zh) \Psi_h(zh)).*r.*(1-r/ab_p).*\exp(-2*r/ab_p),0,inf);

% We can make a change of variable (see numerical methods in C) to smooth
% the singularity at r=0: I'm still trying to get this to work right..
% \text{RadiusEqn} = @(ab_p,B) hbar^2/(8*mexc)*(1-6*(ab_p/(2*Mag_L(B))).^4)-\]
qe.^2./(ephs0.*eps_r).*\triplequad(@(r,ze,zh) \text{Psi}(ze,E_e,me).^2.*\text{Psi}(zh,E_h,mh).^2)./sqrt((r.^2+(ze-zh).^2).*r.*(1-(r.^2)/ab_p).*\exp(-2*(r.^2)/ab_p),-1E5,1E5,-1E5,1E5,0,1E10);

% Loop for different magnetic fields will begin here
% Use a root finding method to actually find what value of ab is a root
% Still working on this one, so it's commented out for now:
% ab_p = fzero(@(ab_p) \text{RadiusEqn}(ab_p,Mag_B), [ab/2 ab]);
The resultant binding energy.

% Integral not converging
% Eb_p = \( -\frac{3}{16} \hbar^2 \frac{a_p^2}{m_{exc}/M_{Mag}(B)^4} - \frac{\hbar^2}{2m_{exc}a_p^2} + \frac{4}{a_p^2} \frac{\epsilon_e^2}{\epsilon_{phs0} \epsilon_r} \cdot \text{quadgk}(r) \cdot \text{InPlane}(r, \Psi_e(ze), \Psi_h(zh)) \cdot \exp(-2r/a_p) \cdot 0, \infty) - \hbar^2 (\omega_e(B) (le + 1/2) + \omega_h(B) (lh + 1/2)) \);

% The wavefunction overlap and the oscillator strength.
% Overlap = \( \text{quadl}(x) \cdot \Psi_e(x) \cdot \Psi_h(x), -1E5, 1E5 \);

V1 = \( V0/(a_p/a) \)^2;

% High field limit
Eb_B = \( \frac{\epsilon_e}{4\pi \epsilon_{phs0} \epsilon_r} \sqrt{\pi} (1 - \frac{1}{2} \frac{2/3}{\sqrt{\pi}} - \frac{5/2}{\pi^2} \frac{1}{\sqrt{\pi}} \cdot QW_L/M_{Mag}(B)) \);

ab_B = \( \frac{\epsilon_e}{4\pi Eb_B(\epsilon_{phs0} \epsilon_r)} \);

V_B = \( V0/(ab_B/(ab)) \)^2;

for B_cnt = 1:length(Bappl)
    Mag_B = Bappl(B_cnt);
    Eb = Eb_B(Mag_B);
    V1 = V_B(Mag_B);
    if (Mag_B == 0)
        V1 = V0;
        Eb = Eb0;
    end
end

%% Last, get the new dispersion relations
%% MicrocavityDispersion;
%% Phonon Scattering rate from k to k0
qll = k;
Ephn = real(Elp-Elp(1));
qz = sqrt((Ephn/(\hbar*cs)).^2-(qll).^2);
Ie = (1+((mh*qll*ab)/(2*mexc)).^2).*(-3/2);
Ih = (1+((me*qll*ab)/(2*mexc)).^2).*(-3/2);
Gq = De*Ie-Dh*Ih;

%% Illustrative purposes only- the Fermi's golden rule to ground state.
%% Actual rate equations will be formulated later
W_Phon_Mag(B_cnt,:) = \( 2\pi/\hbar \cdot \text{abs}(\Xi(1) \cdot \sqrt{Ephn/(2 \cdot NW_S \cdot QW_L \cdot \text{density} \cdot cs^2)} \cdot Gq \) \^ 2;

%plot(k,W_Phon_Mag(k));

%% Polariton Scattering rate from k to k0
%% Fermi's golden rule to ground state.
W_Pol_Mag(B_cnt,:) = \( 2\pi/\hbar \cdot \text{abs}(6 \cdot ab_B(Mag_B) \cdot (2 \cdot NW_S \cdot Eb \cdot \Xi(1)) \)^ 2;
%plot(k,W_Pol_Mag(k))
end
% Loop ends here, will now have the rates from k->0 for varied B-field.
%% Plotting
figure;mesh(k,Bappl,W_Phon_Mag)
ylabel('Magnetic Field (T)');xlabel('k_\perp');title('Fermi Golden Rule for Scattering from state k_\perp to the ground state by phonons')

figure;mesh(k,Bappr,W_Pol_Mag)
ylabel('Magnetic Field (T)');xlabel('k_\perp');title('Fermi Golden Rule for Scattering from state k_\perp to the ground state by polaritons')
APPENDIX B

FDTD SIMULATION IN A GaN-NANOWIRE MICROCAVITY

B.1 The parameters

$n_{\text{SiO}_2}=1.5$, $n_{\text{TiO}_2}=3.66$, $n_{\text{GaN}}=2.5$. Designed wavelength $= 365$ nm, cavity thickness $= 90$ nm (as opposed to be $121$ nm, tailored to compensate the higher index of GaN nanowire), nanowire radius $= 30$ nm, nanowire length $= 400$ nm. 7.5 pairs of top and bottom DBRs respectively and no substrate (in the air).

B.2 The NW embedded cavity resonances

The parameters I’m using are slightly different from my previous simulations in which I was setting $n_{\text{TiO}_2}=2.2$ which is not quite true according to the data we’ve got online. I also placed more monitors at different locations to make sure no resonance is missing. The mode I found here is no longer guessed from numerous random modes in the leaky region. This simulation only assumed symmetry in the cavity growth direction but didn’t restrain the symmetric in the horizontal directions. This time we get two polarization modes confined in the nanowire. X polarized mode corresponds to E field perpendicular to the nanowire. Y polarized mode corresponds to the E field parallel to the nanowire direction. Although the Q factor of the X polarized mode (400) is two times of the Y polarized mode (200), I tend to think that the Y polarized mode is more likely to be the mode we saw, because the energy of it is closed to the exciton energy. However, due to the refractive index setup and other issues the simulation is different from the real sample whose refractive indexes are not sure. It’s hard to say how much we’ve shifted from the real sample. The best way is to look at the polarization data to see the
relative orientation of the nanowire to the ‘X axis’ in the manuscript. The nanowire orientation is the Y axis. Z axis is the DBR growing direction.

The following figures are for the X polarized mode. One can see the discontinuity on the NW boundary.

Assuming that the mode of interest is the Y polarized mode. This mode has higher energy concentration than the previous index setup (for which I got 1.9% energy concentration) as one can see from the mode profiles below.

The energy concentration is 7.7% (compared to the previous result 1.9%) which is actually already much higher than what we would originally think if the existence of the nanowire is just
treated as a perturbation. The normalized E field integral in this region is 3.3e-5 Vm\(^{1/2}\) for 3.638 eV. In comparison the GaN bulk cavity has an energy concentration of 20.6\% with the normalized E field integral being 4.4e-5 Vm\(^{1/2}\) for 3.465 eV. Therefore the normalized E field integration ratio will be around \(\frac{4.4}{3.3} \times \left(\frac{3.638}{3.465}\right)^{1/2} = 1.4\). If the beam splitting effect is the same for both situations, the oscillator strength of the nanowire will be 1.4 times of the bulk GaN, assuming that the exciton wave function have uniform probability distribution.

The following is the cavity structure and the field distribution of the GaN bulk cavity in the reference *Phys. Rev. B* 2006, 73, 033315. The refractive indices I have been using for this are listed as \(n_{\text{SiO}_2} = 1.5\); \(n_{\text{Si}_3\text{N}_4} = 2.1\); \(n_{\text{GaN}} = 2.5\); \(n_{\text{InN}} = 2.9\); \(n_{\text{AlN}} = 2.2\). \(n_{\text{botdbrlow}} = 0.85*n_{\text{AlN}} + 0.15*n_{\text{InN}}\); \(n_{\text{botdbrhigh}} = 0.2*n_{\text{AlN}} + 0.8*n_{\text{GaN}}\); \(n_{\text{topdbrlow}} = n_{\text{SiO}_2}\); \(n_{\text{topdbrhigh}} = n_{\text{Si}_3\text{N}_4}\).

![Field distribution of GaN bulk cavity](image)

**B.3 Comparison with empty cavity**

The structure of the 90nm width cavity (remove the nanowire) and its reflection spectrum are shown below. The Q factor according to the reflectivity is 1800. The nanowire shifts the resonance to 340 nm and degrades the Q factor to ~200.
APPENDIX C

CALCULATION OF POLARITON DIFFUSION LENGTH IN TRAP

C.1 Evaluation of average polariton mass

\[
\frac{1}{m} = \frac{|X|^2}{m_{exc}} + \frac{|C|^2}{m_{cav}}, \quad \text{since } m_{cav} < m_{exc} \quad m \geq m_{cav}, \quad \text{where} \quad \frac{1}{|C|^2} = \frac{1}{n} \sum_{i=1}^{n} \frac{1}{|C_i|^2} \text{ is the inverse of the}
\]

photon fraction averaged along the trap and the cavity mass \( m_{cav} = \frac{E_{cav}(k_\parallel = 0)}{c^2 / n_c} \), where \( E_{cav} \)

(k_\parallel = 0) is the cavity energy at zero in-plane vector and n_c is the refractive index of the cavity material. Upon calculation \( m = 2.53 \times 10^{-5} m_0 \) where \( m_0 \) is the free electron mass.

C.2 Evaluation of average polariton lifetime in the ground state

\[
\tau_0 \approx \frac{\tau_{cav}}{|C|^2} = 0.44 \text{ ps}, \quad \text{where } \tau_{cav} \text{ is the cavity photon lifetime estimated from the cavity Q.}
\]

C.3 Diffusion length

In two dimensions, the diffusion length is given by \( l_D = \sqrt{D \tau_0} \), where the diffusion constant \( D \) is given by \( D = \frac{2k_B T}{m} \tau_{sc} \), where \( \tau_{sc} \) is the scattering time of polaritons with phonons and is equal to 20 ps in GaN at room temperature. The estimated diffusion length is equal to 61 \( \mu \text{m.} \)
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