Fundamental Properties of Metallic Nanolasers

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FUNDAMENTAL PROPERTIES OF METALLIC NANOLASERS

by

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A dissertation submitted in partial fulfillment of the requirements
for the degree of Doctor of Philosophy
in CREOL/The College of Optics & Photonics
at the University of Central Florida
Orlando, Florida

FallTerm
2018

Major Professor: Mercedeh Khajavikhan
ABSTRACT

The last two decades have witnessed tremendous advancements in the area of nanophotonics and plasmonics, which has helped propel the development of integrated photonic sources. Of central importance to such circuits is compact, scalable, low threshold, and efficient coherent sources that can be driven at high modulation frequencies. In this regard, metallic nanolasers offer a unique platform. Their introduction has enabled confinement of light at a subwavelength scale and the ultra-small size of the modes afforded by these structures allows for cavity enhancing effects that can help facilitate thresholdless lasing and large direct modulation bandwidths. In this report, I present my work on the study of the fundamental properties of metallic nanolasers. I start with a rate equation model to predict threshold behavior and the modulation response of metallic nanolasers. Next, I explain the second-order coherence ($g^2(\tau)$) measurement setup that was built, based on a modified Hanbury-Brown and Twiss experiment, to assess the intensity autocorrelation of various optically pumped metallic nanolasers. These studies concluded that metallic coaxial and disk-shaped nanolasers are capable of generating truly coherent radiation. Subsequently, design considerations are taken into account for electrically pumped coaxial nanolasers. This has led to the demonstration of electrically injected coaxial and disk-shaped nanolasers at cryogenic temperatures. Lastly, the appearance of collective behaviors in metallic nanolasers lattices is explored. Individually supporting modes that are highly vectorial by nature, when such cavities are fabricated in close proximity to one another, coupling through their overlapping fields results in the formation of a set of supermodes.
The tendency of the system to minimize the overall loss leads to each element of the lattice having a geometric dependent field distribution and helps promote single-mode lasing. We show both through simulations and experimentally that this effect can lead to the direct generation of vector vortices.
To my Grandpa Bill.
ACKNOWLEDGMENTS

First and foremost, I would like to thank my advisor Professor Mercedeh Khajavikhan for her enduring support throughout my PhD career. With the help of her thoughtful guidance, knowledge, and encouragement, I was able to challenge myself to grow and become a better scientist and person, making my time at CREOL truly rewarding. I am grateful for having the opportunity to have worked with her.

I would also like to give my thanks to Professors Demetrios Christodoulides and Patrick LiKamWa for their time, helpful advice, and stimulating discussions. Also I would like to express my gratitude to Professor Reza Abdolvand for his insightful commentary and the generous usage of his equipment. Additionally, I would also like to thank Professor David Hagan for the guidance and counseling he provided me, which proved especially constructive.

I would like to express my appreciation to my friends and colleagues: Hossein, Sara, Absar, Amin, Midya, Monica, Fedor, Ashutosh, Nick, Mohammad, Pawel, Jinhan, Enrique, Polo, Jason, Nathan, Asaf, and to many more who helped make my time at CREOL fun, rewarding, and an unforgettable time in my life.

Lastly I would like to thank my family and friends back home. First to my parents Kent and Linda who have provided me unwavering love and support my entire life. Also to the Ruhs family, my Aunt Cindy and Uncle Steve, and to my cousins Alyssa, Michael, Ryan, Dustin, and Sarah, who have been like a second family to me. To my high school Physics teacher, Mr. Wally Woelber, who showed me how fun science could be and perhaps unknowingly helped
set me down this career path. Additionally, my childhood friend Chet and his wife Cassie who are always a breath of fresh air and provide some much needed laughter. Finally, I would like to thank my two furry friends Frank and Sandy who always made it a joy to come home.
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CHAPTER 1: INTRODUCTION

In order to keep up with the ever-growing demand in big data processing required by large data centers, cloud computing, and Moore’s law, computer processors are drifting towards evermore parallelism [1]. However, it is predicted that electronic interconnects will soon not be able to keep up with the demand constraints for communication between these systems, mainly being cost, energy consumption, and crosstalk [2,3]. On the other hand, it is predicted that photonic integrated circuits could replace these electrical interconnects [4]. Such a circuit would require a compact, scalable, and efficient source. In this regards, nanolasers are an attractive prospect [5], they have been demonstrated to operate as thresholdless devices and are predicted to have large direct modulation responses.

The past two decades have seen remarkable progress in the use of metal in optics, ranging from metamaterials to the realization of subwavelength laser resonators. The formation of surface plasmon polaritons (SPPs) at the surface of the metal and dielectric interfaces allows for metallic cavities to be reduced below the diffraction limit in all three dimensions simultaneously. In turn, the associated small mode-volume can lead to cavity enhancing effects that help facilitate the lasing process. Provided that the cavity is judiciously designed so that there is a sparse set of modes within the gain bandwidth and also an increased spontaneous emission rate (Purcell effect), a close to unity spontaneous emission coupling factor ($\beta$) can be achieved. Given that $\beta$ has a value near unity, very low lasing thresholds can be realized, despite the relatively lower quality factors affiliated with the metallic cladding.
In this dissertation, an overview on the fundamental properties of metallic nanolasers is presented. Chapter 2 discusses the lasing process in metallic nanolaser disks and metallic coaxial nanolasers, as well as the type of modes supported in these cavities. A simplified rate equation is formulated in Chapter 3. The analysis provided here predicts the dependence of the quality factor, mode confinement, spontaneous emission coupling ratio, and the Purcell effect on the lasing threshold as well and the high direct modulation bandwidths expected from these small lasers. In Chapter 4, the second-order-coherence properties of metallic nanolasers are assessed using a modified Hanbury Brown and Twiss interferometer setup. I describe how to build the setup and then use it to measure the intensity autocorrelation to find the second-order coherence of metallic coaxial and disk nanolasers. Demonstrating that these devices are truly capable of generating coherent radiation. Chapter 5 details design consideration for electrically injected metallic nanolasers. It takes into account heat considerations as well as FEM simulation to identify eigenfrequencies that have high quality factors and large modal confinement. The fabrication procedure for electrically injected nanoalasers is detailed from start to finish. This chapter finishes with the demonstration of coaxial metallic nanolasers operating up to a temperatures of 140 K. Chapter 6 provides an overview of coupled metallic nanolaser lattices. A description of the coupling mechanism in a pair of nanolasers is first described. Next this technique is applied to an array of N-coupled nanolasers in a ring like chain. This leads to the generation of vector vortices. Lastly, vector vortices are demonstrated in metallic nanolaser arrays. The report finishes with concluding remarks in Chapter 7.
CHAPTER 2: METAL-BASED NANOLASERS

2.1. Introduction to metallic nanolasers

Until the past decade, most efforts to create ultra-small lasers concentrated on dielectric semiconductor cavities. In this regard, a myriad of approaches have been adopted to shrink the cavity size, such as vertical cavity surface emitting lasers (VCSELs), micropillars, microdisks, and photonic crystals [6–10]. However, these structures come with various drawbacks. Although VCSELs, micropillars, and microdisks are relatively miniature in stature, the modes in such devices are not always well confined to the laser cavity, making very dense integration of independently operating devices without mode coupling between lasers impractical. Photonic crystal lasers avoid this issue by confining the mode to the active region quite well, but do so at the expense of total cavity size, which can be quite large when factoring in all the Bragg periods that help form the resonator. The second obstacle to overcome is more fundamental, the diffraction limit $\lambda_0/2n_{eff}$ [11]. At best, essentially dielectric in construct, these sources can be scaled-down to subwavelength size in two dimensions. On the other hand, the plasmonic property of metals at optical frequencies allows for the support of highly confined subwavelength modes [12].

The first demonstration of a metallic cavity was an electrically injected active nanowire [13]. The gain medium was encased by a thin electrically isolating layer and was then coated with gold. This quickly led to the development of many other configurations [14–40]. However, the majority of these metal clad lasers support photonic modes that are similar to those observed in dielectric structures [13–25,40]. In these
devices, the metal helps confine light to the active region, but the mode itself has little overlap with the lossy metallic cladding, allowing these arrangements to support modes with moderate quality (Q) factors. Although these sources are slightly subwavelength in dimensions, as the size of the cavity is further reduced, the cut off frequencies for these modes are quickly approached. This in turn drastically reduces the associated Q factor and thus prevents lasing action from occurring.

For a resonator to support deeply subwavelength lasing, the cavity modes must strongly interact with the metallic cladding [26–30,32,33,35,37,38]. The absorption from the metal inevitably results in modes with lower Q factors. Naively, this would lead one to think it implausible to reach the lasing threshold. However, the ultra-small mode volumes offered by these resonators have important ramifications in nanolaser design, bringing about effects that help facilitate lasing. If the cavity is carefully designed so that the spontaneous emission rate is enhanced by the Purcell effect ($F$) [41] and there is only a single mode within the gain bandwidth, a close to unity spontaneous emission coupling ratio can be achieved [42]. In which case, the laser is thresholdless [43]. The first demonstration of a thresholdless laser was shown by Khajavikhan et al. [30] in 2012 utilizing a metallic coaxial cavity structure.

Coaxial cavities are commonly found structures at microwave frequencies and support the TEM mode that has no cut-off frequency [44]. Comprising of a central cylindrical piece of metal surrounded by a symmetric piece of dielectric, which is surrounded by a hollowed out piece of cylindrically shaped metal. It is then terminated by air to provide the impedance mismatch for reflection. A resonator is formed when the length is $l\lambda/2n_{eff}$,
where \( l \) is an integer number greater than zero. At microwave frequencies, where the metal essentially behaves as a perfect conductor, \( n_{\text{eff}} \) is equal to the refractive index of the sandwiched dielectric and the shortest cavity is \( \lambda/2n \). However, at optical wavelengths, the metal no longer acts as a perfect conductor. Surface plasmon polaritons form along the metallic and dielectric interfaces, allowing for the shrinking of the cavity in this direction. Due to this plasmonic property, a coaxial cavity can be reduced in all three dimensions simultaneously [30,45].

A metallic coaxial nanolaser is depicted in Fig. 1. At the center of the cavity is a metallic (silver) rod surrounded by a ring of semiconductor gain (six InGaAsP quantum wells) that is encapsulated by silver. The top and bottom sides are terminated by a SiO\(_2\) and air plug to help improve the mode confinement. The SiO\(_2\) is deposited to stop the formation of unwanted plasmonic modes at this interface. The lower air plug allows for optical pumping and for outcoupling of light. Furthermore, the silver acts as an effective heat sink, helping the devices to lase under CW pumping and at room temperatures.

![Figure 1 Schematic of a coaxial nanolaser cavity](image-url)
This coaxial cavity exhibits many of the promising properties associated with an ‘ideal’ nanolaser. It is capable of supporting ultra small confined modes, despite being subwavelength. This in turns leads to a large Purcell factor as well as a large portion of the spontaneous emission being coupled into the laser mode. Due to the small cavity size, high $F$, and lower Q-factor associated with these cavities, it is expected that these devices will have large direct modulation bandwidth, eliminating the need for stand-alone on chip modulators. Finally, the reduced radiative lifetime due to the Purcell factor lowers the total carriers able to recombine through non-radiative channels. As a result, coaxial nanolasers are expected to display a lower sensitivity to Auger and Surface recombination processes intrinsic to semiconductor lasers. These aspects will be further discussed in chapter 3 through rate equation formulations and chapter 4 for heat considerations.

The occurrence of several competing modes in the broad gain bandwidth provided by most semiconductors is, in general, undesirable. Below the lasing threshold, the spontaneous emission that facilitates the lasing process will be divided into numerous channels. When above the lasing threshold, multiple modes tend to oscillate as the device is pumped at high powers – a hindrance that can cause power fluctuations and instabilities.

To further elucidate on this approach for threhsoldless lasing, one needs to look more closely at the parameters governing the Purcell and $\beta$ factors. The Purcell factor represents the enhancement of the radiative decay rate of an emitter confined within a cavity environment to that of the bulk and is defined as,

$$ F = \frac{3}{4\pi^2} \left( \frac{\lambda_0}{n} \right)^3 \left( \frac{\min(Q,Q_h)}{V_m} \right) $$  \hfill (2.1)
where \((\lambda_0/n)\) is the wavelength in the material, \(Q_h\) is the quality factor associated with the homogeneous lineshape of the gain medium, and \(Q\) and \(V_m\) are the quality factor and mode volume of the cavity, respectively. Similarly, the spontaneous emission coupling factor \(\beta\) is defined as the ratio of spontaneous emission into the lasing mode to the total spontaneous emission generated by the gain medium, and can be written as

\[
\beta = \frac{F^{(1)}}{\sum_k F^{(k)}}
\]

(2.2)

where the lasing mode is indicated by \(k = 1\), and the summation is over all modes, including confined cavity modes as well as the free space radiation modes. The spontaneous emission coupling factor can be brought close to the theoretical limit of unity by increasing the Purcell factor associated with the lasing mode, reducing the number of cavity modes that coincide with the gain spectrum, and lastly by blocking the coupling to the radiation modes through the use of a metallic cladding (Fig. 2 a)). If \(\beta \approx 1\), almost all the spontaneous emission is funneled into the lasing mode and the device is considered to be thresholdless (Fig. 2 b)).

A metallic coaxial nanolaser provides an alluring approach for realizing such a source. Due it’s the ultra-small mode volume, the Purcell factor is large and only a sparse few modes are supported. In Fig. 3, a 3-D FEM coaxial resonator with outer and inner radii of 190 nm and 115 nm, and SiO\(_2\) and air plugs of 20 nm is simulated. In this geometry, a lone TEM-like mode falls within the spectral window of gain.
Figure 2  Spontaneous emission in a laser cavity. a) A standard laser cavity, light can emit to the continuous spectrum of radiation modes, into other cavity modes, and also into the actual lasing mode. b) A thresholdless resonator. All the light emission is funneled into the lasing mode.

Figure 3  3D FEM COMSOL simulation of a metallic coaxial resonator (outer radius: 190 nm, inner radius 115 nm, air and SiO2 plugs of 20 nm. A single TEM-like mode falls within the spectral window of gain. The next nearest supported modes are depicted to either side.
2.2. Modes of metallic nanolasers

Metallic nanolasers, in general, support modes similar to those observed in a hollow perfectly conducting circular waveguide. Due to the cylindrical nature of the cavity, it is easiest for formulate the electric field and magnetic field equations in terms of cylindrical coordinates, and these formulations must satisfy the Helmholtz equation, which can be deconstructed into three single variable equations and solved for independently [46]. The \( \phi \) component and the \( Z \) component will both form a set of harmonic equations and have the familiar sine and cosine solutions, while the radial component separates into Bessel’s equation. In order for the field equations to be finite valued at \( r = 0 \), the Bessel function of the first kind \( J_n(k_r r) \) must be used. The solution will be of the form, \( \Psi(r, \phi, z) = R(r)\Phi(\phi)Z(z) \). Equation 2.3 provides the solution to Helmholtz equation in cylindrical coordinates.

\[
\Psi_{k_r,n,k_z} = J_n(k_r r) \begin{cases} \sin(n\phi) \\ \cos(n\phi) \end{cases} e^{ik_z z} \tag{2.3}
\]

These modes can be classified into two categories, transverse magnetic (TM) or transverse electric (TE), in which case, there is no magnetic/electric field for the \( z \)-component, respectively. The TM components are given as below:

\[
E_r = \frac{\partial^2 \Psi}{\partial r \partial z} \tag{2.4}
\]

\[
E_\phi = \frac{1}{r} \frac{\partial^2 \Psi}{\partial \phi \partial z} \tag{2.5}
\]

\[
E_z = \left( \frac{\partial^2}{\partial z^2} + k_z^2 \right) \Psi \tag{2.6}
\]

\[
H_r = \frac{1}{r} \frac{\partial \Psi}{\partial \phi} \tag{2.7}
\]
\[ H_\phi = -\frac{\partial \Psi}{\partial r} \quad (2.8) \]
\[ H_z = 0 \quad (2.9) \]

For the case of the TM mode, the electric field must be equal to zero at the outer wall \( r = R_0 \) of the metal. In this case it imposes the condition \( J_n(k_r R_0) = 0 \). Since it is a Bessel function there are an infinite amount of solutions, and the appropriate value of \( k_r R_0 \) must be selected to satisfy the boundary conditions and is called \( x_{n,r} \). For example, a TM01 mode will have the solution of \( J_0 \left( \frac{x_{0,1}}{R_0} r \right) e^{ik z} \).

Likewise, the TE modes can also be found following a similar procedure. However, in this case \( E_\phi \) must be equal to zero at \( r = R_0 \).

\[ E_r = -\frac{1}{r} \frac{\partial \Psi}{\partial \phi} \quad (2.10) \]
\[ E_\phi = \frac{\partial \Psi}{\partial r} \quad (2.11) \]
\[ E_z = 0 \quad (2.12) \]
\[ H_r = \frac{\partial^2 \Psi}{\partial r \partial z} \quad (2.13) \]
\[ H_\phi = \frac{1}{r} \frac{\partial^2 \Psi}{\partial \phi \partial z} \quad (2.14) \]
\[ H_z = \left( \frac{\partial^2}{\partial z^2} + k^2 \right) \Psi \quad (2.15) \]

Since \( E_\phi = \frac{\partial \Psi}{\partial r} \), one can conclude that the derivative of the Bessel function must be equal to zero, \( J'_n(k_r R_0) = 0 \). In which case the solutions will have the form \( J_n \left( \frac{x'_{n,r}}{R_0} r \right) \left\{ \sin(n\phi) \right\} \cos(n\phi) e^{ik z} \).

Noticeably, in both the TE and TM cases there is a degeneracy, except for the TM0r and TE0r modes. Moreover, the modes that are supported in the cavity are limited by the magnitude
of the radius of the disk, and shrinking of the radius too much can lead to no well confined modes. Also of note is that these modes here are meant to describe a perfectly conducting metal and there will be slight deviations when this approximation is no longer valid, such as in metallic nanolasers operating at optical frequencies. However, in general these modes work well to describe the modes in a metallic nanolasers and will be used extensively in Chapter 6.

In addition to the modes described above that both coaxial and disk shaped nanolasers share, the coaxial devices also support an additional mode. A TEM like mode (both $E_z = 0$ and $H_z = 0$) that exhibits no cutoff frequency is supported. The equation for such a mode can be readily arrived at by considering the inner metal cable to be an infinitely long linear charge and an infinite long linear current [47]. In which case one arrives at an electric and magnet field of:

$$\vec{E} = \frac{e^{ikz}}{r} \hat{r}$$ \hspace{1cm} (2.16)

$$\vec{H} = \frac{\epsilon_0 \epsilon_r e^{ikz}}{\mu_0} \hat{\phi}$$ \hspace{1cm} (2.17)
CHAPTER 3: RATE EQUATION ANALYSIS

The demand for a smaller footprint, lower power consumption, and higher modulation bandwidth has fueled a host of activities in developing nanolasers as one of the key components of photonic integrated circuits [12]. One approach to overcome the challenges facing laser miniaturization is to use new designs based on metallic nanocavities [9,14,15,19–28]. In this section, a rate equation model is provided in order to study metallic nanolasers. Utilizing this tool, the threshold and output power, as well as the frequency response of a metallic nanolaser can be predicted with reasonable accuracy.

3.1. Rate Equations

At first glance, the low Q-factor of the resonator may insinuate that a nanoscale coaxial cavity requires a high pump power to reach the onset of lasing. However, a careful analysis shows that due to the high mode confinement ($\Gamma$), close to unity spontaneous emission coupling ratio, and large Purcell factor ($F$), such structures can indeed display both low threshold and unprecedentedly broad modulation bandwidths – two traits that are usually at odds with one another. Here we introduce a simplified rate-equation model that could further elucidate the laser dynamics in such arrangements.

\[
\frac{dn_c}{dt} = \frac{I}{q} - \frac{F\beta}{n_sp \tau_sp} \Gamma n_p n_c - \frac{F}{\tau_sp} n_c - \frac{1}{\tau_nr} n_c \tag{3.1}
\]

\[
\frac{dn_p}{dt} = \frac{F\beta}{\tau_sp n_sp} \Gamma n_p n_c + \frac{F}{\tau_sp} \beta n_c - \frac{1}{\tau_p} n_p \tag{3.2}
\]
Here $n_c$ and $n_p$ are the total electron-hole pair and the number of photons, $I$ is the injection current, $q$ is the elementary charge, and $\tau_{sp}$ and $\tau_{nr}$ are the bulk spontaneous and non-radiative lifetimes. The cavity lifetime is $\tau_p = Q \lambda / 2\pi c$, where $\lambda$ is the wavelength and $c$ the speed of light in vacuum, Lastly $n_{sp} = (f_c (1 - f_v)) / (f_c - f_v)$, where $f_c$ and $f_v$ are the Fermi-Dirac function in the conduction and valence bands [42,48–50]. These equations also include the relation of the Einstein A and B coefficients, in which the spontaneous emission and stimulation emission rates are directly proportional to one another. So an increase of the spontaneous emission rate characterized by the Purcell factor will also equally increase the stimulated emission rate [51,52]

At steady-state, the threshold current and the number of photons in the laser mode are given as:

\[
I_{th} = \frac{2\pi cn_{sp}}{\lambda} \frac{q}{\beta \Gamma Q} \left(1 + \frac{\tau_{sp}}{\Gamma \tau_{nr}}\right) \tag{3.3}
\]

\[
n_p = \frac{1}{2} \left[ \frac{I}{q \tau_p} - \frac{n_{sp}}{\beta \Gamma} \left(1 + \frac{\tau_{sp}}{\Gamma \tau_{nr}}\right) \right] + \frac{I}{q \tau_p} \frac{n_{sp}}{\beta \Gamma} + 1 / 4 \left[ \frac{I}{q \tau_p} - \frac{n_{sp}}{\beta \Gamma} \left(1 + \frac{\tau_{sp}}{\Gamma \tau_{nr}}\right) \right]^2 \tag{3.4}
\]

It can be seen from Eq. (3.3) that the threshold is inversely proportional to the product of $\beta$, $\Gamma$ and $Q$. On the other hand, the Purcell factor contributes in reducing the threshold only through increasing the decay rate of the spontaneous emission in respect to that of the non-radiative processes. However, in most semiconductor gain systems where
A light-emitting device is considered a laser if stimulated emission dominates the output power. Typically, for a laser, the Light-Injection (L-I) or Light-Light (L-L) curve in the logarithmic-logarithmic (log-log) scale is expected to consist of three distinct regions. At low pump intensities, the device is in the photoluminescence (PL) regime, where it operates as an LED and the slope of the L-I or L-L curve is unity. At higher pump levels, it experiences a sudden increase in the power allocated to the future lasing mode – creating a nonlinear kink that is known as the amplified spontaneous emission regime (ASE) and the slope of the log-log curve is larger than one. By further increasing the pump energy, the slope returns to unity and the device operates in the lasing mode. Figure 4a) and Fig. 4 b) display the Light-Current (L-I) curves for two lasers: one is the coaxial nanolaser as presented in Fig. 1 with a spontaneous emission coupling factor of unity ($\beta \sim 1$), and the other one is a typical microscale laser (parameters are extracted from [53] for a vertical cavity surface emitting laser (VCSEL)). While in simulation, the trajectory of the spontaneous and stimulated emission curves (dash-dotted dotted and dashed-dotted lines) can specify the threshold power, for a device with $\beta \rightarrow 1$, the lack of a discernible kink in the total output power makes it virtually impossible to determine the threshold via L-I or L-L measurements. Such a soft transition from PL to lasing is known as thresholdless behavior. Using the parameters provided in Fig. 4, the corresponding lasing thresholds for the coaxial nanolaser and the VCSEL are 1.65 $\mu$A ($\sim 2$ kA/cm$^2$) and 0.794 mA ($\sim 700$ A/cm$^2$), respectively.
Figure 4  Light-Current curve for a thresholdless coaxial nanolaser a) and a VCSEL b). Green (solid) is the total power in the mode, blue (dashed) the stimulated power contribution, and red (dash-dotted) the spontaneous power contribution. Using Eq. 3.3, the threshold current of the nanolaser and the VCSEL are 1.65 µA and 0.794 mA, respectively. Plot of the 3dB modulation frequency as a function of injection current for the nanolaser c) and VCSEL d). The nanolaser exhibits a modulation response of over 1 THz.
3.2. Direct modulation response

The direct modulation behavior of a single mode laser diode can be derived from a small signal analysis \( n_c \rightarrow n_c + \Delta n_c, n_p \rightarrow n_p + \Delta n_p, \text{ and } I \rightarrow I + \Delta I \) performed on Eq. (3.1 & 3.2) [53]. The impulse response of the system is defined as:

\[
H(f) = \frac{f_r^2}{f_r^2 - f^2 + i\xi f} \quad (3.5)
\]

Here \( f_r \) is the relaxation frequency and \( \xi \) is the damping coefficient, which are given as below

\[
(2\pi f_r)^2 = \left( \frac{FF\beta n_p}{\tau_{sp}\tau_{sp}} + \frac{FF\beta n_c}{\tau_{sp}\tau_{sp}} \right) + \left( \frac{1}{\tau_p} - \frac{FF\beta n_c}{\tau_{sp}\tau_{sp}} \right) \left( \frac{FF\beta n_p}{\tau_{sp}\tau_{sp}} + \frac{F}{\tau_{sp}} + \frac{1}{\tau_{nr}} \right) \quad (3.6)
\]

\[
2\pi \xi = \left( \frac{1}{\tau_p} - \frac{FF\beta n_c}{\tau_{sp}\tau_{sp}} \right) + \left( \frac{FF\beta n_p}{\tau_{sp}\tau_{sp}} + \frac{F}{\tau_{sp}} + \frac{1}{\tau_{nr}} \right) \quad (3.7)
\]

Of particular importance is to find the 3dB frequency \( f_{3dB} \), where \( |H(f)|^2 = 1/2 \),

\[
f_{3dB} = \sqrt{f_r^2 - \frac{\xi^2}{2}} + \frac{1}{2} \sqrt{8f_r^4 - 4f_r^2\xi^2 + \xi^4} \quad (3.8)
\]

Figure 4 c) and Fig. 4 d) show the 3 dB bandwidth as a function of the injection current for the nanolaser and VCSEL. While at ten times the threshold power, a typical microscale VCSEL is expected to show a modulation bandwidth of up to 30 GHz, a coaxial nanolaser can potentially show a much larger bandwidth on the order of a few hundreds of GHz. Such unprecedented large modulation bandwidth is perhaps one of the most compelling drives behind the research towards developing such nanoscale lasers.
CHAPTER 4: SECOND-ORDER COHERENCE OF METALLIC NANOLASERS

In this chapter the second-order coherence properties of metallic coaxial and disk-shaped nanolasers are explored near and above their classically defined lasing threshold. Section 4.1 describes second-coherence as it pertains to light, Section 4.2 describes the coaxial laser cavity design and its first-order optical properties. In Section 4.3 an approach for measuring the second-order coherence of broad linewidth radiation sources, using a Hanbury Brown and Twiss interferomic method, is established and used to determine the nature of the emitted light from a number of multiple quantum-well metal-clad nanolasers.

4.1. Second-order coherence

An unambiguous measure to determine the nature of a given emission is the second-order coherence function:

\[ g^2(\tau) = \frac{\langle I(t)I(t+\tau) \rangle}{\langle I(t) \rangle^2} \] (4.1)

which is an intensity correlation function of the radiation that measures the time dependent amplitude of the emission. The intensity fluctuations of light are in general classified as chaotic/bunched \((g^2(0) > 1)\), coherent/Poissonian \((g^2(0) = 1)\), or lastly anti-bunched/sub-Poissonian \((g^2(0) < 1)\) [54]. Generally, bunched light is characterized as photons emitted within a timescale on the order of the temporal coherence \((\tau_c)\) (Fig. 5 a)), coherent light as photons that are emitted at statistically random intervals (Fig. 5 b)), and anti-bunched, the emission of a single photon at a time (Fig. 5 c)).
In this regard, the second-order coherence differs fundamentally from the first-order coherence function \( g^1(\tau) \), which serves as a description of phase. For example, a white thermal source spectrally filtered to such a degree that the temporal coherence is equal to that of a laser may be deemed "classically coherent." However, in this case, the amplitude fluctuations associated with the statistical nature of the photons’ arrival would still be present in the emitted light. The second-order coherence function can be used to further characterize the emission properties of light – in particular, lasers. For a laser device, below threshold, light is expected to be super-Poissonian (Fig. 6 regime I) with the amplitude fluctuating on a timescale that is inversely proportional to the linewidth of the emission, or \( \tau_c \). As the laser is pumped at even higher powers, a larger share of radiated light is generated through stimulated emission (Fig. 6 regime II). Near the classically defined threshold the emission transitions to a coherent state; i.e., the photons’ arrivals follow a Poissonian distribution (Fig. 6 regime III) \[55\]. The transition from chaotic to coherent emission in a laser can be characterized by using the Siegert relation \[56\] to yield the second-order coherence of the source. Where the Siegert relation is given as follows:

![Figure 5](image.png)

An example of the arrival of photons for different classifications of light. a) bunched light (red circles), packets of photons radiate within timescales on the order of the temporal coherence. b) coherent radiation (green circles), the photons emit at statistically random times, obeying a Poissonian distribution. c) anti-bunched light (blue circles), photons are emitted one at a time.
\begin{equation}
g^2(\tau) = 1 + |g^1(\tau)|^2 \tag{4.2}
\end{equation}

Since the light emitted from a laser has the following Lorentzian lineshape:

\begin{equation}
L(\nu) = \frac{1}{2\pi(\nu-\nu_0)^2 + (\frac{1}{2}\Delta\nu)^2} \tag{4.3}
\end{equation}

where \( \nu \) is the frequency, \( \Delta\nu \) is the full width at half maximum (FWHM) of the frequency, and \( \nu_0 \) is the central frequency. Its Fourier transform in the time domain is given as:

\begin{equation}
g^1(\tau) = e^{-i2\pi\nu_0\tau - \pi\Delta\nu|\tau|} \tag{4.4}
\end{equation}

With \( \pi\Delta\nu = \frac{1}{\tau_c} \), resulting in \( g^2(\tau) = 1 + |g^1(\tau)|^2 = 1 + e^{\frac{2|\tau|}{\tau_c}} \). An example of the second-coherence function is plotted in Fig. 7, depicting bunching (Fig. 7 a)), coherent light (Fig. 7 b)), and anti-bunching (Fig. 7 c), this is plotted with a Gaussian lineshape).
Figure 6  An L-L curve for a single moded typical laser with $\beta \ll 1$. In regime I, the light generated is almost entirely from spontaneous emission and the photons are bunched. In regime II, lasing action begins to occur and the light is a mixture of stimulated and spontaneous emission. In regime III, the total radiated light is dominated by the stimulated emission and the photon statistics are Poissonian. Spontaneous emission is denoted as a red dashed line, stimulated emission as a blue dashed line, and the total emission as a solid green line. Bunching is depicted as a red circle, coherent radiation as a blue circle.

Figure 7  Example of the second-order coherence function for a) bunched, b) coherent, and c) anti-bunched light.
4.2. Cavity design

In recent years, there has been tremendous progress in the development of metallic and metallo-dielectric nanoscale lasers [13,17,18,20,24,26–33,35–38,40,57–61]. These advances are largely motivated by their small footprint and potential for high-speed operation, which makes such nanolasers great candidates for on-chip sources in photonic integrated circuits [61]. Through the use of metal as cladding, the volume of the laser cavity can be reduced to subwavelength dimensions without significantly compromising the mode confinement ($\Gamma$). If designed properly, the portion of the spontaneous emission coupled into the lasing mode ($\beta$) can even approach unity, in which case the laser is said to be thresholdless [30].

Determining the onset of coherent emission in such high-$\beta$ resonators can prove challenging [55,62–65]. Typically, a light-light (L-L) or light-current (L-I) curve, where the output power is measured as a function of incident light or injection current, can be used to resolve whether a light-emitting device is a laser. In general, when the L-L or L-I curve is plotted in a logarithmic scale, it will exhibit an “S” shape, which consists of three regions. At the lower left end is the photoluminescence (PL) dominant region where the L-L or L-I curve can be represented by a line having a theoretical slope of 1, in the middle is a sharp jump in the intensity due to the prevalence of amplified spontaneous emission (ASE), and at the upper right end is the lasing region where the curve regains its unity slope. However, these three regimes are only readily discernible in lasers having a low spontaneous emission coupling factor ($\beta \ll 1$). As $\beta$ increases, the sharpness due to the ASE begins to soften. When $\beta \rightarrow 1$, the nonlinearity from the ASE completely disappears and the ensuing curve becomes
a line in its entirety [30,66,67]. Consequently, merely assessing the L-L or L-I curve is no longer a viable approach for determining the lasing threshold. It should be noted that the situation depicted above, with unity slopes in the PL and lasing regions, is only valid under the assumption that most processes involved are radiative and no thermal roll-over is present. In the majority of semiconductor lasers, where the nonradiative recombination processes cannot be neglected, it is expected that the slope of the lines will deviate from unity at the lower end of the PL (due to surface recombination), and at the higher end of the lasing regime (due to Auger recombination) – a set of trends that can make the L-L or L-I curve of a light-emitting diode (LED) appear like that of a laser.

So far, lasing operation has been demonstrated in a number of metal-coated nanocavities [13,18,24,28,30,57,58]. However, there has been a debate as to whether the light emitted from these structures is truly coherent. Generally, such viewpoints are motivated by the relatively broad linewidth of the emission, and the lack of readily distinguishable regions in the L-L curve. This is particularly the case in metallic coaxial nanolasers that can simultaneously exhibit a high β, a low quality factor (Q-factor), and a high Γ [30].

Figure 8 a) provides a schematic of the coaxial nanolaser under study. It is composed of a metallic rod (R_{core}: 50 nm) surrounded by a metal-coated semiconductor ring (Δ: 200 nm, h₂: 210 nm). The gain-medium (ring) consists of six vertically stacked quantum wells with an overall height of 200 nm, each composed of a 10 nm thick well (Inₓ=0.56Ga₁₋ₓAsᵧ=0.938P₁₋ᵧ) sandwiched between two 20 nm thick barrier layers (Inₓ=0.734Ga₁₋ₓAsᵧ=0.57P₁₋ᵧ). The quantum wells are covered by a 10 nm thick InP overlayer for protection. The upper and lower ends
of the ring are terminated by silicon dioxide (SiO$_2$) and air plugs (heights $h_3$: 30 nm and $h_1$: 20 nm, respectively). The PL spectrum of the bare quantum-well system is measured at several pump powers at a temperature of 77 K. The PL spectrum is depicted in the spectral window of gain in Fig. 8 b) at a pump power comparable to that required to reach lasing operation in our devices. The modal content of the nanolaser is obtained using electromagnetic (EM) simulations based on the finite element method (FEM) and is displayed in Fig. 8 b). The EM simulations are performed with material parameters at a temperature of 77 K (permittivity of silver, $-90 - 1.0i$; quantum-well gain system, 11.35; InP, 9.8; SiO$_2$, 2.2). Our simulations indicate that the coaxial resonator under study supports three modes within the gain bandwidth of the active medium: two degenerate whispering-gallery-type modes at 1303 nm as well as a gap-plasmon-like mode at 1373 nm. It should be noted that small variations of permittivities (<2%) and dimensions (<5%) around the nominal values do not change the modal content within the gain bandwidth (in particular, only their corresponding wavelengths, and to a small extent their Q-factors, are affected). In this simulation, $\Gamma$ is determined through an appropriate normalized overlap integral, and the effective modal volume is calculated as

$$V_m = \int_{V_a} \frac{dV \epsilon_g(r) |E(r)|^2}{\max(\epsilon_g(r) |E(r)|^2)}$$

where $V_a$ is the volume of the active region, and $\epsilon_g = \frac{d(\omega \epsilon)}{d\omega}$ [68]. Although all three modes can potentially participate in the lasing process, the higher Q-factor, as well as the larger $\Gamma$ of the set of the degenerate modes, places them first in line to reach the lasing
threshold. The spontaneous emission coupling factor for this cavity is found by calculating the emission from a randomly oriented dipole in a random location within the active region. β is estimated as the ratio of the emitted power at the wavelength of the desired lasing mode to the total power radiated by the dipole, weighted by the photoluminescence profile of the bare quantum-well system. For the modes at 1303 nm, the calculated β is divided by half to account for the degeneracy. This procedure is repeated for 10 random dipoles, and the results are averaged to find a β ~ 0.048. It should be noted that the above value of β is still considerably larger than most micro-scale semiconductor lasers, e.g., for VCSELs β < 10^{-3}. While coaxial structures with different dimensions can exhibit higher spontaneous emission coupling factors, in this work, we focused our attention on the above device because of its higher output power due to its improved out coupling. The higher output power allows us to study the second-order coherence properties closer to the threshold condition.

Figure 8 Coaxial nanolaser geometry and modes. a), Illustration and b), modal content of the metallic coaxial nanolaser. The resonator supports three modes within the gain bandwidth of the active medium: a pair of degenerate modes at 1303 nm and a gap-plasmon-type mode at 1373 nm. Q and Γ denote the quality factor and the extent of energy confinement to the semiconductor region, V_m, the effective modal volume. The color bar shows normalized |E|^2, where E is the electric field. Nominal permittivity values are used in this simulation.
4.3. Experiment

The coaxial laser is fabricated using the procedure described in Section 6.3 and characterized in a micro-photoluminescence (μ-PL) setup to collect the evolution of the spectrum (Fig. 9 a)), the L-L curve (Fig. 9 b)), and the linewidth (Fig. 9 c)). The nanolasers are pumped optically using a continuous wave (CW) single-mode fiber laser operating at 1064 nm. All nanolasers reported in this paper are cooled to a temperature of 77 K – mainly to boost the laser efficiency in order to be able to perform the subsequent second-order coherence measurements. The spectral evolution of the above laser is shown in Fig. 9 a). At lower pump powers the gap-plasmon mode at 1373 nm is the first resonance to appear in the PL, because spectrally it is closer to the gain peak. However, due to its smaller Q-factor and lower Γ, this mode does not reach the threshold condition. As the pump power increases, one of the modes at 1303 nm emerges – ultimately dominating over the other modes. This behavior of the modes is in excellent agreement with the simulation results of Fig. 8 b). The L-L curve of this laser is plotted in a logarithmic scale in Fig. 9 b) and in a linear scale in the inset. The three characteristic regions of the logarithmic L-L curve are emphasized with red lines plotted in Fig. 9 b) – the PL near the lower left corner, the ASE in the center, and the lasing in the upper right. The upsurge in output power that is expected in the ASE region begins around 20 μW and softens at 70 μW, where the device appears to transition into lasing operation. The emission power from the laser continues to increase, until ultimately Auger recombination and to some extent thermal roll-over become predominant, causing the output power to decrease. It should be noted that the output power reported in Fig. 9 b) presents the actual power collected off the sample via an objective with a numerical aperture
(NA) of 0.42 and intensity transmission of \( \sim 57\% \) at \( \sim 1300 \text{ nm} \). Due to the limited NA of the objective lens, it is estimated that the power at the exit aperture of the laser is about 10 times greater than the values provided in Fig. 9 b). Finally, the measured linewidth of the emitted light is plotted in Fig. 9 c), showing a sharp decrease in the emission linewidth until it levels off at around 80 \( \mu \text{W} \). The inset of Fig. 9 c) displays the Lorentzian fit used in determining the linewidth. Far above threshold, as the pump power increases, the linewidth slightly broadens. This behavior is almost universal in all the nanolasers we studied, both coaxial and disk-shaped, and to some extent may be attributed to the optical pumping scheme that introduces heating and carrier fluctuations. For the above reported laser, the minimum measured linewidth is \( \sim 0.7 \text{ nm} \). This relatively broad linewidth is a byproduct of the large \( \Gamma \) and high \( \beta \). In this regard, a large portion of the spontaneous emission lands in the lasing mode. This noise is then amplified by the gain due to the high mode confinement. In fact, as suggested in a new study, an effective strategy to achieve an ultra-narrow linewidth in semiconductor lasers is through reducing both \( \Gamma \) and \( \beta \) [69].
To further investigate the nature of the radiation from nanolasers, a modified HBT setup is prepared. Here, the light under study is split by a 50:50 coupler, and guided into two arms of the interferometer where each arm is equipped with a single photon avalanche diode (SPAD). Upon the arrival of a photon, the first SPAD triggers a time-correlated single photon

Figure 9 Characterization of the nanolaser under CW optical pumping at a temperature of 77 K. a), the spectral evolution of the laser, b), the light-light curve in a logarithmic scale and linear scale (inset), and c), the linewidth versus pump power. The Lorentzian fit used to estimate the linewidth is depicted in the inset of c).

To further investigate the nature of the radiation from nanolasers, a modified HBT setup is prepared. Here, the light under study is split by a 50:50 coupler, and guided into two arms of the interferometer where each arm is equipped with a single photon avalanche diode (SPAD). Upon the arrival of a photon, the first SPAD triggers a time-correlated single photon
counting (TCSPC) module operating in start–stop mode, setting the start point. When the
second SPAD detects a photon, a stop signal is sent to the TCSPC module, and the resulting
time delay \( \tau \) shows the arrival correlation of the photons. For chaotic light, at zero time delay
\( (\tau = 0) \) a coincidence peak is expected – a direct result of photon bunching. On the other
hand, for coherent radiation with a Poissonian distribution, the resulting correlation
function is expected to be unity for all time delays. It should be noted that, similar to other
interferometric setups, the aforementioned coherence properties may only be observed
within the coherence time of the emission, with the maximum second-order coherence value
at \( \tau = 0 \) being given by the relation:

\[
g^2(0) = 1 + \frac{\tau_c}{2\tau_r} \left( 1 - e^{-\frac{2\tau_r}{\tau_c}} \right)
\]

where \( \tau_r \) is the temporal resolution of the detectors. Consequently, in order to measure the
second-order coherence of broad linewidth sources, either the detection system must have
very good timing resolution (in the order of a few tens of femtoseconds) or the emission
must first be spectrally filtered such that its temporal coherence becomes larger than the
timing resolution of the SPADs and TSCPC module [56]. For example, Fig. 10 a) depicts a
situation where the temporal coherence is much greater than the resolution of the detectors
\( (\tau_c = 10\tau_r) \), in this scenario, the maximum \( g^2(0) = 1.91 \). However, if \( \tau_c \ll \tau_r \) (Fig. 10 b)), it
can be seen that the maximum detectable \( g^2(0) \) is near unity in this case even though the
source is chaotic. Limited by the timing resolution of the currently available single photon

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counters, we chose to spectrally filter the radiation to extend the coherence time of the radiation under study [70]. It is known that spectral filtering when performed out of the laser cavity cannot alter the higher-order coherence function of a noncoherent beam to resemble that of coherent light [71].

The experimental setup, which includes both a μ-PL characterization station and a modified HBT interferometer, is illustrated in Fig. 11 a). The HBT interferometer is incorporated into the μ-PL setup via a kinematic mirror that redirects the light through a spectral filtering stage. The filter is composed of a cascaded arrangement of a diffraction grating and a Fabry–Perot resonator (Thorlabs SA210-12B) to increase the temporal coherence to \( \sim 3.75 \) ns, well beyond the resolution of the SPADs (IDQ ID220, resolution: 240 ps). A linear polarizer is used to remove undesired polarization components. The spectrally filtered light is then collected using a single-mode fiber, split into equal parts with a 50:50 directional coupler, and directed to the SPADs through fibers each equipped with a variable

![Figure 10](image.png)

Figure 10 Dependence of maximum \( g^2(0) \) on the temporal resolution of the single photon detectors in relation to the temporal coherence of the measured emission. a) When \( \tau_c \gg \tau_r \) (\( \tau_c = 10\tau_r \) in this scenario) \( g^2(\tau) \) is near its maximum value of 2 when measured bunched light. b) When \( \tau_c \ll \tau_r \), the maximum detectable \( g^2(0) \) is severely limited.

The experimental setup, which includes both a μ-PL characterization station and a modified HBT interferometer, is illustrated in Fig. 11 a). The HBT interferometer is incorporated into the μ-PL setup via a kinematic mirror that redirects the light through a spectral filtering stage. The filter is composed of a cascaded arrangement of a diffraction grating and a Fabry–Perot resonator (Thorlabs SA210-12B) to increase the temporal coherence to \( \sim 3.75 \) ns, well beyond the resolution of the SPADs (IDQ ID220, resolution: 240 ps). A linear polarizer is used to remove undesired polarization components. The spectrally filtered light is then collected using a single-mode fiber, split into equal parts with a 50:50 directional coupler, and directed to the SPADs through fibers each equipped with a variable
optical attenuator (VOA). Lastly, the SPADs are connected to a TCSPC module (PicoHarp 300) to collect the time-correlated histograms. The overall loss in the setup, associated with the HBT interferometer, is found to be $\sim 69$ dB – this includes both the losses related to the optical components after the kinematic mirror ($\sim 36$ dB) as well as the power filtered out due to the linewidth narrowing ($\sim 33$ dB, ultimately depending on the spectral linewidth of the radiation).

In order to establish the second-order coherence capability of the setup, we first measure the $g^2(\tau)$ for a commercially available laser (Agilent 81460A). The laser is set to generate an output emission at 1550 nm with a linewidth of 50 MHz (using the coherence control module) – schematically shown in Fig. 11 b). In this case the measured $g^2(\tau)$ function is a flat line across all time delays – confirming that the source is indeed a laser (Fig. 11 c)). We then measure the second-order coherence function for a commercially available ASE source (Amonics ALS-CL-20). The output from the ASE source has a spectral width of $\sim 50$ nm centered at $\sim 1550$ nm as displayed in Fig. 11 d) – yielding a coherence time on the order of 100 fs. Without spectral filtering, the $g^2(\tau)$ of the ASE source also resulted in a flat line (featureless) – falsely resembling that of a coherent source.
However, by using the filtering scheme incorporated in the setup, the emitted radiation from the ASE source is narrowed down to ∼85 MHz. For this spectrally narrowed emission the second-order coherence function is no longer a line. Instead, it clearly shows a coincidence peak of $g^2(0) = 1.864 \pm 0.025$ (Fig. 11 e)). In this case, the collected coincidence data (presented with dots in the figure) are fitted using the Siegert relation (Eqn. 4.2). From this fitting, the temporal coherence was estimated to be $\tau_c = 3.75$ ns – agreeing well with the linewidth expected from the cascaded diffraction grating and the Fabry–Perot filter. The fact that a broadband ASE source with intrinsic spectral linewidth of ∼50 nm is capable of demonstrating a $g^2(0)\sim1.864$ confirms that the current setup can be used for characterizing the second-order coherence properties of an arbitrary source with a broad linewidth.
After calibrating the HBT setup to reliably characterize the statistical nature of the light from an arbitrary source with broad linewidth, the second-order coherence measurements are performed for a number of coaxial and disk-shaped lasers with various radii. It should be noted that in order to be able to compare the results and to ensure the accuracy of the measurements, most of the intensity correlation data are collected over the same period of time (~7 min) and count rate (~50 KHz).

Figure 12 displays the measured second-order coherence function at different pump levels, along with the logarithmic single-shot emission spectrum, for the coaxial nanocavity described in Section II. Far above threshold at a pump power of 215 μW, the measured $g^2(\tau)$ is a flat line, where the fit suggests a $g^2(0) = 1.009 \pm 0.038$ (Fig. 12 a)) – confirming that the coaxial structure under study is, to a good approximation, capable of generating coherent radiation. A similar measurement at a pump power of 79.7 μW shows a slightly $g^2(0)$ of $1.037 \pm 0.039$ (Fig. 12 c)). Finally, the second-order coherence measured barely below the classically defined threshold, at a pump power of 66.4 μW, yields $g^2(0) = 1.081 \pm 0.033$ (Fig. 13 e)). Due to the limited output power at a pump power of 66.4 μW, the presented data were collected during a period of 20 min and at a count rate of 20 KHz. Even for this measurement, $g^2(0)$ is still considerably smaller than 2 – suggesting that the transition from chaotic to coherent is quite gradual. All the measurements are fitted in the same manner as the ASE data, maintaining the previously determined coherence time of 3.75 ns. Further investigation of the characteristics of the emission at yet lower pump powers could not be carried out with our current setup due to the limited output power near the threshold and the low efficiency of the detectors.
The second-order coherence function of the nanoscale coaxial laser reported in Fig. 12 matches the theoretical predictions for lasers with a high spontaneous emission coupling factor ($\beta$). Where just below threshold a small yet notable component of spontaneous emission is present, while at and well above threshold this contribution diminishes and the emission approaches that of an ideal coherent source [55]. Surprisingly, even around threshold, the radiation from this device appears to be quite coherent.

We also measured the second-order coherence function for a number of disk-shaped nanocavities with various radii. These cavities share an almost identical structure to the coaxial nanolaser with the exception that the silver core is replaced with the gain material. Figure 13 displays the second-order coherence functions along with logarithmic scale single-
shots of the emission spectra for two of the example disk-shaped resonators (with radii of 250 nm and 900 nm). The 250 nm radius disk with a single-mode emission is studied above threshold. The resulting intensity coincidence peak is $g^2(0) = 1.022 \pm 0.038$, confirming that this light-emitting device can generate coherent radiation (see Figs. 13 a) and 13 b)). The electromagnetic simulations for this cavity suggest that the spontaneous emission coupling factor is on the order of $\beta = 0.22$. Next, the larger disk with a radius of 900 nm is investigated (see Figs. 13 c) and 13 d)). For this device, the measured $g^2(0)$ is $1.485 \pm 0.043$.

As is clear in Fig. 13 d), the disk resonator with a radius of a 900 nm supports several competing modes. It seems that the simultaneous presence of multiple modes can cause the emission to become more chaotic [70]. Whether the observed $g^2(0) > 1$ is an indication that this device is operating below threshold, or the peak appearing at zero time delay is instead caused by the beating between independent modes, is yet to be fully investigated. Future investigations to solve these fundamental questions will, however, require detectors with higher sensitivity and/or better timing resolutions. It should be noted that the deviation from $g^2(0) = 1$ in larger nanocavities was regularly observed when characterizing nanolasers with multi-moded spectra.

The areas of nanophotonics and plasmonics have progressed tremendously in the past couple of decades. Undoubtedly, the introduction of metallic structures has opened a path toward light confinement and manipulation at the subwavelength scale – a regime that was previously thought to be out of reach in optics. Of central importance in this endeavor is to devise subwavelength light-emitting devices that can power up future nanoscale photonic circuits. The metal-clad coaxial and disk-shaped nanoresonators can provide viable
platforms to implement such sub-wavelength sources. They support ultra-small cavity modes and offer large mode-emitter overlap as well as multifold scalability. In addition, because of their small size and high Purcell factor, metallic nanolasers are expected to show large direct modulation bandwidths. Furthermore, coherent radiation generally has a lower relative intensity noise (RIN) in comparison to incoherent light from LEDs; hence metallic nanoscale lasers are expected to operate more reliably as high-speed devices [72].

4.4. Conclusion

In this chapter, the results of our measurements for the second-order coherence functions of coaxial and disk-shaped nanoscale lasers (with InGaAsP multiple quantum-well gain systems) were detailed. These measurements were accomplished by establishing a

![Figure 13](image)

Figure 13  Second-order coherence measurements results for nanoscale disk-shaped lasers. The $g^2(\tau)$ measurements of the emitted light from the disk-shaped nanolaser as well as the corresponding emission spectra in a semi-logarithmic scale for disk radii of a),b), 250 nm and c),d), 900 nm.
setup to reliably characterize the intensity correlation function for broad linewidth sources at telecommunication bands. In order to ensure the capability of our setup to measure $g^2(\tau)$, the second-order coherence is first measured for a spectrally broad ($\sim 50$ nm) commercial ASE source. In addition, by optimizing the design and modifying the fabrication process, we developed a number of nanolasers that are capable of generating relatively high output powers (few to few tens of microwatts) and could operate under CW pumping for an extended period of time.

The second-order coherence measurement results presented in this chapter unambiguously confirm that nanoscale coaxial and disk-shaped metallic cavities can generate coherent radiation ($g^2(0) \sim 1$). These studies may in turn shed light on the quantum properties of the emission from metallic nanoscale light sources.
CHAPTER 5: ELECTRICALLY INJECTED NANOLASERS

Small sized and scalable lasers that exhibit low threshold and high-speed direct modulations are expected to play an important role in the future of data processing and transmission. As detailed in earlier sections, metallic nanolasers have shown themselves to be a very promising candidate in this regard. However, in many of these demonstrations the lasers were optically pumped and/or lased at cryogenic temperatures. For most settings, this is not a practicable option. Instead, it is essential to generate the gain through means of electrical injection at room temperature. This chapter goes over design considerations to meet this goal and also shows the demonstration of electrically injected metallic nanolasers operating at temperatures up to 140 K.

5.1. Electrically injected metallic nanolaser design

A schematic of an electrically injected coaxial nanolaser is provided in Fig. 14. The cavity is similar in design to that shown in Fig. 1, but with a few notable differences. For starters, the wafer structure needed to be modified to allow for the injection of holes and electrons. Thus the gain region is cladded by p- and n-type InP, and the contact layers are composed of p+ InGaAsP and n+ InGaAs. This cladding increases the dimensions of the structure in the vertical direction, increasing the overall size of the cavity. On the other hand, the use of direct current electricity as the pumping mechanism causes the devices to experience higher operating temperatures than their optically pulsed cohorts, due to resistivity heating in the cladding layers. Moreover, an electrically insulating dielectric layer is deposited on the entire surface of the device – isolating the laser diode electrically from
the surrounding silver. Unfortunately, this dielectric layer also acts as a thermal insulator and can lead to less effective heat dissipation. However, the encapsulating metal provides an effective method of dissipating excess heat and if properly designed, this is not expected to be a major issue in these devices. For structural support of the n-contact wire, a layer of Benzocyclobutane (BCB) is spun and cured on the sample. The last major difference associated with these devices is that they are now mounted on a TO-8 header package. More details on the fabrication process are provided in Section 5.2.

5.1.1 Surface recombination

Surface recombination is a non-radiative recombination process that plays a degrading role in all semiconductor lasers, but is more acute in small devices such as nanolasers. In this process, an electron in the conduction band drops to an intermediate state between the conduction and valence band, and then again falls to the valence band. The
energy that the electron in the conduction band had is now converted to thermal energy as the electron did not recombine with a hole. Making this process reducing the electrical injection efficiency and also increased the heat in the laser, which provides further complications.

Surface recombination is brought about in semiconductor lasers due to abrupt changes in crystal structure, which can result in dangling bonds, such as when a material is etched. Due to the high quality of MBE growth now days, stacks of differing semiconductors have well defined interfaces and negligibly contribute to this surface recombination. Furthermore, the characterization of surface recombination rate is directly linked to a surface recombination velocity, \( \nu_s \). Where a high surface recombination velocity denotes that the material will have many traps for holes and electrons, and leads to a larger recombination rate.

In larger semiconductor lasers, surface recombination can be viewed as a minor issue, due to the significantly larger surface area to volume ratio of these cavities. However, as the size of the cavity is reduced, it is more likely that electrons and holes will be located near the etched surfaces, leading to an increased surface recombination rate. An example of two active gain regions of a disk and coaxial nanolaser are provided in Fig. 15 a) & b). It is observed from this that in the case of the coaxial nanolaser that the surface recombination will play a larger factor than in the disk geometry for cavities of equal outer radii.
As the size of the device decreases, the surface area to volume ratio becomes larger and surface recombination becomes a larger aspect.

The equation governing the surface recombination rate at high injection levels is given below [53]:

$$ R_{sr} = \frac{\text{Area}}{\text{Volume}} v_s N $$  \hspace{1cm} (5.1)

Where $R_{sr}$ is the surface recombination density rate, and $N$ is the carrier density. As seen from the equation, this is a linear relationship since either one electron, or one hole is involved in the capturing process, and the magnitude of the equation heavily depends on the area to volume ratio and the surface velocity. However, given that cavities support modes at certain eigenfrequencies, which entirely depend on device geometry, it is not feasible to alter the surface area to volume ratio. In this regard, efforts have focused on techniques to lower the surface recombination velocity. The surface recombination lifetime can be defined as:

$$ \tau_{sr} = \frac{\text{Volume}}{\text{Area}} \frac{1}{v_s} $$  \hspace{1cm} (5.2)
Common techniques for passivating the surface in these nanolaser structures are the deposition of the dielectrics Si₃N₄ [14] and SiO₂ [15] via plasma enhanced chemical vapor deposition (PECVD), or Al₂O₃ through atomic layer deposition (ALD) [73]. Another method for reducing the surface recombination is immersing the lasers in ammonium sulfide (NH₄)₂S, a process that has been successfully utilized in VCSELs [74]. In Table 1, the numbers were computed for a coaxial device with outer radius of \( R_2 = 400 \, \text{nm} \) and inner radius \( R_1 = 200 \, \text{nm} \), that lases at a current density level of \( N = 6 \times 10^{18} \, \text{cm}^{-3} \), using the technique outlined in [60] to passivate the surface of a bulk In₀.₅₃Ga₀.₄₇As active material. Initially, the current lost due to surface recombination, given as:

\[
I_{sr} = q \frac{N_{th}}{\tau_{sr}} Volume
\]  

is large, especially when considering the ideal relatively low threshold associated with a device of this size (~2 µA ignoring non-radiative recombinations). Moreover, the ammonium sulfide treatment reduces the surface velocity by a factor of 4, in turn reducing the current by the same factor. This technique used in addition to a SiO₂ deposition has been shown to reduce the surface recombination current to levels nearing the threshold. Considering a typical value of radiative recombination lifetime on the order of \( \tau_r \sim 1 \, \text{ns} \) in bulk semiconductor materials at this wavelength, it becomes obvious that increasing the surface recombination lifetime \( \tau_{sr} \) is of importance. It should be noted that reducing the surface recombination by deposition of dielectrics can also have impacts on heat dissipation that can severely degrade the performance of devices and will be further expanded on in Section 5.1.3.
Table 1  Effect of surface passivation on current devoted to surface recombination at threshold.

<table>
<thead>
<tr>
<th>Treatment</th>
<th>$v_s$ (cm/s)</th>
<th>$\tau_{sr}$ (ns)</th>
<th>$I_{sr}$ (μA)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Initial</td>
<td>$2 \times 10^4$</td>
<td>0.5</td>
<td>217</td>
</tr>
<tr>
<td>(NH₄)₂S</td>
<td>$5 \times 10^3$</td>
<td>2.0</td>
<td>55</td>
</tr>
<tr>
<td>SiO₂</td>
<td>$2.6 \times 10^2$</td>
<td>38.5</td>
<td>2.82</td>
</tr>
</tbody>
</table>

This is for a coaxial resonator of outer radius $R_2 = 400 \text{ nm}$ and inner radius $R_1 = 200 \text{ nm}$. All values are assuming a threshold current density $N = 6 \times 10^{18} \text{cm}^{-3}$. The wasted current due to surface recombination is shown to reduce by $\sim 2$ orders of magnitude using the methods outlined in [75].

5.1.2 Auger recombination

Contrary to surface recombination which is a result of fabrication processes, Auger recombination in a non-radiative processes that is associated with the band structure of semiconductor materials. Here, electrons and holes collide into one another, sending each other to different energy states. Because it involves multiple electrons or holes, such a process is more detrimental when the carrier density is large. The non-radiative recombination density rate associated with Auger recombination is given as the formula below [53]:

$$R_{Aug} = CN^3$$  \hspace{1cm} (5.4)

Where C is a constant that is determined experimentally, and N is again the carrier density as before. The Auger recombination rate is next computed for the geometry of a coaxial nanolaser with radius of $R_2 = 400 \text{ nm}$ and inner radius $R_1 = 200 \text{ nm}$, and gain height of $h =$
300 nm. Again it is assumed that the threshold current density is $N_{th} = 6 \times 10^{18} \text{cm}^{-3}$. At threshold, the amount of current lost due to Auger recombination is derived and given as:

$$I_{Aug} = qC N_{th}^3 V_a,$$

(5.5)

where $V_a = 1.13 \times 10^{-13} \text{cm}^3$ is the volume of the gain region. For a $C = 8 \times 10^{-29} \text{cm}^6/\text{s}$ (bulk InGaAs)[76] and the other parameters as given before, $I_{Aug} = 313 \mu A$.

Clearly as seen from the above analysis, a low threshold gain is needed in order for room temperature operation in these devices. If a large carrier inversion is necessary, much of the current injected into the device will be funneled into non-radiative recombinations. Also from the previous work in Chapter 3, it is expected that a high Purcell Factor will significantly reduce the threshold associated with these small lasers, and a significant portion of the non-radiative recombination rate will instead be funneled into the lasing mode.

5.1.3 Heat considerations

Heat dissipation, and more importantly, heat generation are both major factors in metallic nanolaser design. Due to the miniature dimensions associated with these lasers, the current injected into such a small area brings about a large resistance, generating heat in the pillars above and below the active region. Also, as shown before, if surface recombination is not reduced, this causes a need for larger injection currents, and these injected electrons and holes both lose energy going from one state to the next, proportional to the difference in the quasi Fermi levels in each band, a value slightly larger than the bandgap, $E_g$. This can also be said to be true about Auger recombination. Lastly, the incorporation of a dielectric layer to
electrically isolate the nanolaser from the metal cladding has a blanketing effect that traps heat within the pillar, mostly allowing for heat dissipation along the InP pillars of the device.

As shown in section 5.1.1 and 5.1.2, a significant amount of current in these small devices is committed to surface recombination and to Auger recombination when considering the longer wavelength bulk InGaAs gain region. A plot of the current committed to non-radiative recombinations is shown in Fig. 16. At low carrier densities, surface recombination dominates, while as the inversion increases, the cubic factor related to Auger recombinations is the leading non-radiative recombination method. Moreover, assuming that the energy difference between the conduction and valence band is approximately 1 eV, the plot in Fig. 16 can easily be converted to energy generation inside the cavity (change the mA axis to units of mW) due to non-radiative recombination. From this it can be readily observed that heat generation in the cavities can be quite large, and heat management considerations should be incorporated into the design of the nanolasers.

![Figure 16](image)

**Figure 16** Current devoted to non-radiative recombinations as a function of the carrier density for a coaxial device with active gain geometry of outer radius $R_2 = 400 \, nm$, inner radius $R_1 = 200 \, nm$, etc.
and height of \( h = 300 \text{ nm} \). At lower carrier densities surface recombination dominates, at larger densities Auger recombination is the major contributor.

Heat dissipation in metallic nanolaser cavities is largely confined to the InP surrounding the active gain region. The electrically insulating dielectric layer has a low thermal conductivity compared to InP (\( \sim 1 - 6 \frac{W}{mk} \) compared to \( 68 \frac{W}{mk} \), respectively) and acts as a thermal shield. A coaxial structure of \( R_2: 400 \text{ nm}, R_1: 200 \text{ nm} \), and active region (InGaAs) \( h: 300 \text{ nm} \) is studied for heat dissipation. The active region is surrounded by appropriately doped InP, and the entire structure is coated by a 50 nm thick dielectric layer, which is further surrounded by silver. The structure of the remaining layers are the same as given in Table 3. Table 2 provides the thermal conductivities used in the simulation; SiO\(_2\) and Si\(_3\)N\(_4\) thermal conductivities are as found in [73], while Al\(_2\)O\(_3\) and AlN were taken from [77]. These values for thermal conductivity are lower than the values found bulk materials because of the amorphous nature of the film depositions and because of the thinness of the layers involved.

**Table 2** Thermal conductivity of thin film electrically insulated dielectrics

<table>
<thead>
<tr>
<th>Material</th>
<th>Si(_3)N(_4)</th>
<th>SiO(_2)</th>
<th>Al(_2)O(_3)</th>
<th>AlN</th>
</tr>
</thead>
<tbody>
<tr>
<td>Thermal Conductivity [W/mK]</td>
<td>0.7</td>
<td>1.1</td>
<td>2.3</td>
<td>5.6</td>
</tr>
</tbody>
</table>

The thermal conductivity of these thin films are lower than in their bulk counterparts because of the amorphous nature and the thinness of the deposited layers. This table provides the thermal conductivity of the electrically insulating thin film dielectrics.
A COMSOL FEM heat transfer simulation is carried out and shown in Fig. 17. Here, the bottom side of the structure is a heat sink kept at 295 K. Heat generation in the active region due to non-radiative recombinations and also resistivity heating of the p-doped InP is considered at a current double threshold ($350 \mu A$ threshold corresponding to $N = 6 \times 10^{18} \tau V^{-3}$). The maximum temperature is reported in the figures. In the lower thermally conductive SiN, the maximum temperature is found to be 303.5 K for a 50 nm thick shielding layer. However, as the thermal conductivity increases, more heat is able to dissipate through this layer. In the AlN a maximum temperature of 298.1 K is found. In these simulations all parameters are kept the same (current, heat generation due to non-radiative recombinations) in order to provide a 1 to 1 comparison between the shields, and the heating is based on operating at twice the threshold. In an actual structure the dielectric layers will alter the cavity mode, and causes slight changes in the quality factor and the confinement. However, in the case that the devices are fabricated imperfectly and do not have completely smooth sidewalls or vertical surfaces, the quality factor will be degraded and larger currents will be needed. Moreover, this requires even higher carrier inversions and currents. For a device that has a carrier inversion threshold of $8 \times 10^{18} cm^{-3}$ (threshold current of 1 mA) operating at twice the threshold current, heat dissipation is now a serious concern due to the larger amount of current diverted to Auger recombination. A heat transfer simulation for this scenario is shown in Fig. 18 for a coaxial nanolaser with the same dimensions as in Fig. 17. For higher heat generation the choice of insulating dielectric has a clear implication in heat dissipation, where the AlN coated nanolaser heats almost 3 times less than the SiN device.
Figure 17  Temperature distribution with varying dielectric electrically insulating shields of 50 nm thickness accounting for heat generated from non-radiative recombination and resistivity heating in the bottom p-doped pillar at a current of 700 $\mu$A. The substrate is a heatsink kept at 295 K. a) Si$_3$N$_4$ has the lowest thermal conductivity and the most heating. b) SiO$_2$. c) Al$_2$O$_3$ d) AlN. For low threshold devices the choice of the shielding dielectric is negligible is the heat dissipation process.
Figure 18  Temperature distribution, at a current of 2 mA, with varying dielectric electrically insulating shields of 50 nm thickness accounting for heat generated from non-radiative recombination and resistivity heating in the bottom p-doped pillar. The substrate is a heatsink kept at 295 K. a) Si₃N₄ has the lowest thermal conductivity and the most heating. b) SiO₂. c) Al₂O₃. d) AlN. The choice in dielectric electrical insulator has a clear effect on the maximum temperature and the higher thermal conductivity of AlN provides for the best heat dissipation.

5.2. Electrically injected fabrication

In fabricating the electrically pumped coaxial nanolasers, other successful nanolaser fabrication recipes are closely followed. The electrically pumped wafers will be ordered from
OEpic. The main differences from the optically pump fabrication process are the additions of an insulating layer and the changes to the wafer to allow for proper electrical contact formation gold. The wafer structure can be seen below in Table 3.

Table 3  The molecular beam epitaxially grown wafer structure of the electrically pumped coaxial nanolasers.

<table>
<thead>
<tr>
<th>Layer</th>
<th>Material</th>
<th>Thickness (nm)</th>
<th>Dopant</th>
</tr>
</thead>
<tbody>
<tr>
<td>8</td>
<td>InGaAs</td>
<td>125</td>
<td>Si (n~2E19 cm(^{-3}))</td>
</tr>
<tr>
<td>7</td>
<td>InP</td>
<td>235</td>
<td>Si (n~5E18 cm(^{-3}))</td>
</tr>
<tr>
<td>6</td>
<td>InP</td>
<td>235</td>
<td>Si (n~1E18 cm(^{-3}))</td>
</tr>
<tr>
<td>5</td>
<td>InGaAs</td>
<td>300</td>
<td>undoped</td>
</tr>
<tr>
<td>4</td>
<td>InP</td>
<td>125</td>
<td>Zn (p~1E18 cm(^{-3}))</td>
</tr>
<tr>
<td>3</td>
<td>InP</td>
<td>725</td>
<td>Zn (p~5E18 cm(^{-3}))</td>
</tr>
<tr>
<td>2</td>
<td>InGaAsP</td>
<td>135</td>
<td>Zn (p~2E19 cm(^{-3}))</td>
</tr>
<tr>
<td>1</td>
<td>InP</td>
<td>100</td>
<td>undoped</td>
</tr>
<tr>
<td>0</td>
<td>InP</td>
<td>substrate</td>
<td>undoped</td>
</tr>
</tbody>
</table>

100 nm of undoped InP is grown on an undoped InP substrate. A highly p-type doped InGaAsP layer of 135 nm is next grown (Zn \(p\sim2E19cm^{-3}\)). Followed by two layers of p-type InP with thickness of 725 nm and 125 nm, respectively (Zn \(p\sim5E18cm^{-3}\) and \(p\sim1E18cm^{-3}\), respectively). The gain medium is 300 nm of undoped bulk InGaAs. Two n-type layers of InP are grown on top of the gain (thickness of 235 nm each, doping of Si \(n\sim1E18cm^{-3}\) and \(n\sim5E18cm^{-3}\), respectively). Lastly, a heavily n-type doped InGaAs layer is grown (Si \(n\sim2E19cm^{-3}\)).

Figure 18 details the fabrication of the electrically injected coaxial nanolasers depicted in Fig. 14. This same process is also used in the fabrication of disk-shaped nanolasers as well. The epitaxial grown wafer (Fig. 18 a)) described above is cleaned and Flowable Oxide (FOx-16) is spun and then soft baked, resulting in an 800 nm thick film. Coaxial nanolasers with differing inner and outer radii, along with nanodisks are written by electron beam exposure. The electron beam alters the FOx-16 and turns it into a structure
similar to SiO₂. Two beakers of teramethylammonium hydroxide (TMAH) and one beaker of isopropyl alcohol (IPA) are prepared. The die is next immersed into the first beaker for 15 seconds, removed upon which the position of the tweezers is quickly changed, and then plunged into the second beaker for an additional 15 seconds. Immediately afterwards the sample is submerged into the IPA and subsequently rinsed with IPA. The FOx-16 which was exposed to the electron beam now remains and serves as a mask during the next process, reactive ion etching (Fig. 18 b)). To perform the dry etching a mixture of H₂:CH₄:Ar is used with gas proportion of 40:4:20 sccm, with RIE power of 150W at a chamber pressure of 30 mT. The sample is etched until there is only a small amount of p-type InP left (less than 300 nm). The wafer is then cleaned with oxygen plasma to remove organic contaminations and the polymers that build up during the dry etch process. A 50 sccm flow of O₂ was used with an RIE power of 150 W and a chamber pressure of 50 mT. A scanning electron microscope image of a coaxial device is provided in (Fig. 18 c)). A thin high index dielectric shield is next deposited onto the sample. NR7-3000PY photoresist is spun onto the sample and the p-contact lithography is performed. Afterwards the sample is dipped into a 1:1 solution of HCl:H₃PO₄ to fully expose the heavily p-doped InGaAsP layer. P-contact deposition follows: 5 nm of Ti and 300 nm of Au are deposited by means of a thermal evaporator, with lift off following using RR41 (Fig. 18 d)). NR7-3000PY is again spun onto the sample, soft baked for 1 min at 150°C, developed in RD6 for 2+2 seconds, flood exposed in the mask aligner, and soft baked for an additional minute at 100°C. A dry etch process using O₂ plasma is performed until the FOx-16 pillars protrude slightly from the NR7-3000PY. The Si₃N₄ shield on top of the FOx-16 is dry etched, and the sample is immersed in buffered oxide etch (BOE)
for removal of the FOx-16. Nickel, Au-Ge eutectic, and Au are next deposited to form a thin n-contact in thicknesses of 5 nm, 10 nm, and 30 nm, respectively, subsequently liftoff is performed (Fig. 18 e)). NR7-3000PY is spun and lithography performed again in order to deposit the Ag squares to form the metallic nanocavities. A thin layer of Cr is first deposited to promote adhesion (8 nm) and approximately 1300 nm of Ag is deposited to create the cavities, followed by a lift off process (Fig. 18 f)). A 3 μm layer of BCB is next spun onto the sample and cured. The BCB is dry etched until the Ag pillar on top of the cavities protrudes from the BCB (Fig. 18 g)). Following this, NR7-3000PY is again spun onto the sample to form the n-contact wires, with metallic composition of Ni/Au-Ge eutectic/Au and thickness 5 nm/150 nm/300 nm (Fig. 18 h)). The sample is then mounted to a 16 pin TO-8 header with a 3 mm hole drilled in the middle of it, and wire bonding from the TO-8 pins to the contact pads is next performed (Fig. 18 i)). It should be noted that the inclusion of the Au-Ge eutectic in the last deposition step is to provide a harder surface under the gold in order to bond the wires to the pins of the header.
Figure 19  Fabrication process of electrically injected metallic nanolasers. a) The MBE grown wafer. b) 800 nm of FOx-16 negative tone ebeam resist is spun onto the die, which is next patterned by ebeam lithography. The FOx-16 serves as a mask for the subsequent reactive ion etching. c) SEM image of a coaxial nanolaser after the RIE process. d) high-index dielectric is deposited and deposition of the p-contact (5 nm Ti, 300 nm Au). e) A thin n-contact is deposited on the top of the device (5 nm Ni, 10 nm Au-Ge eutectic, 30 nm Au). f) Formation of the silver Cavities. 8 nm of Chromium is deposited as an adhesion layer, followed by 1300 nm of Ag. g) BCB is spun, cured, and etched back until the silver pillars protrude from the BCB. h) n-contact wires are deposited, 5 nm Ni, 150 nm Au-Ge eutectic, and 300 nm Au. i) The sample is mounted to a 16 pin TO-8 header. Wire bonding from the contact pads to the header pins.
5.3. Experimental results

5.3.1 Electrically injected nanoalser characterization setup

The devices are characterized in a micro-electroluminescence setup similar to what is shown in Fig. 14 a), with the exception that the nanolasers are pulsed pumped (100 ns pulse, 50 kHz repetition rate) with a current driver (ILX Lightwave LDP-3840B). The samples are cooled to cryogenic temperatures using a liquid nitrogen cooled cryostat (Janis ST-500). Figure 20 depicts the schematic of the micro-electroluminescence characterization setup. The sample is inserted into a continuous-flow microscopy cryostat (Janis ST-500) equipped with electrical feedthroughs, and cooled with liquid nitrogen to a temperature of 77 K. The lasers are electrically pumped using a current driver (ILX Lightwave LDP-3840B) generating pulse widths of 116 ns at a repetition rate of 50 kHz. The laser diode is connected to the current source in series with a 100 Ω resistor. The average voltage across this 100 Ω resistor is measured with a voltmeter and divided by 100 Ω for the average current value. This is further divided by the duty cycle to arrive at the equivalent CW current. For proper pump shaping and finer current resolution, a circuit with only a 50 Ω resistor is connected in parallel to the laser diode and 100 Ω resistor (Fig. 21). Output power of the lasers were measured with an IR photodiode (Newport Model 818-IR). The measured output power is divided by the duty cycle for an equivalent CW power.
Figure 20  Micro-electroluminescence characterization setup. The sample is pumped with a pulsed current driver. The emission is collected by a 50x objective and directed into a monochromator with an attached InGaAs array detector, or to a cooled InGaAs camera or power-meter. An ASE source is used in conjunction with a rotating diffuser for purposes of imaging the sample surface.

Figure 21  Circuit schematic used for pulsed electrical injection. The circuit consists of a pulsed current driver and two parallel series, one with a 50 Ω resistor and the other with a 100 Ω resistor and the laser diode. The average voltage is measured across the 100 Ω resistor for current measurements.
Electrically injected nanolaser characterization

The first device under study is a coaxial metallic nanolaser with an inner silver radius of \( R_{\text{core}} = 200 \text{ nm} \), a gain ring of thickness 800 nm, an outer radius of \( R_{\text{outer}} = 1000 \text{ nm} \) (Fig. 22 a)), and also a 50 nm film of SiN providing electrical insulation. It is fabricated in the manner previously described in Section 5.2. A COMSOL FEM simulation using the Wave Optics module was carried out to find the modal content of the cavity at a temperature of 77 K (permittivities; InGaAsP: 11.56; InGaAs: 12.46; InP: 9.86; Ag: -90–i, Si\(_3\)N\(_4\): 3.92). The normal component of the electric field at a plane at the midpoint of the gain region for the mode with the largest quality factor (1066) and high mode confinement (0.612) is provided in Fig. 22 b). This mode is found at a wavelength of 1382 \( \text{nm} \). Defining the gain threshold as below in Eqn. 5.6:

\[
g_{\text{th}} = \frac{2\pi n_g}{\lambda \Gamma_{\text{E}} Q}
\]  

(5.6)

where \( n_g \) is the group velocity, \( \lambda \) is the freespace emission wavelength, \( \Gamma_{\text{E}} \) the amount of light confined to the gain region, \( Q \) the quality factor, and assume a group index of 3.2, the simulated gain threshold is \( g_{\text{th}} = 223 \text{ cm}^{-1} \). The nanolaser is next characterized at a temperature of 77 K. The spectral evolution of the emission is provided in Fig. 22 c). At a pump current of 200 \( \mu \text{A} \), the mode has clearly emerged and lasing action is occurring. The light-current curve provided in Fig. 22 b) further confirms this behavior. The temperature of the nanolaser was next elevated to 140 K by a heater inside the cryostat and lasing is also observed (Fig. 22 e) & Fig. 22 f)), but at a higher injection current.
An even smaller coaxial nanolaser is next studied. It has an inner radius of $R_{\text{core}} = 500$ nm, gain thickness of 400 nm, outer radius of $R_{\text{outer}} = 900$ nm (Fig. 23 a)), and again a SiN shield of 50 nm thickness. Figure 23 b) provides the mode profile of the normal component of the electric field for this cavity ($\lambda: 1331$ nm, $Q: 997$, and $\Gamma_E: 0.562$). The expected gain threshold is $g_{\text{th}} = 270 \, cm^{-1}$. The sample is characterized first at a temperature of 77 K. The spectral evolution and the light-current curve are depicted in Fig. 22 c) and Fig. 22 d), respectively. Due to a combination of larger gain threshold and also because the lasing mode is located so far away from the peak of the gain (1430 nm at 77 K), the threshold current in this nanolaser is larger than in the device of the previous paragraph. The temperature is next raised to 125 K and the device lases, but at a higher current (Fig. 22 e) & Fig. 22 f)). Neither laser operated at a higher temperature than at the values mentioned above. This is attributed to higher loss in the metals and also to the thermal onset of Auger recombination, which has an exponential relationship with temperature [78,79].
Figure 22 Characterization of a coaxial nanolaser a) with dimensions of $R_{\text{core}} = 200$ nm, gain thickness 800 nm, and outer radius $R_{\text{outer}} = 1000$ nm. b) the normal component of the electric field at the midpoint of the gain region. The values below are the mode's wavelength, quality factor, and energy confinement to the gain region. c) Spectral evolution and d) LI curve at a temperature of 77 K. e) & f) provide the spectral evolution and LI curve, respectively, at a temperature of 140 K.
Figure 23 Characterization of a submicron coaxial nanolaser a) dimensions of $R_{\text{core}} = 500$ nm, gain thickness 400 nm, and outer radius $R_{\text{outer}} = 900$ nm. b) the normal component of the electric field at the midpoint of the gain region. The values listed here are the associated wavelength, quality factor, and energy confinement to the gain region. c) Spectral evolution and d) LI curve at a temperature of 77 K. e) & f) provide the spectral evolution and LI curve, respectively, at a temperature of 125 K. The current threshold in this device is larger due to the higher gain threshold and also its increased distance from the peak of the gain.
CHAPTER 6: METALLIC NANOLASER ARRAYS

For many applications, the scalar treatment of the electric field of light (linear, elliptical, and circular polarizations) is adequate in describing the behavior of a system. When two resonators are brought close together into close proximity to another, these scalar properties serve well to describe the dynamics of a coupled photonic molecule. However, there are circumstances where the electric field cannot be described in such a simple fashion, and the full vectorial nature of a light needs to be considered. Such is the case of metallic cladded nanolasers, which support lasing modes featuring highly vectorial electric fields (as described in Section 2). Interestingly, despite the highly lossy metal separating them, when two such cavities are brought into close proximity to one another a coupling between the two resonators is established. This coupling gives rise to a set of supermodes that have nearly equal energy confinement factors, but vastly differing quality factors. In turn the supermodes have largely varying gain thresholds, which promotes single-mode lasing in the coupled system for the supermode of the highest quality factor. In this section, we demonstrate that through the judicious incorporation of resonator design and coupling the realization of metallic nanolaser lattices that oscillate in a single-mode and are also generators of a vector vortices.

6.1. Metallic nanolaser lattice.

It is well established that the coupling of indistinguishable components can lead to the arrangement of systems with vastly different characteristics compared to the principal element itself. For example, a single atom has altered properties in comparison to a molecule
formed by a number of the same atoms. In a similar fashion, when comparing two identical optical resonators that are coupled to one another, it could be expected that they too have characteristics differing than that of the individual constituent elements. In particular, metallic nanolasers support modes of complex and intricate vectorial fields; and when in close proximity to additional identical resonators the coupled system can exhibit new non-trivial features. For example, due to the lossy nature of the metal between cavities, a complex coupling develops. In turn, this gives rise to consequential properties, such as supermodes of differing quality factors, and even the generation of topological vortices. While investigations of coupled systems exhibiting scalar electric fields has been extensively studied, much less is known about coupled systems accompanied with vector field components. In this study, we report on the observation of a vector vortex in metallic nanolaser lattices by exploiting the loss associated with coupled of metallic cavities.

Vortices are ubiquitous in nature, revealing themselves in settings ranging from the circulation of water in a pond elegantly described by Leonardo da Vinci, to hurricanes, to even the clear blue sky. Such singularities in the sky are referred to as an optical singularity, a flourishing field in optics; singularities can arise in phase [80–84], polarization [82,85–91], or even the Poynting vector [92,93], to name a few examples. However, the creation of these vortices generally require a table top setting and large and bulky optical elements, such as quarter-wave plates, half-wave plates, polarizers, beamsplitters, spiral phase plates, dove prisms, spatial light modulators, q-plates, and metasurfaces, usually in used in some combination with one another [82,94–103]. However, direct generation is even more problematic and doing so at microchip scale levels has remained elusive due to the difficulty
of including an intracavity element to the nano/microscale resonators [83,104–106]. The method detailed in this chapter demonstrates how to generate these beams at the chip-scale level by taking advantage of a polarization rotation between adjacent elements induced by a complex coupling in a lattice of metallic nanolasers.

6.2. Coupling in metallic nanolasers

It has also been shown that some classes of metallic nanolasers are capable of generating radially and azimuthally polarized beams [30,107], revealing the vectorial nature of the field. However, although these resonators have been widely studied, there is still little known about the collective behaviors of such miniature lasers when placed in close proximity to one another.

6.2.1 7-element lattice simulation

In order to address these questions, we first consider a 7-element array of nanolasers as illustrated in Fig. 24 a). It is comprised of a central nanodisk surrounded by six additional identical elements that form a hexagonal lattice. Each cavity has a radius of 850 nm, is encapsulated by silver, and is separated from the adjacent elements via a 50 nm metallic silver gap. The gain medium consists of six InGaAsP quantum wells (overall height: 200 nm) covered by a 10 nm InP layer for protection. The top and bottom sides of the disk are terminated by a 50 nm SiO2 and a 30 nm air plug, respectively. The fabrication procedure is detailed in Section 6.3. A scanning electron microscope (SEM) of the structure before silver deposition is shown in Fig. 24 b). Figure 24 c) displays the normalized electric field and the polarization resolved intensity distribution of the principal supermode inside this 7-disk
structure at a plane located at the center of the gain region. Each peripheral cavity supports the TE\textsubscript{42} mode. The eigenfrequencies of the system are found using finite element method (FEM) (COMSOL Multiphysics 3D modules). From the simulation, it appears that despite being comprised of seven individual elements, this lattice tends to predominantly emit in a single-mode fashion. In this coupled metallic nanolaser lattice, minimizing the overlap of the field with the metallic lossy gap has important ramifications in determining the structure of the beam. In particular, one may notice that in this arrangement, the polarization of the field rotates from one element to the next in order to establish the lowest loss. This is in sharp contrast to a linear chain of coupled cavities, in which the same effect forces the array to operate in the out of phase mode.

As evident in the figure, for the dominant supermode the field inside the periphery cavities is rotated by $\varphi = 2\pi/3$ radians (Fig. 24 d) is a copy of the right most element in Fig. 24 c) with arrows included to more easily track the rotation) – and there is negligible field in the central element. Thus indicating a vector vortex with a topological index $n = 2$. A vector vortex is defined as the number of $2\pi$ rotations the polarization makes in one encirclement of the singularity [90,91,108]:

$$n \equiv \frac{1}{2\pi} \oint_{C} \nabla \theta(r) \cdot dr$$  \hspace{1cm} (6.1)

where $\theta(r)$ is the angle the polarization vector makes with the horizontal x-axis and C is a closed path around the singularity point. Furthermore, there is a strong relationship between a vector vortex and the topological charge associated with orbital angular momentum (OAM) that can be describe by the higher-order Poincaré sphere [109] (HOPS).
This sphere is similar to the more familiar Poincaré sphere describing only polarization, but in the HOPS the spins at the poles of the sphere also are coupled with opposite but equal OAMs, with the equator being a special case describing an azimuthally twisting inhomogeneous linear polarization that is undefined at the center of the beam. Moreover, the generation of a vector vortex of index \( n \) is also accompanied by the generation of OAM of \( l = \pm n \), where \( l \) is the topological charge.

Figure 24 Seven element nanoscale laser lattice. a) Illustration of the 7-element lattice. b) SEM image of the array before silver deposition. c) Normalized electric field of the lasing mode. The central element has negligible intensity. The polarization of the electric field is overlayed with the field magnitude. Each of the periphery cavities experiences a \( 2\pi/3 \) rotation with respect to its neighboring elements. d) The right most element in 1c is copied and rotated 120°. The arrows are included to more easily track the rotation.
6.2.2 2-element lattice, symmetric and anti-symmetric modes

To further elucidate on the single-mode behavior, one may revert to a simpler two-element system; a pair of coupled cavities cladded by silver, of the same dimensions as given above (850 nm radius, 50 nm gap). Due to the lifting of the degeneracy, it is expected that each resonance of the system splits into two, resulting in a symmetric mode which experiences maximal field overlap with the metal gap, and an anti-symmetric mode that minimally interacts with the lossy silver. Therefore, in this arrangement, the modes are expected to encounter a certain level of discrimination once optical gain is introduced, making the odd mode the first in line to lase by a relatively wide margin. A FEM simulation is next performed and displayed in Fig. 25, showing that the anti-symmetric mode (Fig. 25 a)) has a much larger quality factor ($Q = 1004$) than the symmetric mode (Fig. 25 b)) ($Q = 612$). This anti-symmetric supermode also has a higher quality factor than a lone isolated resonator ($Q = 766$). More details on the nature of the coupling can be found in Section 6.2.3. For characterization of the lasers, a micro-photoluminescensce setup is employed, further details on the setup are provided in Section 6.4. FEM simulations of the emission intensity profile in the Fresnel region provided in Fig. 25 c)-e) along with the observed intensity profiles in Fig. 25 f)-h), and the spectral evolution is presented in Fig. 25 i) confirming the simulation results. At a pump intensity of $0.63 \ kW/cm^2$, a single mode begins to emerge. As the pump is increased, this resonance dominates and even at pump intensities greater than times higher than the threshold, the device remains single-moded.
Figure 25  Single mode lasing in a 2-element arrangement. a) The anti-symmetric mode has minimal overlap with the lossy metal and has a higher quality factor of 1004 b) The symmetric mode supported by the system has maximum overlap with the silver. Its associated q-factor is 612 c) The simulation intensity in the Fresnel region. d) & e) Polarization resolved intensities. f) The observed emission profile of the two element system. g) & h) The observed polarization resolved intensity distribution. i, Spectral evolution. At low pump intensities many modes are observed, as the pump is further increased the lasing mode dominates and the lattice remains single-moded at pump intensities 30 times greater than the threshold.

6.2.3  Coupling mechanism between elements

Due to the high optical losses present in the silver cladding layer, the optical coupling between successive elements in our platform occurs through the air layer on top of the structure. To systematically verify this, we compared simulation results for our structure when the top layer is air vs. when the array is covered by silver. The results are summarized in Fig. 26, where the even and odd vectorial supermode profiles associated with the modes of the two-element arrangement are calculated together with their resonance frequencies using FEM methods (COMSOL Multiphysics).
In contrast to the 2-element system where the lasing modes in the two cavities have a \( \pi \) phase difference to one another, the coupling of a circular chain of \( N \) elements has a more elegant solution. Let's examine the case of \( N \) elements forming a circular lattice \([110]\) and focus on two adjacent elements such as in Fig 27. In comparison to the \( j^{th} \) element, the \( j^{th} + 1 \) element experiences a geometric rotation of \( 2\pi/N \), so that the total rotation returns to its starting point after \( N \) elements (i.e. the total geometric rotation is \( 2\pi \)). The circle in the figure is the region of interest wherein the coupling between the elements occurs and will be elaborated on further more below.
Figure 27  Two elements of an N element lattice. There is a geometric rotation between the $j^{th}$ and $j^{th} + 1$ cavity of $2\pi/N$. The circle between the elements is the location of the majority of the coupling.

We will next consider the field components inside each resonator and pay particular attention to the TE$_{nm}$ modes, since the mode of concern is a TE$_{42}$ mode. Equations 2.10-2.15 characterize each component of the electrical and magnetic field, but are listed again below in more explicit form and now describe the fields inside the $j^{th}$ element:

$$E_{r,j} = -\frac{1}{r} \frac{\partial \psi}{\partial \phi} = \frac{n}{r} J_n(k_r r) e^{ik_z z} \sin(n\phi + \phi_j) \quad (6.2)$$

$$E_{\phi,j} = \frac{\partial \psi}{\partial r} = \frac{k_r}{2} [J_{n-1}(k_r r) - J_{n+1}(k_r r)] e^{ik_z z} \cos(n\phi + \phi_j) \quad (6.3)$$

$$E_{z,j} = 0 \quad (6.4)$$

$$H_{r,j} = -\sqrt{k^2 - k_r^2} \times k_r \frac{1}{2\omega \mu_0} [J_{n-1}(k_r r) - J_{n+1}(k_r r)] e^{ik_z z} \cos(n\phi + \phi_j) \quad (6.5)$$

$$H_{\phi,j} = \frac{n}{\omega \mu_0 r} J_n(k_r r) e^{ik_z z} \sin(n\phi + \phi_j) \quad (6.6)$$

$$H_{z,j} = \frac{i k_r^2}{\omega \mu_0} J_n(k_r r) e^{ik_z z} \cos(n\phi + \phi_j) \quad (6.7)$$
After describing the field in each cavity the Poynting vector can next be written as:

\[ P_{L,j} \propto \frac{1}{2} \Re \left\{ \overrightarrow{E} \times \overrightarrow{H}^* \right\} = \frac{1}{2} \Re \left\{ \overrightarrow{E}_{av,j} \times \overrightarrow{H}_{av,j}^* \right\} = \frac{1}{2} \Re \left\{ -E_{\rho,av}H_{\phi,av}^* + E_{\phi,av}H_{\rho,av}^* \right\} \] (6.8)

Where \( \overrightarrow{E}_{av} \) and \( \overrightarrow{H}_{av} \) are the average electric and magnetic fields at the midway point between the cavities as shown in Fig. 28.

![Figure 28 Illustration demonstrating the overlapping of the fields in the array. \( \overrightarrow{E}_{av} \) and \( \overrightarrow{H}_{av} \) are average fields at the midway point between the cavities.](image)

The two addition parts of Eqn. 6.8 are next calculated separately and will later be combined to form the total Poynting vector.

\[ \frac{1}{2} \Re \{-E_{r,av}H_{\phi,av}^*\} \]

\[ = -\frac{1}{2} \sum_j \left\{ \frac{1}{2} \left[ E_{r,j}(\phi = \pi) - E_{r,j+1}(\phi = -\frac{2\pi}{N}) \right] \times \frac{1}{2} \left[ H_{\phi,j}(\phi = \pi) - H_{\phi,j+1}(\phi = -\frac{2\pi}{N}) \right] \right\} \]

\[ = -\frac{1}{2} \sum_j \left\{ \frac{1}{2} \left[ \frac{k^2-k_r^2}{\omega \mu_0 r^2} \int_0^2(k_r r) \times \frac{1}{4} \left\{ \sin(n\pi + \phi_j) - \sin\left(\frac{2\pi}{N} n + \phi_{j+1}\right) \right\} \right]\right\} \]

\[ = -P_1 \sin^2 \left[ -\frac{n\pi(N+2)}{2N} + \frac{\phi_{j+1} - \phi_j}{2} \right] \cos^2 \left[ \frac{n\pi(N-2)}{2N} + \frac{\phi_{j+1} + \phi_j}{2} \right] \] (6.9)
Here \( P_1 = \frac{1}{2} \sqrt{\frac{k^2 - k_r^2}{\omega \mu_0 r^2}} J_n^2(k_r r) \). Similarly \( \frac{1}{2} \text{Re}\{E_{\phi,av}H_{r,av}\} \) can be simplified to Eqn. 6.10:

\[
\frac{1}{2} \text{Re}\{E_{\phi,av}H_{r,av}^*\} = -P_2 \sin^2 \left[ -\frac{n\pi(N+2)}{2N} + \frac{\phi_{j+1} - \phi_j}{2} \right] \sin^2 \left[ \frac{n\pi(N-2)}{2N} + \frac{\phi_{j+1} + \phi_j}{2} \right]
\]  \( (6.10) \)

With \( P_2 = \frac{k^2_r}{4\omega \mu_0} [J_{n-1}(k_r r) - J_{n+1}(k_r r)]^2 \). Noticeably, both equations share the common term \( \sin^2 \left[ -\frac{n\pi(N+2)}{2N} + \frac{\phi_{j+1} - \phi_j}{2} \right] \). The total Poynting vector at the midpoint between adjacent cavities for an N element ring like lattice is written as:

\[
P_L \propto \sum_{j=1}^{N} \sin^2 \left[ -\frac{n\pi(N+2)}{2N} + \frac{\phi_{j+1} - \phi_j}{2} \right]
\times \left\{ P_1 \cos^2 \left[ \frac{n\pi(N-2)}{2N} + \frac{\phi_{j+1} + \phi_j}{2} \right] + P_2 \sin^2 \left[ \frac{n\pi(N-2)}{2N} + \frac{\phi_{j+1} + \phi_j}{2} \right] \right\}
\]  \( (6.11) \)

Since the metal between the two cavities is lossy, the mode that lases in the arrangement will be the configuration with the highest quality factor. The modes will experience the lowest loss when the amount of light between the cavities is at a minimum, i.e. when the Poynting vector between the cavities is at its lowest value, i.e. when Eqn. 6.12 or Eqn. 6.13 are at their minimum.

\[
\sin^2 \left[ -\frac{n\pi(N+2)}{2N} + \frac{\phi_{j+1} - \phi_j}{2} \right]
\]  \( (6.12) \)

\[
\cos^2 \left[ \frac{n\pi(N-2)}{2N} + \frac{\phi_{j+1} + \phi_j}{2} \right] + \sin^2 \left[ \frac{n\pi(N-2)}{2N} + \frac{\phi_{j+1} + \phi_j}{2} \right]
\]  \( (6.13) \)
Defining the rotation between adjacent elements as $\Delta \phi_t = \phi_{j+1} - \phi_j$, one arrives at the condition that the rotation of the field between adjacent elements will satisfy Eqn. 6.14 corresponding with Eqn. 6.12, or Eqn. 6.15, corresponding to Eqn. 6.13:

$$\Delta \phi_t = -\frac{\pi}{n} \pm \frac{2q\pi}{n} \quad (6.14)$$

$$\Delta \phi_t = \pm \frac{2\pi}{N} \pm \frac{2q'\pi}{n} \quad (6.15)$$

Where $q$ and $q'$ are separate integer numbers.

To further elucidate on the coupling in a larger lattice it is of interest to consider a six-element lattice in the shape of a hexagon, where each cavity supports a TE$_{42}$ mode. In this arrangement, the geometric rotation between adjacent elements is $\pi/3$. From Eqn. 6.14 $\Delta \phi_t = -\frac{\pi}{4} \pm \frac{2q\pi}{4} = (\frac{2q+1}{4})\pi$. However this rotation between the cavities does not satisfy the condition that the total phase change around the periphery must be equal to $2M\pi$, where $M$ is an integer number. This is similar to geometric frustration[111] seen in systems with anti-ferromagnetic spins. Figure 29 quickly illustrates the idea behind this in a simpler 3-element anti-ferromagnetic lattice, where the arrows denote either positive or negative spin. The energy of the system is at a minimum when the spins of the nearest neighbors are antiparallel. In this arrangement, the spin in the bottom right corner of the triangular lattice does not prefer to be in the up or down position, demonstrating geometric frustration. In the scenario that Eqn. 6.14 cannot be satisfied, Eqn. 6.15 is the minimum rotation between elements. In this case $\Delta \phi_t = \pm \frac{2\pi}{3} \pm \frac{2q'\pi}{n} = \pm \frac{2n\pi}{3}$, resulting in a total rotation in the polarization of $4\pi$. 
Figure 29 Geometric frustration in a 3-element antiferromagnetic arrangement. In order to minimize the energy of the system the arrows denote the spin and should point in opposite directions. However, in this particular arrangement, whichever direction the bottom right arrow points, one of its nearest neighbor will also be pointing in the same direction.

6.2.4 Characterization of 7-element lattice

Similar single-mode behaviors have also been observed in lattices with larger number of elements. Figure 30 a) depicts the spectral evolution for the 7-element lattice described above. At a pump intensity of $0.17 \, kW/cm^2$ the lasing mode emerges, as the pump is further increased, this mode dominates the spectrum and the array remains single-moded, even at pump powers exceeding 30 times the threshold. The light-light (L-L) curve associated with this device is plotted in Fig. 30 b). (green lines and circles). For comparison, a single metallic nanolaser with a radius of 850 nm is fabricated on the same wafer and its L-L curve is also plotted (purple line and diamonds) in Fig. 30 b). When considering the additional six elements and the differences between the radiation patterns, the array’s efficiency is found to be a factor of $\sim 5$ greater than a single element. To establish that the emission is associated to the mode from the FEM simulation, the observed intensity profile (Figs. 30 c)-e)) is studied
and compared to the simulated emission pattern in the Fresnel region (Figs. 30 f-h)), demonstrating excellent agreement. For further confirmation of the vector vortex, the OAM associated with the two spins are examined. As the vector vortex is of index $n = 2$, it is expected that the beam is a combination of topological charges $l = \pm 2$. In addition to the self-interference technique commonly used for determining the OAM order, we also adopt the approach based on the far-field diffraction pattern through an equilateral triangular aperture. In this latter technique, if the light carries OAM with a charge of $l$, a triangle with $|l| + 1$ spots along each side is formed, with its direction determining the sign of the charge. The emission is filtered for spin using a cascading zero-order quarter-wave plate and linear polarizer and then passed through the equilateral triangle, where a lens facilitates the far-field diffraction. The measurement is shown in Figs. 30 i) & j), where clearly $l = +2$ & $l = -2$ are shown in the figures, respectively, showing excellent agreement with the theory.
Figure 30 Characterization of the 7-element lattice vector vortex. a) Spectral evolution of the lattice. The device remains single-moded, even at pump intensities greater than 30 times the threshold. b) Comparison of the light-light curve of the 7-element lattice (green line and circles) and a single individual element (purple line and diamonds). The 7-element lattice has an output intensity ~7 times greater than the single element. The slope efficiency of the array is increased by a factor of ~5. c) The observed mode profile. d) & e), The horizontal (d)) and vertical (e)) component of the emission reveal the vectorial nature of the beam. f), g), & h), The observed emission patterns agree well with the FEM simulated intensity distribution at the Fresnel region. i) & j), Orbital angular momentum measurements. The observed far-field diffraction pattern through the triangular aperture reveal OAM's of \( l = 2 \) & \( l = -2 \), as expected from a vector vortex of topological index \( n = 2 \).
6.3. Fabrication procedure of metallic nanolaser lattice

The fabrication of metallic nanolaser lattices is depicted below in Fig. 31. The wafer is fabricated at OEpics Inc. and consists of six quantum wells of In$_{x=0.56}$Ga$_{1-x}$As$_{y=0.938}$P$_{1-y}$ (10 nm) sandwiched between two 20 nm thick barrier layers of In$_{x=0.734}$Ga$_{1-x}$As$_{y=0.57}$P$_{1-y}$ with an overall height of 200 nm, grown on a p-type InP substrate (Fig. 31 a)). The quantum wells are covered by a 10 nm thick InP over-layer for protection. Hydrogen silsesquioxane (HSQ) solution in methyl isobutyl ketone (MIBK) is used as a negative electron beam resist. A 50 nm resist is spun onto the wafer for the subsequent fabrication process, which is then soft baked (Fig. 31 b)). The arrays are written by electron beam exposure. The electron beam alters the HSQ and turns it into a structure similar to SiO$_2$. Two beakers of teramethylammonium hydroxide (TMAH) and one beaker of isopropyl alcohol (IPA) are prepared. The die is next immersed into the first beaker for 15 seconds, removed upon which the position of the tweezers is quickly changed, and then plunged into the second beaker for an additional 15 seconds. Immediately afterwards the sample is submerged into the IPA and subsequently rinsed. The HSQ which was exposed to the electron beam now remains and serves as a mask during the next process, reactive ion etching (Fig. 31 c)). To perform the dry etching a mixture of H$_2$:CH$_4$:Ar is used with gas proportion of 40:4:20 sccm, with RIE power of 150W at a chamber pressure of 30 mT. The wafer is then cleaned with oxygen plasma to remove organic contaminations and the polymers that build up during the dry etch process. A 50 sccm flow of O$_2$ was used with an RIE power of 150 W and a chamber pressure of 50 mT (Fig. 31 d)). After this, a 1000 nm layer of silver is deposited onto the sample using electron beam evaporation at a pressure of 5 $\times$ 10$^{-7}$ Torr at a rate of 0.1 Å/s for the first 400
nm, at which point the rate is ramped up to 1 Å/s over a period of 30 minutes (Fig. 31 e)). SU-8 is then used to bond the silver side to a glass substrate for mechanical support (Fig. 31 f)). The sample is baked for 3 minutes at 95°C, exposed, and then baked for 1 min at 65°C and another 2 min at 95°C. Lastly, the sample is wet etched in hydrochloric acid to remove the InP substrate (Fig. 31 g)). Scanning electron microscope (SEM) images of the two element lattice after dry etching is show in Fig. 31 h).

Figure 31 Fabrication process of a two-element metallic nanolaser lattice. This same process is used in other geometries as well. The bottom right corner provides a legend to the materials of the structure. a) Cleaned wafer with InGaAsP Quantum wells grown on an InP substrate. b) A thin layer of negative tone HSQ ebeam resist is spun onto the sample. c) The wafer is patterned by ebeam lithography and the resist is developed. d) A dry etching process is used to define the lattice. e) 1 μm Ag is deposited by means of ebeam evaporation. f) Sample is mounted and bonded to a glass microscope slide silver side down with SU-8. g) Sample is immersed into HCl to remove the InP substrate. h) SEM image of the two element lattice after dry-etching, before the silver is deposited.
6.4. Characterization setup schematic

For characterization of the lasers, a micro-photoluminescence setup is employed (Fig. 32). The samples are pumped with a 1064 nm single mode fiber laser that is focused (~30 µm pump diameter) onto the pattern using a 50x objective (N.A. of 0.42). The samples are cooled in a cryostat to a temperature of 77.4 K. The emission is collected with the same objective and directed into a linear array InGaAs detector for spectral measurements, or to an InGaAs camera to observe the emission pattern, selected via a mirror placed on a kinematic stage. An ASE source with a rotating diffuser is utilized to image and locate the sample pattern. A removable power meter is inserted into the setup for power measurements of the nanolaser lattices.
Figure 32 Characterization microphotoluminescence setup schematic. The samples are pumped with a 1064 nm fiber laser and focus onto the sample with a 50x objective (N.A. 0.42). Light is either directed to an InGaAs camera to observe emission patterns, or to a monochromator with an attached LN2 cooled InGaAs linear array detector. The emission power is collected at the location of the power meter near the monochromator. The triangular aperture used to reveal the OAM of the radiated beams is removable so as to not obstruct imaging the sample surface and to collect proper modal profiles.

6.4.1 Comparison of OAM measurement methods

In order to measure the OAM of the emission, we use the approach first proposed and demonstrated by Hickmann et al. [112], where the far-field diffraction pattern is examined after passing the beam through an equilateral triangular aperture (Fig. 33 a)). A 2-f system is installed after the triangle to take the Fourier-transform of the aperture, yielding the far-
field diffraction pattern. A quarter-wave plate and linear polarizer are used to filter for the spin handedness of the beam. If the light carries OAM, a triangle with $|l| + 1$ spots along each side is formed (Fig. 33 b-e)), the direction that the pattern is pointing denotes if the OAM is positive or negative. For example, an $l = +1$ results in a triangle with two sides, pointing to the right. A similar pattern is observed for $l = -1$, but instead it points to the left. The same trend is observed for $|l| = 2$, but in this case there are three spots formed along each side of the diffraction pattern. We then characterized a vector vortex of order $n = 2$, and find that when filtering for left/right circular polarizations, find both $l = 2$ & $l = -2$, as expected (Fig. 33 f & g)).
Figure 33  Zoomed OAM measurement setup schematic. a) The far-field diffraction pattern passed through a triangular aperture reveals the topological charge \( l \) associated with a given beam. b)-e), The triangle will have \(|l| + 1\) lobes on each side, while the direction provides an assessment of the sign of the charge (left is negative and right is positive). f),g) A vector vortex of charge \( n = -2 \) is passed through the triangular aperture and filtered for the left/right circular polarizations. OAM values of \( l = +2 \) & \( l = -2 \) were detected, as expected.

In addition to the OAM measurements recorded using the triangular aperture technique presented in this paper, measurements were also recorded using a self-interference technique. A Mach-Zander interferometer was constructed (Fig. 34 a)) and
operated in a quasi-parallel fashion and imaged onto an InGaAs IR camera. The same beam as in the triangle measurement \((n = -2)\) supporting a mode with an OAM of \(l = +2\) & \(l = -2\) is used. The forked interference pattern is shown in Fig. 34 b) & Fig. 34 c), clearly showing a \(|l| = 2\) topological charge. When the other spin is filtered for, the direction of the forks in Fig. 34 b) flip in Fig. 34 c), indicating the OAM has the same magnitude but a different sign. The advantage to the triangular aperture approach is that the topological magnitude and the sign of the charge can quickly be assessed.

Figure 34  Self-interference technique. a) A schematic of the quasi-parallel Mach-Zander interferometer used in the self-interference measurement. b),c), The same beam used in Fig. 28 is tested. The self-interference clearly shows the fork pattern indicating a topological charge of \(|l| = 2\). In b) & c) the spin polarizations are being filtered for and the fork patterns split in opposite directions, denoting the change in sign.
6.5. Conclusion

In conclusion, we have observed a vector vortex in metallic nanolaser lattices. Due to the lossy nature of the coupling between the cavities, the lattices operate in a single-mode. Additionally, the vector vortex index can be modified by altering the geometry and size of the elements of the lattice, so that different modes are resonant in the system. We noticed that such arrays generally exhibit higher slope efficiencies in comparison to their individual constituent lasers. Importantly, these lattices could help pave the way for a new generation of nanoscale light sources that can realize arbitrary emission properties such as polarization patterns and topological protection.
CHAPTER 7: CONCLUDING REMARKS

In this work, the fundamental properties of metallic nanolasers were explored. Due to the use of a metallic cladding their size is capable of being reduced to subwavelength dimensions. This also leads to cavity enhancing effects such as a large Purcell factor and a large spontaneous emission coupling ratio. Due to this higher $\beta$-factor and the lossy metallic cladding, there had even been a argument as to whether or not these nanolasers were even capable of lasing. To settle this debate we fabricated and tested various metallic coaxial and disk shaped nanolasers that had an InGaAsP quantum well gain medium. The devices were optically pumped and the emission was assessed via a modified Hanbury Brown and Twiss interferometer in order to characterize the second-order coherence ($g^2(\tau)$) of the radiation. The results of this experiment demonstrated that these miniature sources were undoubtedly capable of lasing.

The prospects of these devices operating under electrical injection were also examined. Due to large modal confinement, and in turn high Purcell factor and spontaneous emission coupling factor, and additionally the enhancement of the stimulated emission by the Purcell effect, low thresholds (μA) and large direct modulation bandwidths (100’s GHz) are predicted. Moreover, design considerations and demonstration of electrically injected metallic nanolasers operating at cryogenic temperatures were shown.

Lastly, the collective properties of coupled metallic nanolaser lattices were investigated. Due to the vectorial nature of the modes inside these cavities and the lossy
metal cladding, a complex coupling occurs in which the fields inside the cavities orient themselves to minimize the loss. In a pair of cavities this results in a supermode where the fields are $\pi$ out of phase between the two resonators. However, if the size of the array increases and forms a closed ring like shape, a more elegant coupling occurs that accounts for geometrical rotations and minimization of losses, and can lead to the generation of vector vortices in coupled metallic nanolaser lattices.
REFERENCES


