

COLLOIDAL QUANTUM WELL LIGHT-EMITTING WAVEGUIDES

A THESIS SUBMITTED TO
THE GRADUATE SCHOOL OF ENGINEERING AND SCIENCE
OF BILKENT UNIVERSITY
IN PARTIAL FULFILLMENT OF THE REQUIREMENTS FOR
THE DEGREE OF
MASTER OF SCIENCE
IN
ELECTRICAL AND ELECTRONICS ENGINEERING

By
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July 2023

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We certify that we have read this thesis and that in our opinion it is fully adequate, in scope and in quality, as a thesis for degree of Master of Science.

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ABSTRACT

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M.S. in Electrical and Electronics Engineering

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July 2023

Micro/nanoscale semiconductor light-emitting devices of colloidal nanocrystals offer low-cost solutions while delivering high performance in ambient lighting systems, displays, and photonic circuits. Colloidal quantum wells (CQWs) are excellent candidates as active materials for these optoelectronic devices owing to their superior properties including suppressed Auger recombination, large absorption cross-section, and narrow emission linewidth. In this thesis, as our first study, we proposed and demonstrated dual-color lasing using heterostructures of CQWs as the gain media in an all-solution-processed dual-color optical cavity for the first time. Here, a multilayered waveguide architecture consisting of green- and red-emitting CQWs, separated with a transparent low refractive index colloidal spacing layer of silica nanoparticles (NPs) suppressing otherwise detrimental nonradiative energy transfer between them, enabled amplified spontaneous emission (ASE) simultaneously in two colors at the threshold level of $\sim 17 \mu\text{J}/\text{cm}^2$. We further adapted this multilayer waveguide configuration to a whispering-gallery-mode (WGM) cavity by fabricating a microdisk structure directly out of these layered CQWs-NPs-CQWs colloids. The resulting device showed dual-color multimode lasing both at 569 and 648 nm at the same time with the threshold of $\sim 106 \mu\text{J}/\text{cm}^2$. Then, as the second study of this thesis, we developed a colloidal waveguide light-emitting diode (LED) structure of CQWs that changes the direction of light from the surface to the edge of the device by combining the active CQW region with a slit-shaped waveguide architecture that confines the light within the emissive layer and guides it through the lateral axis. Driving this LED waveguide of $900 \mu\text{m}$ in length by $150 \mu\text{m}$ in width at a current density level of $5.6 \text{ A}/\text{cm}^2$, we observed the output emission reached a luminance level of $\sim 20,400 \text{ cd}/\text{m}^2$. These unique waveguiding architectures integrated into the light-emitting devices of CQWs hold great promise for on-chip photonic applications including

CQW dual-color excitation for biological imaging and CQW LED-based photonic integrated circuits.

Keywords: Colloidal waveguides, colloidal quantum wells, colloidal light-emitting devices, optical gain, dual-color lasers, edge-emitting light-emitting diodes.



ÖZET

KOLOİDAL KUANTUM KUYULARI TABANLI IŞIK YAYAN DALGA KILAVUZLARI

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Elektrik ve Elektronik Mühendisliği, Yüksek Lisans

Tez Danışmanı: Hilmi Volkan Demir

Temmuz 2023

Koloidal nanokristallerle oluşturulmuş mikro/nano ölçekli yarıiletken ışık yayan cihazlar, ortam aydınlatma sistemlerinde, görüntüleme uygulamalarında ve fotonik devrelerde yüksek performans için düşük maliyetli çözümler sunmaktadır. Koloidal kuantum kuyuları (KKK'ler), bastırılmış Auger eksiton birleşimi, geniş soğurma kesiti ve dar ışıma çizgi genişliği gibi üstün özellikleri nedeniyle bu optoelektronik cihazlar için mükemmel aktif malzeme adaylardır. Bu tez kapsamında, ilk çalışma olarak, tümü çözeltilde gerçekleştirilen çift renkli bir optik çınlaçta kazanç ortamı olarak KKK'lerin heteroyapılarını kullanarak çift renkli lazer ışıması gösterdik. Burada, yeşil ve kırmızı renklerde ışıyan KKK'lerin, aralarındaki aksi takdirde zararlı ısınımsal olmayan enerji transferini bastırmak için silika nanoparçacıklar (NP'ler) ile oluşturulan şeffaf, düşük kırılma indeksli koloidal açıklık katmanı ile ayrılmasıyla oluşan çok katmanlı bir dalga kılavuzu mimarisi, aynı anda iki renkte $\sim 17 \mu\text{J}/\text{cm}^2$ eşik seviyesine sahip kendiliğinden ışıma yükseltilmesi (KIY) sağladı. Bu çok katmanlı dalga kılavuzu konfigürasyonunu, doğrudan bu katmanlı KKK-NP-KKK koloidlerinden bir mikrodisk yapısı üreterek fısıldayan-galeri-kip (FGK) çınlacına uyarladık. Ortaya çıkan aygıt, aynı anda $\sim 106 \mu\text{J}/\text{cm}^2$ eşik seviyesiyle 569 ve 648 nm'de çift renkli çok modlu lazer ışıması gösterdi. Daha sonra, bu tez kapasımdaki ikinci çalışma olarak, aktif KKK bölgesini, ışığı yayıcı katman içine hapseden ve yanıl eksen boyunca yönlendiren yarık biçimli bir dalga kılavuzu mimarisiyle birleştirerek, ışığın yönünü cihazın yüzeyinden kenarına değıştiren KKK'lerden oluşan bir koloidal dalga kılavuzlu ışık yayan diyot (LED) yapısı geliştirdik. Bu 900 μm uzunluğunda ve 150 μm genişliğindeki dalga kılavuzu şeklindeki LED'e 5,6 A/cm²'lik bir akım yoğunluğu sürüldüğünde, çıkış ışıma $\sim 20.400 \text{ cd}/\text{m}^2$ 'lik bir parlaklık seviyesi gösterdi. KKK tabanlı ışık yayan cihazlara tümleştirelebilen bu benzersiz dalga kılavuzu mimarileri, biyolojik görüntüleme için KKK tabanlı çift renkli uyarma

ve KKK kullanılan LED tabanlı fotonik tümleşik devreler gibi ip st fotonik uygulamalar iin byk umut vaat etmektedir.

Anahtar szckler: Koloidal dalga kılavuzları, koloidal kuantum kuyuları, koloidal ışık yayan aygıtlar, optik kazanç, ift renk lazerler, kenardan ışıyan ışık yayan diyotlar.



Acknowledgement

I am sincerely grateful to my advisor, Prof. Hilmi Volkan Demir, for his unwavering support, invaluable guidance, and profound vision throughout this thesis. Working in his research group has been an incredible learning experience.

I would also like to thank my jury members Asst. Prof. Erdinç Tatar and Prof. Evren Mutlugün for their valuable comments and suggestions.

I would like to sincerely thank all the past and present members of Demir Research Group. In particular I want to thank Özgün Akyüz, Emre Ünal, Dr. Savaş Delikanlı, Dr. Tatiana Lastovina, Dr. Mohammed A. Ibrahim, Dr. Zeynep Dikmen, Dr. Aysan Khaligh Vazirabadi, Rabia Önen, Farzan Shabani, Furkan Işık, Taylan Bozkaya, Taha Haddadifam, Rumon Miah, Tuğba Karahan, Anıl Çağrı Atak, Furkan Şahin, Sobhan Gholami, Mohsin Waris, Tevfik Bülent Kanmaz, and Hakan Akgün for being genuine friends and excellent co-workers.

I would like to give special thanks to the LED Team, Dr. Betül Canımkbey, İklim Bozkaya, and İlayda Özkan. Their intimate friendship and endless support have always enlightened my path during these challenging studies.

Finally, I want to express the deepest love to my mother, my father, and my siblings for their selfless support in every conditions and circumstances. Their unwavering presence and encouragement have been invaluable to me, and I am forever grateful for their love and support.

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Chapter 1

Introduction

Light is the fundamental source of vision that enables humans to perceive the universe and collect visual information about their surroundings, from detecting nearby objects to observing distant planets and microscale entities. For thousands of years, stars and fire were the primary illumination source for mankind. The first artificial light generation was achieved by Humphry Davy in the early 19th century by passing an electrical current through a carbon piece [1]. Since then, for two hundred years, continuous progress in artificial light sources has been demonstrated. During these efforts, several materials, such as electroluminescent phosphors [2], organic dyes [3], and rare earth elements [4], were utilized for various light generation applications, including ambient lighting, display systems, and solid-state lasers. However, these systems usually suffer from inefficiency, shorter lifespan, and limited color control.

Integration of semiconductors into light generation systems during the second half of the 20th century enabled to obtain low-cost, high-performance light-emitting devices with sensitive color tuning features. Owing to semiconductors' direct bandgap transition, higher emission efficiencies are reached. Since the energy bandgap can be engineered by tailoring the size and composition of the semiconductor, precise color tuning has been possible. Besides, thanks to the ability of compact-size fabrication, semiconductor-based devices present design flexibility as well as low production costs. Having the abovementioned advantages, semiconductors, including III-V compounds [5], organic molecules [6], perovskites [7], and colloidal nanocrystals (NCs) [8], have been widely utilized for various light-emitting devices as active materials.

During many years of research, several semiconductor-based light-emitting devices with different functions were demonstrated. For example, semiconductors are commonly used in luminescent solar concentrators (LSCs) [9], which are designed to increase the efficiency of solar energy conversion by absorbing the sunlight and guiding its concentrated emission to the solar cell. Optically pumped lasers based on semiconductor materials, which receive the excitation energy from an external light source and generate coherent, monochromatic, and directional light by optical amplification, are widely found [10]. Besides, the semiconductors

are the essential materials as the active layer of light-emitting diodes (LEDs) and laser diodes that generate light by electrical excitation [11].

Among the semiconductors widely employed for light-emitting applications, colloidal NCs have attracted great interest due to their unique electronic and optical properties. Thanks to the controllable quantum confinement effect, spectral features of the colloidal NCs can be manipulated by precisely adjusting the size, shape, and composition. Also, low production costs and solution processability make colloidal NCs excellent candidates for light-emitting devices in large-scale manufacturing [12]. A class of colloidal NCs having a quasi-two-dimensional shape is called colloidal quantum wells (CQWs). Compared to their zero- and one-dimensional counterparts, these specific nanocrystals offer a narrower emission bandwidth owing to their suppressed inhomogeneous broadening and more suppressed Auger recombination thanks to their non-confined lateral dimensions [13]. Therefore, CQWs present great potential as active semiconductor materials for light-emitting devices.

In this thesis, we designed, fabricated, and characterized two types of CQWs-based light-emitting devices integrated into optical waveguide architectures to control the emission properties of devices. The first device we presented is an optically-pumped laser in a waveguide that simultaneously exhibits lasing in two colors. For this purpose, we combined red- and green-emitting CQWs into a single multi-layered waveguide showing efficient optical gain properties with a cavity structure consisting of a microdisk. In addition to our dual-color laser, we demonstrated a CQW LED structure integrated into a waveguide slit to obtain electroluminescence emission from the device edge. We sandwiched the CQWs-based emissive layer between two reflective metals so that the generated light was confined within the waveguide, and the surface-emission was not allowed. Thus, the findings of this thesis indicate that CQWs have an excellent capability of utilization with nano- and micro-scale state-of-the-art photonic architectures to develop several types of waveguide-based light-emitting devices.

The rest of the thesis is organized as follows: In Chapter 2, we present scientific information about the theory of materials, techniques, and devices utilized in this thesis. Chapter 3 explains the details of the experimental methods applied for fabrication and characterization processes. In chapter 4, we demonstrate dual-color ASE and lasing using multilayered waveguide and microdisk architectures. In Chapter 5, we propose and exhibit an LED architecture of CQWs from a slit-shaped active waveguide structure that directs the emission to the device edge. Finally, Chapter 6 concludes this thesis and gives an outlook for the future studies.

Chapter 2

Scientific Background

2.1 Colloidal Semiconductor Nanocrystals

In this section, we explain the fundamental properties of colloidal nanocrystals by giving brief information about the quantum confinement theory, heterostructures of colloidal quantum wells (CQWs), their nonradiative energy transfer mechanism, and the optical gain structures.

2.1.1 Quantum Confinement Effect

Colloidal nanocrystals, which are composed of inorganic semiconductor materials, feature unique optical and electronic properties thanks to their small sizes, typically measured in nanometers. Shrinking semiconductors into nanoscale dimensions causes the quantum confinement of electron-hole pairs inside them. In a colloidal nanocrystal, the quantum confinement effect directly modifies the absorption and emission spectra, radiative and nonradiative recombination rates, and relaxation time [14].

In the case of bulk semiconductor material, a large number of atoms crowd together and significantly increase the number of energy states inside the semiconductor. This substantial rise eventually causes the formation of a continuous-like band of states. However, when the size of the particle becomes smaller and its size approaches the exciton Bohr radius, defined as the physical distance between the electron and the hole in an exciton, spatial confinement occurs, and the energy states separate from each other [15]. As shown in Figure 2.1, the smaller nanoparticles have a larger bandgap, which causes a blue shift in the emission spectra, presenting that the confinement effect has a size-tunable impact.

Colloidal nanocrystals formed by group II-VI semiconductors are available with different dimensions. The 0D variation of the materials, also called quantum dots, has an almost spherical shape and hence presents the confinement in all three dimensions. Similarly, 1D nanorods and 2D nanoplatelets exhibit quantum confinement in two dimensions and one dimension, respectively (Figure 2.2). Since the size of the colloidal nanocrystals is comparable to the exciton Bohr radius, Equations 2.1 and 2.2 can be employed to calculate the confinement energy [16].

$$\Delta E \approx \frac{\hbar^2 \pi^2}{2\mu R^2} \quad (2.1)$$

$$\frac{1}{\mu} = \frac{1}{m_e} + \frac{1}{m_h} \quad (2.2)$$

where ΔE is the exciton energy shift in the states due to the confinement effect, $\hbar$ is the reduced Planck's constant, R is the radius of the crystallite, μ is the reduced exciton mass, and m_e and m_h are the effective masses of the electron and the hole.

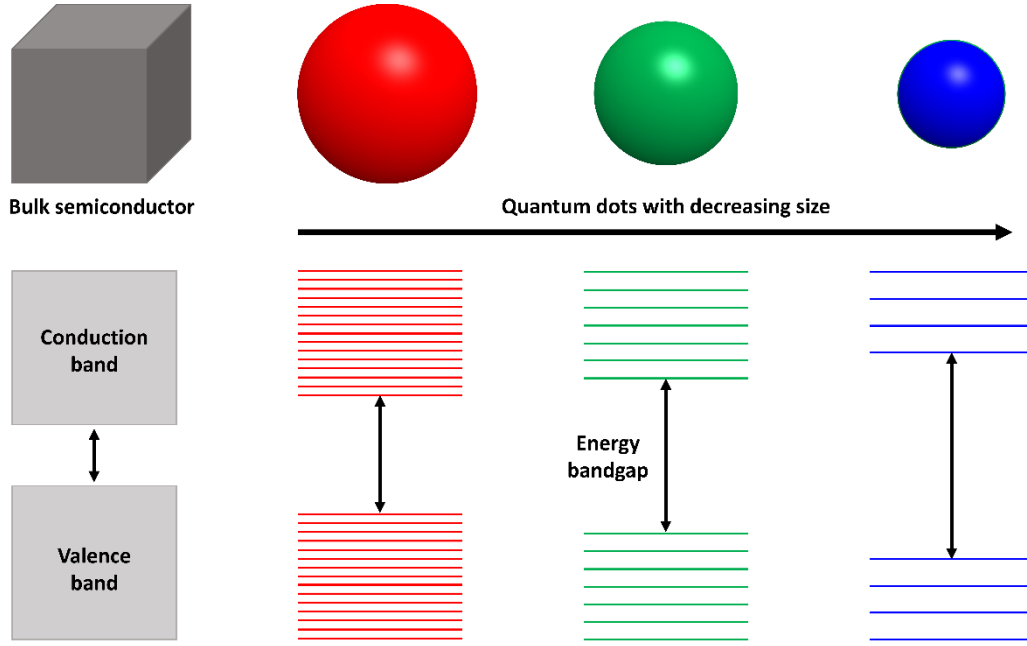


Figure 2.1: Schematic of energy band diagrams for bulk and nanocrystal structures.

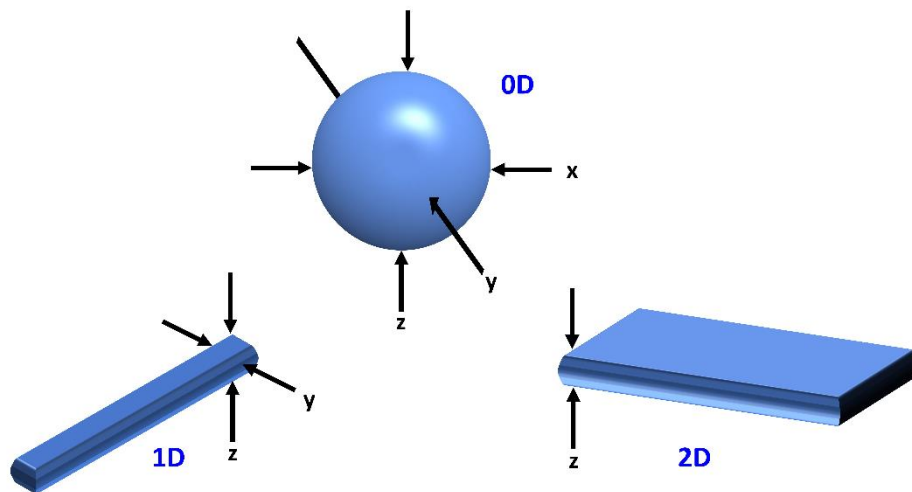


Figure 2.2: Schematics of semiconductor nanocrystals with different dimensions of quantum confinement.

2.1.2 Colloidal Quantum Wells (CQWs)

CQWs are two-dimensional semiconductor nanocrystals with relatively large lateral sizes in tens of nanometers and small vertical thicknesses of few nanometers. Due to their plate-like shapes caused by the ultrathin vertical dimension, they are also called colloidal nanoplatelets. Since their lateral sizes are larger than the exciton Bohr radius, which is typically in the range of a few nanometers, CQWs do not exhibit a quantum confinement effect in the lateral dimensions [17]. The absence of the confinement significantly suppresses the Auger recombination in the lateral dimensions and improves the optical properties by reducing the nonradiative recombinations and increasing the absorption cross-section [18].

Unlike the lateral axes, the vertical thickness of the CQWs is much smaller than the Bohr radius; hence, a strong quantum confinement is observed in one dimension. Because the thickness in the vertical axis directly affects the absorption and emission spectra, it is very critical to control the vertical size. During the synthesis of CQWs, growing the monolayers (MLs) one by one provides almost precise thickness control [19]. Since the thickness depends on the discrete number of MLs, acquiring a set of CQWs with a nearly uniform vertical size is possible via synthetic approach.

Owing to its direct effect on quantum confinement, precise control of the number of MLs ensures tuning peak wavelength position in the emission spectra. Increasing the ML number of CQWs relaxes the confinement effect inside the nanocrystals, leading to a red shift of the peak wavelength. Moreover, the uniform distribution of the ML number substantially boosts the homogenous broadening and causes much narrower linewidth in the emission spectra [20]. This sharp photoluminescence (PL) emission is characterized by a full-width-half-maximum (FWHM) of approximately 10 nm.

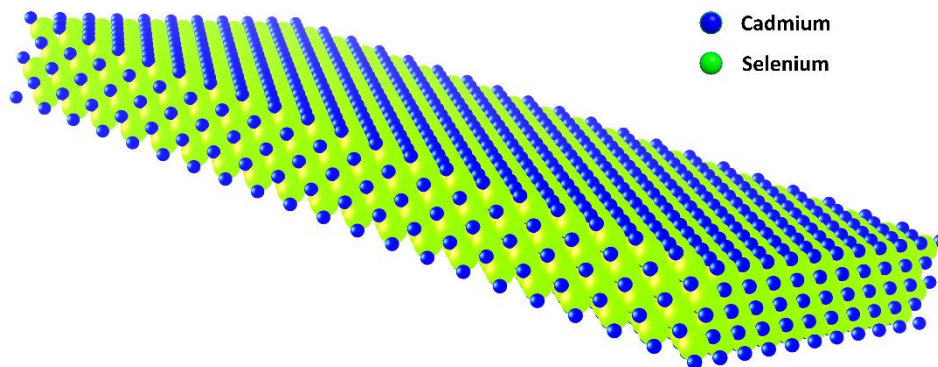


Figure 2.3: Schematic representation of 4.5ML CdSe core CQWs.

Figure 2.3 depicts a visual representation of 4.5 MLs of CdSe core CQWs. In this core architecture, the facets of the structure contain unfilled dangling bonds, which causes to increase in nonradiative recombination and a decrease in overall optical performance due to their highly energetic nature creating electron and hole traps in the surface [21]. To avoid performance degradation and further tailor the optical properties, different types of heterostructures are utilized by passivating the core with a crown, shell, or a combination of both (Figure 2.4).

Core/crown structures are built by laterally growing a crown structure around the core. As the crown does not cover the vertical axis, it does not affect the confinement. Nonetheless, because of their high photon absorption and exciton transfer capabilities, crown structures significantly improve the absorption cross-section of the CQWs [22]. Another heterostructure of CQWs is core/shell, which is constructed by coating the vertical axis of the core with a shell structure. Because changing the vertical thickness of the CQWs impacts the quantum confinement, covering the core with a shell induces a red shift in the emission spectra. Thanks to the shell growing, wherein localization of the electrons and holes in different regions of the heterostructure occurs, energy bands of the CQWs can be engineered [23].

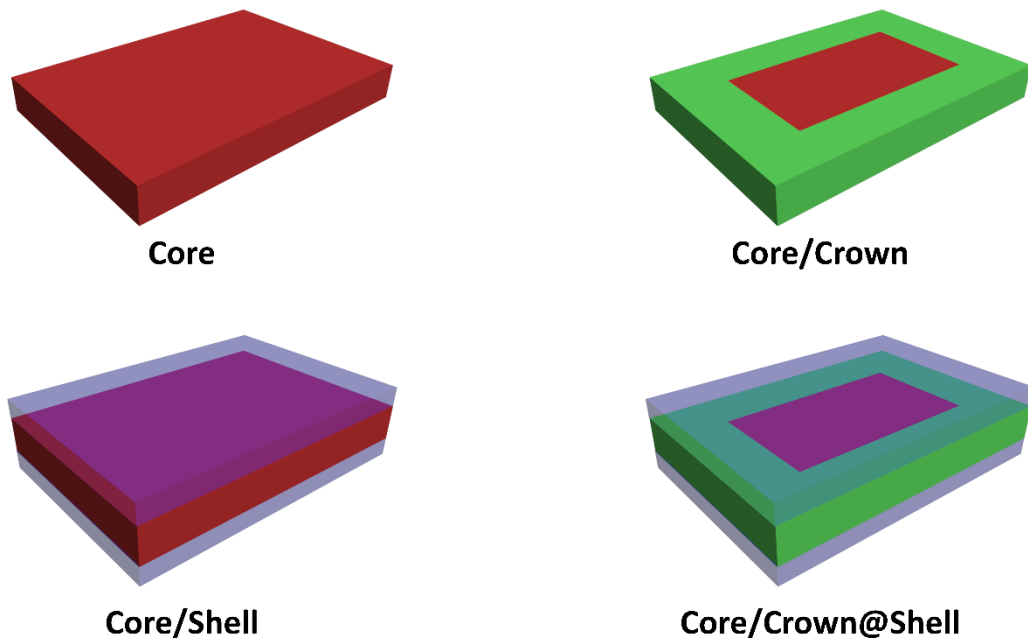


Figure 2.4: Heterostructures of CQWs: core, core/crown, core/shell, core/crown@shell.

2.1.3 Optical Gain

The fundamental mechanism of photon absorption and emission in a matter can be explained by referring to a two-level system. In such a system, consisting of a ground energy state and an excited energy state, the energy level of the atoms can be changed via the absorption or emission of photons. When the atom is at ground level, it can absorb an incident photon and be excited to the upper energy level. This process is called an upward transition. As the excited atom decays to the ground state, it emits a photon with energy that is equal to the energy difference between the two levels of the system. This is known as a downward transition, and the emission is called spontaneous emission.

In a two-level system, N_1 and N_2 are defined as densities of ground and excited energy states, respectively. If the population density difference, $N = N_2 - N_1$, is negative, the medium acts as an attenuator. However, when N is zero, the medium is optically transparent, and when N is greater than zero, population inversion occurs, and the medium behaves as an optical amplifier. Since the excited state has a higher population density, a photon in the system induces another photon to be replicated. This is defined as stimulated emission. Since the emitted photons stimulate the emission of new photons, the intensity of light traveling through the medium increases exponentially [24].

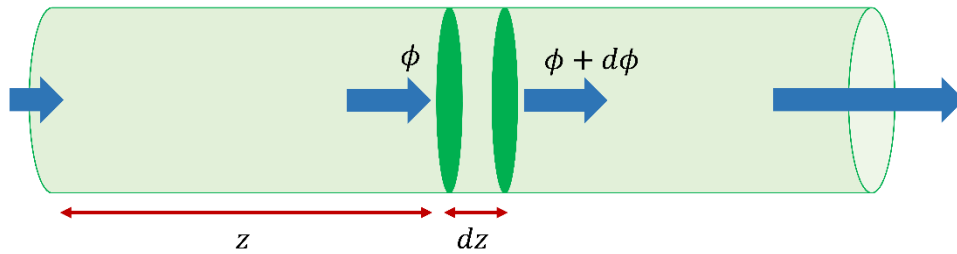


Figure 2.5: Increment of photon-flux density through the gain medium.

Figure 2.5 shows a traveling light inside a medium where population inversion exists. Photon-flux density increment per distance is calculated by

$$d\phi = (N_2 W_i - N_1 W_i) dz \quad (2.3)$$

where W_i is defined as the probability density that a ground state atom absorbs a single photon. $N_1 W_i$ is the density of photons absorbed by ground level atoms, and $N_2 W_i$ is the density of clone photons emitted by stimulated emission. W_i can also be expressed by

$$W_i = \phi \frac{\lambda^2}{8\pi t_{sp}} g(\nu) \quad (2.4)$$

where λ is the wavelength of the light travelling in the medium, t_{sp} is effective spontaneous lifetime, and $g(\nu)$ is normalized lineshape function [25].

Having a $d\phi$ greater than zero indicates the existence of gain in the medium. The net gain per unit length in the photon-flux density is defined as

$$\gamma(\nu) = N \frac{\lambda^2}{8\pi t_{sp}} g(\nu) \quad (2.5)$$

Because the light is amplified exponentially in a gain medium, the overall amplifier gain in a medium with the length d is written as

$$G(\nu) = e^{\gamma(\nu)d} \quad (2.6)$$

Figure 2.6 demonstrates photon-flux density in the medium over traveling distance.

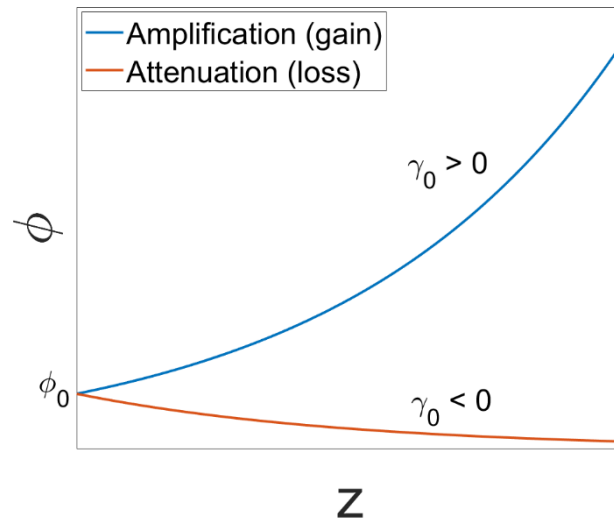


Figure 2.6: Photon-flux density along the traveling direction in gain and loss media.

Optical gain in semiconductor nanocrystals can be modeled utilizing three-level and four-level pumping schemes [26]. Diagrams of these two pumping schemes are given in Figure 2.7. For a three-level system, given the assumptions that initially induced transitions do not exist and $3 \rightarrow 1$ transitions are negligible, steady-state equations can be written as

$$\frac{dN_3}{dt} = 0 = W_p N_1 - \frac{N_3}{\tau_{32}} \quad (2.7)$$

$$\frac{dN_2}{dt} = 0 = \frac{N_3}{\tau_{32}} - \frac{N_2}{\tau_{21}} \quad (2.8)$$

where W_p is the pumping rate and τ_{ik} is the decay lifetime of transition from the i^{th} state to the k^{th} state. N_1 and N_3 are expressed in terms of N_2 as follows:

$$N_1 = \frac{N_2}{W_p \tau_{21}} \quad (2.9)$$

$$N_3 = N_2 \frac{\tau_{32}}{\tau_{21}} \quad (2.10)$$

Given that N_a is the total population density and expressed as $N_a = N_1 + N_2 + N_3$, the equation for population difference in the three-level system is given by

$$N = N_2 - N_1 = N_a \frac{W_p \tau_{21} - 1}{1 + W_p (\tau_{21} + \tau_{32})} \quad (2.11)$$

Therefore, to reach population inversion, pumping rate W_p must be greater than the rate of $2 \rightarrow 1$ transition, $\frac{1}{\tau_{21}}$.

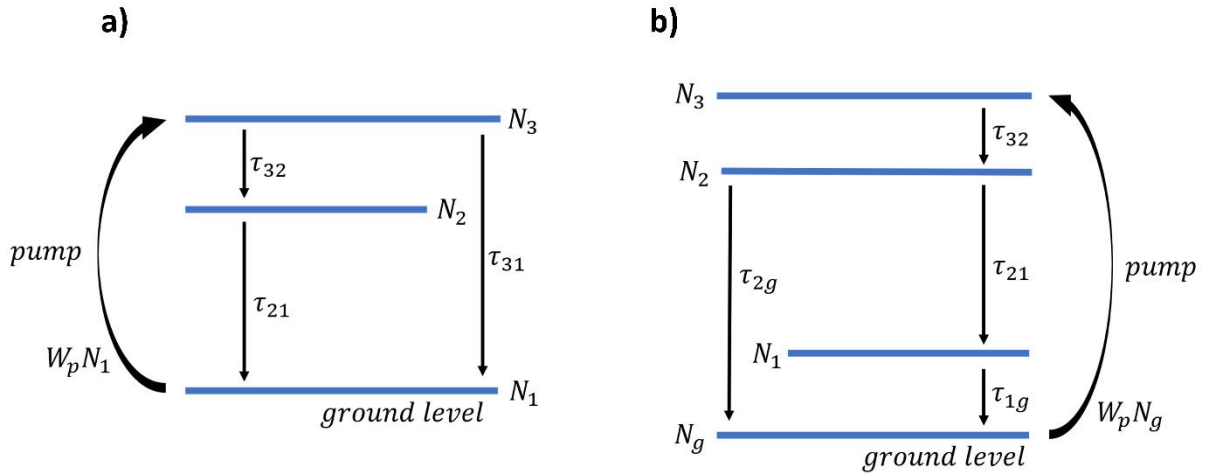


Figure 2.7: a) Three-level and b) four-level pumping schemes, including their energy levels and decay lifetimes.

Similarly, in a four-level system, steady-state equations is written as

$$\frac{dN_3}{dt} = 0 = W_p N_g - \frac{N_3}{\tau_{32}} \quad (2.12)$$

$$\frac{dN_2}{dt} = 0 = \frac{N_3}{\tau_{32}} - \frac{N_2}{\tau_{21}} \quad (2.12)$$

$$\frac{dN_1}{dt} = 0 = \frac{N_2}{\tau_{21}} - \frac{N_1}{\tau_{1g}} \quad (2.13)$$

Hence the population difference is

$$N = N_2 - N_1 = N_a \frac{W_p(\tau_{21} - \tau_{1g})}{1 + W_p(\tau_{1g} + \tau_{21} + \tau_{32})} \quad (2.14)$$

Population inversion in a four-level system is satisfied by having a higher decay lifetime of 2→1 transition than the 1→g transition.

2.1.4 Förster Resonance Energy Transfer (FRET)

FRET is a nonradiative energy transfer (NRET) process modeled and explained by dynamic dipole-dipole interactions, also known as Coulombic coupling [27]. The transfer occurs in the presence of two particles, identified as the donor and acceptor, which are in close proximity. When the donor, represented as an oscillating dipole, is excited by an external source, it induces a local electric field at the site of the acceptor. The induced field generates a transition dipole on the acceptor, and the near-field Coulombic interactions between these two resonant dipoles enable the nonradiative energy transfer from the donor to the acceptor. This nonradiative transfer, which does not involve any photon emission or absorption, causes quenching of the donor's fluorescence. Figure 2.8 illustrates the energy transfer process from the excited donor to the acceptor over a distance R that is much smaller than the wavelength of light, resulting in the acceptor's fluorescence emission.

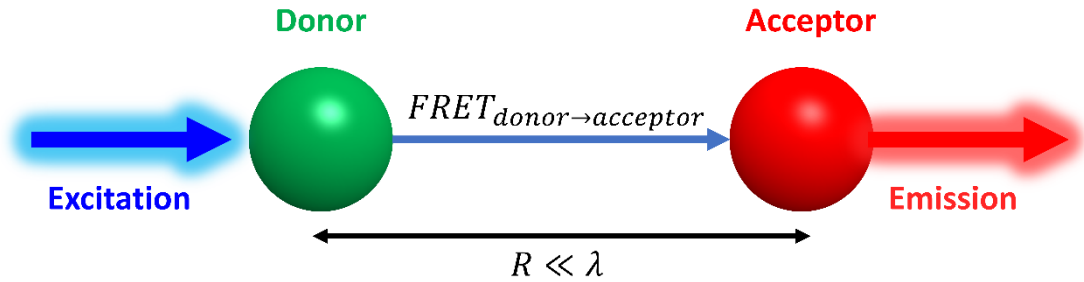


Figure 2.8: Nonradiative energy transfer from the excited donor to the emissive acceptor.

To achieve the resonant condition of energy transfer, the donor's emission spectrum and the acceptor's absorption spectrum must coincide. The rate of the energy transfer is directly proportional to this spectral overlap:

$$k_{FRET} \sim \int_0^{\infty} F_D(\lambda) \epsilon_A(\lambda) \lambda^4 d\lambda \quad (2.15)$$

where $F_D(\lambda)$ is the normalized fluorescence spectrum of the donor and $\epsilon_A(\lambda)$ is the absorption spectrum of the acceptor. Due to this convergence requirement, it can be inferred that, in colloidal systems, energy flows from the nanocrystal with a wider energy bandgap to the one with a narrower energy bandgap. No free carriers are generated during this process because FRET is an exciton transfer process, and electrons and holes are transferred as pairs [28]. Figure 2.9 shows the exciton transfer between donor and acceptor bands when FRET is performed.

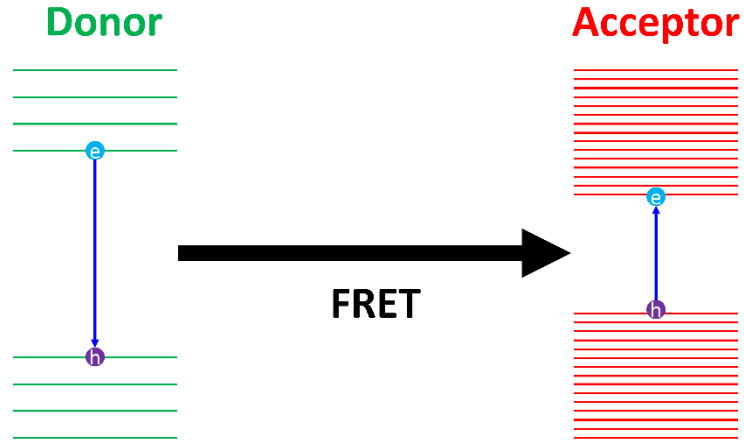


Figure 2.9: FRET process shown along with electron-hole pairs involved in the process in the band diagrams of the FRET pair: donor and acceptor

When FRET prevails, the donor emission lifetime can be expressed as follows:

$$\tau_D = \tau_r + \tau_{nr} + \tau_{FRET} \quad (2.16)$$

where τ_r , τ_{nr} , and τ_{FRET} are radiative decay, nonradiative decay, and FRET lifetimes, respectively. Besides, the efficiency of FRET is expressed as:

$$\eta_{FRET} = 1 - \frac{\tau_{DA}}{\tau_D} \quad (2.17)$$

where τ_{DA} is the donor lifetime when there is an acceptor and τ_D is the donor lifetime when there is no acceptor in the system [29].

2.2 Photonic Device Structures and Lasing

Integrating the colloidal nanocrystals with photonic structures enables the generation of highly efficient amplified spontaneous emission (ASE) and lasing. In this section, we briefly explain the waveguide and cavity structures utilized in colloidal devices and how lasing is achieved using these structures.

2.2.1 Waveguides

To transmit light over long distances, a medium with low optical loss that confines the electromagnetic wave laterally should be employed instead of using free space. Most photonic devices propagating light in extended distances use waveguides of planar dielectrics. This type of waveguide includes a high refractive index core medium and two surrounding media with a lower refractive index. The index difference causes total internal reflections (TIR) inside the waveguide and confines the light in the core during the propagation. Since the intensity of traveling light must be preserved, the attenuation constant of the dielectric materials used in the waveguide must be as small as possible.

Figure 2.10 depicts light rays' transmission, reflection, and refraction inside the waveguide. The refractive index contrast between the core and surrounding layers plays a significant role in light propagation, where TIR is particularly crucial. During the transition of light from low-index medium to high-index medium, the transmitted light angle shifts from the angle of incidence because of refraction. However, if the angle of incidence exceeds the critical angle, θ_c , TIR occurs, and all the light is reflected. The condition of TIR for the incidence angle, θ , is as follows :

$$\theta > \theta_c = \sin^{-1}\left(\frac{n_s}{n_c}\right) \quad (2.18)$$

where n_s and n_c are refractive indices of the surrounding and core media, respectively. The formula indicates that increasing the difference between the densities of the media results in TIR occurring at a smaller incidence angle [30]. Also, this leads to TIR being observed in a broader range of incidence angles, causing the number of electromagnetic modes supported by the waveguide increases.

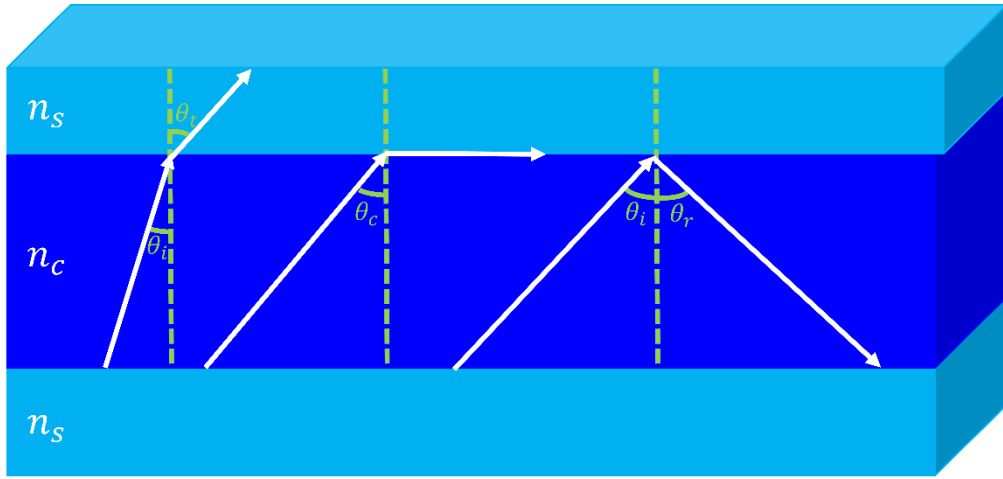


Figure 2.10: Transmitted and reflected light rays inside the waveguide.

In the nonideal case in which the waveguide includes nonnegligible optical loss, the intensity of the light is attenuated inside the medium. Depending on the attenuation constant, α_c , the light power decreases exponentially [31]:

$$P_{out} = P_{in}e^{-\alpha L} \quad (2.19)$$

where P_{in} is the power of light at the input of the waveguide, L is the distance that light travels, and P_{out} is the power of light at the position L . Figure 2.11 shows the change of electric field propagating through a lossy waveguide.

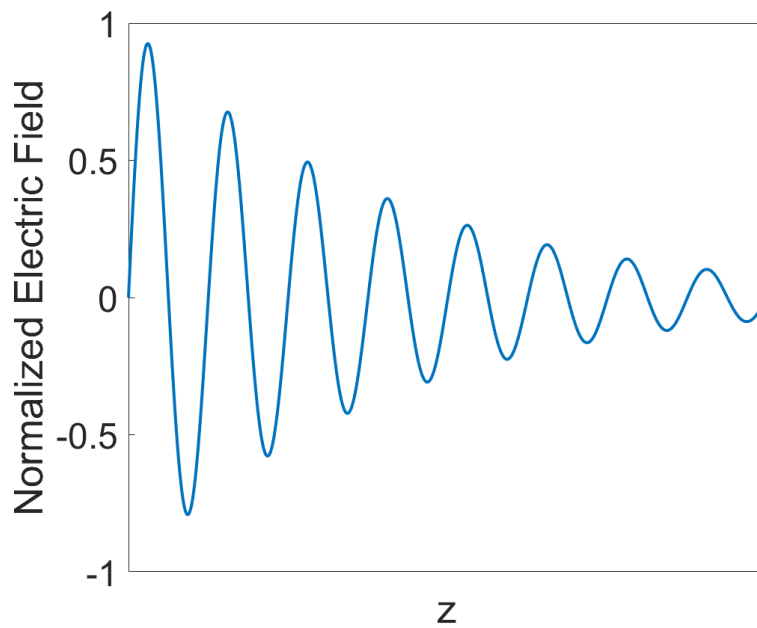


Figure 2.11: Attenuation of electric field propagating through a lossy waveguide.

2.2.2 Whispering-Gallery-Mode (WGM) Resonators

The discovery of WGM resonators dates back to the late 19th century when the confinement of acoustic waves in the galleries was studied. Due to the continuous internal reflections of sound waves from the circular walls, even the low-volume sounds generated inside a gallery can be heard from relatively long distances. The same logic applies to optical waves resonating in circularly shaped photonic structures such as microdisks, microrings, and fibers [32]. By fabricating the photonic structures using low-loss materials without surface imperfections, WGM cavities can yield resonant modes with high-Q factors.

Like planar dielectric waveguides, TIR traps the light inside the WGM-type resonators. Figure 2.12 depicts how the light travels within the microring, microdisk, and microsphere structures. The refractive index difference between the medium used inside the resonator and the medium surrounding the resonator causes TIR at the edges of the structure. When the incident light hits the edge with an angle greater than the critical angle, calculated by Equation 2.18, it reflects at the same angle and stays inside the resonator. Because of the rounded shape of the structure, the incident angle is preserved in each reflection. Therefore, the light continues to reflect from the borders of the resonator without attenuation unless the medium is lossy or contains imperfections on its surface [33].

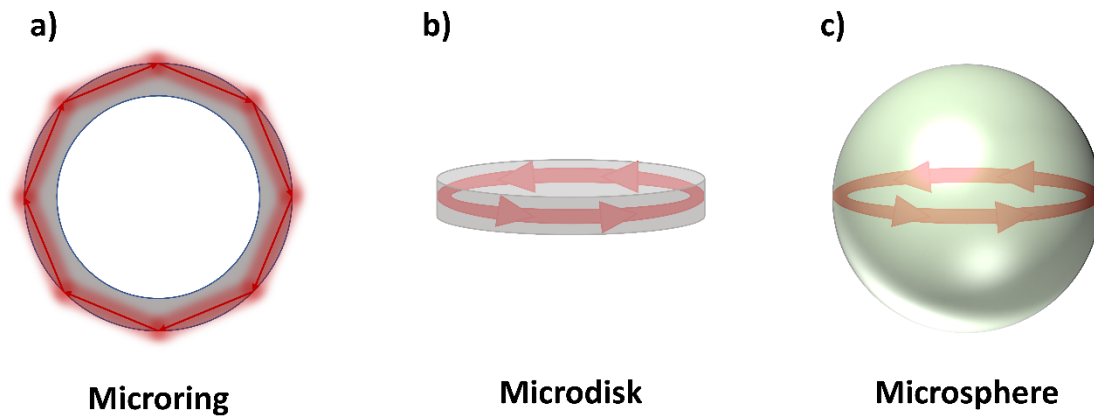


Figure 2.12: Propagation of light subjected to WGM resonance inside a) microring, b) microdisk, and c) microsphere cavity.

Depending on the size and refractive index, a WGM resonator may contain multiple modes. When the structure's diameter increases, the number of the supported resonant modes increases. The following formula finds the spectral positions of the modes:

$$m\lambda_m = 2\pi n_{eff}r \quad (2.20)$$

where λ_m is the wavelength of the m^{th} mode, n_{eff} is the effective refractive index, and r is the radius of the circular structure. Having this formula, free spectral range (FSR), which is defined as the spacing in wavelength between two successive resonant modes, is calculated by simply subtracting the wavelengths of two sequential modes from each other, i.e., $\lambda_m - \lambda_{m-1}$. Thus, increasing the size and the refractive index leads to the reduction of FSR, and hence decreases the spectral distance between the consecutive modes.

2.2.3 Amplified Spontaneous Emission (ASE) and Lasing

ASE is a crucial phenomenon in examining the optical gain properties of luminescent semiconductors. As we have already discussed in Section 2.1.3, where the optical gain mechanism of CQWs is explained, when population inversion occurs in an active medium, photon flux of the light propagating through the excited region increases continuously. Observing ASE on a semiconductor film indicates that the material is in optical gain regime and has the potential to exhibit lasing in a cavity if placed in a cavity.

To acquire a medium that photons can propagate by amplifying the light intensity, an excited waveguide structure is required. A prevalent method for building such a waveguide is placing the active layer on top of a low refractive index, transparent substrate. Since the high refractive index gain medium is sandwiched between the substrate and the air, the light is effectively confined within the medium and guided laterally. The ASE threshold of the semiconductor film is directly related to the confinement factor, defined as the ratio of the mode power coupled to the gain medium to the total power [34]. The confinement factor can also be expressed as follows:

$$\Gamma = \frac{\iint_{al_{min}}^{al_{max}} \text{Re}\{\mathbf{E} \times \mathbf{H}\} dx dy}{\iint_{-\infty}^{+\infty} \text{Re}\{\mathbf{E} \times \mathbf{H}\} dx dy} \quad (2.21)$$

where al_{max} , and al_{min} are the boundaries of the active layer, $\mathbf{E}$ is the electric field, and $\mathbf{H}$ is the magnetic field of the propagating wave. In an active waveguide, to quantify the optical performance, in addition to the material gain, modal gain, which refers to the amount of amplified light per unit length, is a critical parameter and directly proportional to the confinement factor [35]:

$$g_{modal}(\lambda) = \Gamma g_{material}(\lambda) \quad (2.22)$$

During the amplification of emission inside a gain medium, a high proportion of photons are generated by replicating the existing ones. Therefore, the substantial amount of photons that form the output emission are identical. Because the identical photons have the same direction and wavelength, ASE is remarkably directive and has a much narrower linewidth than spontaneous emission. Due to these enhanced optical properties, ASE can be considered as lasing without a cavity since the gain medium produces the photons via stimulated emission despite the absence of a cavity structure in the system.

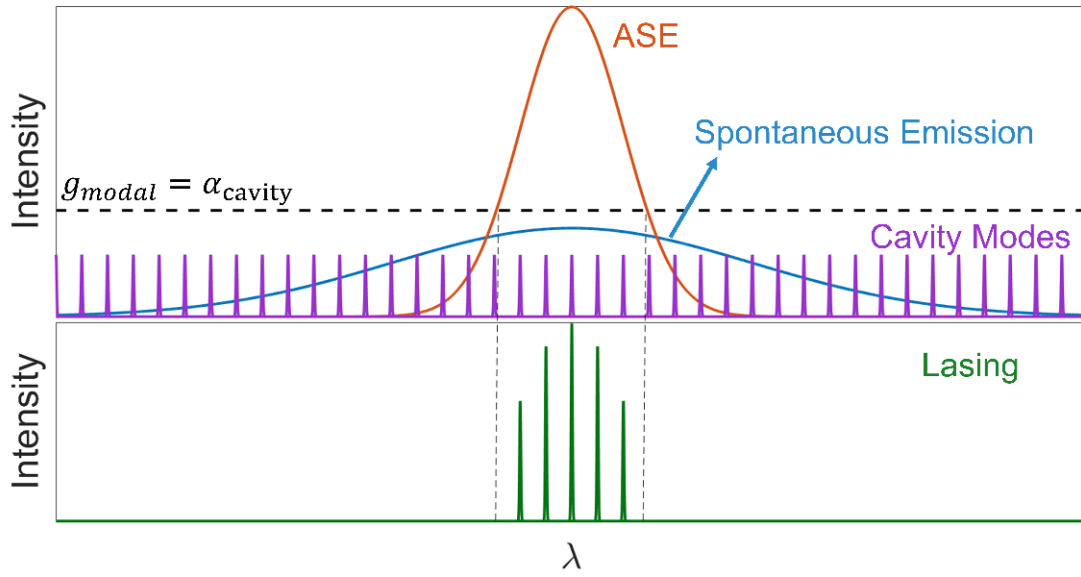


Figure 2.13: Cavity modes, spontaneous emission, ASE, and lasing spectra of a gain material inside a resonator.

Unlike waveguide structures, a cavity traps the light, leading to its resonance within the confined region. Thus, locating the gain material into a well-designed, high-quality cavity exceedingly boosts the directivity and squeezes the linewidth of the emission. The output emission of such a resonator with the gain medium is called lasing. To achieve the lasing, the modal gain should overcome the cavity losses. This condition is expressed as:

$$g_{modal} > \alpha_{reflection} + \alpha_{medium} \quad (2.23)$$

where $\alpha_{reflection}$ is the loss during the reflection of light in the boundaries of the cavity, and α_{medium} is the loss in the medium occurring during the propagation of light. Figure 2.13 illustrates the spectra of spontaneous emission, ASE, and lasing, along with the resonance modes of the cavity. It is noticed that lasing spectrum consists of only the resonance modes within the boundaries of the optical gain region that beats the losses.

To characterize the efficiency of the resonator, cavity photon lifetime, τ_c and quality factor, Q , are useful parameters and have a direct relation between each other and with the cavity losses [36]. The parameters are calculated as follows:

$$\tau_c = \frac{1}{c_n(\alpha_{reflection} + \alpha_{medium})} \quad (2.24)$$

$$Q = 2\pi\tau_c\nu_0 \quad (2.25)$$

where c_n is the speed of light in the cavity, and ν_0 is the resonance frequency. The quality factor can also be expressed as the ratio of the resonance wavelength to its FWHM:

$$Q = \frac{\lambda}{\Delta\lambda} \quad (2.26)$$

2.3 Colloidal Light-Emitting Diodes (LEDs)

Besides optical excitation, electrical pumping can achieve the emission of colloidal nanocrystals. This section explains the working principles and main components of colloidal semiconductor-based light-emitting diodes and the fundamental parameters to evaluate their electrical and optical performance.

2.3.1 Electroluminescence (EL)

EL is the process of photon generation by injecting electrical current into an emissive semiconductor. This process can also be described as the conversion of electrical energy into light. When the current passes through the emissive layer, injected holes and electrons recombine radiatively, resulting in photon emission. The energy of the photons emitted due to EL is the same as the energy difference between the electron and hole states of the emissive material. EL is the underlying phenomenon behind LEDs.

2.3.2 Working Mechanism

LEDs, in general, are the semiconductor structures that emit light based on the EL process. In the structure, the emissive layer is sandwiched between hole and electron supporting layers. Due to the $p-i-n$ architecture, current flow and light emission occur only when a forward voltage is applied. The forward voltage injects positive charges (the voids of electrons, the holes) from the hole-supporting layers and negative charges

(the electrons) from the electron-supporting layers leading to the radiative recombination and photon generation in the active region.

Various light-emitting materials can be employed as the emissive layer of LEDs, such as III-V compound semiconductors, organic polymers, perovskites, and colloidal nanocrystals. Since each of these semiconductor materials has distinct electrical properties, the functional layers used in an active device must be unique and carefully selected based on the specifics of the emissive layer. Engineering the layered architecture considering the parameters such as energy band profile and balance in charge mobilities is of critical importance for efficient light generation.

In a colloidal LED architecture, the charge direction is from the electrodes to the colloidal emissive layer through the charge injection and transport layers. Electrodes, which are conductive materials with suitable work functions, directly transmit the charges from the power source to the hole and electron injection layers. Aluminum, silver, and gold, with work functions of ~ 4.2 eV, ~ 4.5 eV, and ~ 5.3 eV, are the common metals used as electrodes. For the LED devices that emit from their surface, one of the electrodes must be transparent for light extraction. Indium tin oxide (ITO), with a work function of ~ 4.7 eV, is widely used as an electrode to transmit light through the device with negligible optical loss.

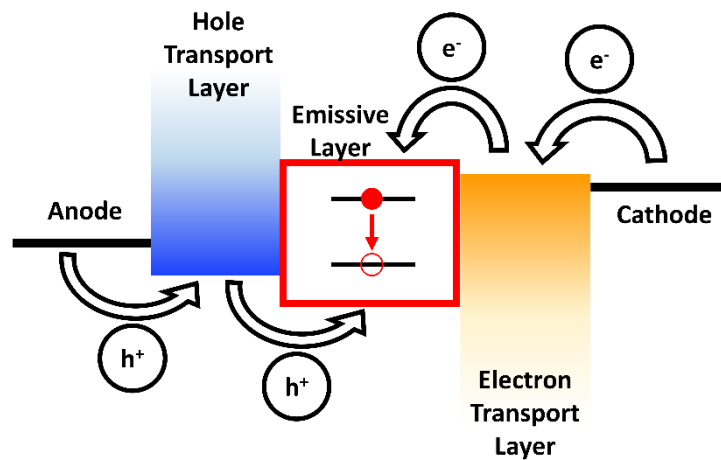


Figure 2.14: Energy band diagram of layers in an LED structure.

The hole injection layer transfers the positive charge carriers from the anode to the hole transport layer. For efficient injection, the hole injection layer must be selected in a way that its valence band edge is adjacent to both the anode's work function and the hole transport layer's valence band edge to avoid the degradation of charge transfer by the potential barrier. Similarly, the valence band edge of the hole transport layer should be close to the emissive layer's valence band edge. The same design approach is valid for selecting electron injection and transport layers, except this time, conduction bands are considered rather than the valence bands. Figure 2.14 displays the charge transfer mechanism in an LED structure.

2.3.3 LED Characterization

2.3.3.1 Current Density – Voltage (J-V) Characteristics

Current density is defined as the current passing through per unit cross-section area. During the operation of an LED device, obtaining the J-V curve provides valuable information about its electrical performance. The curve depicts the behavior of current density under an increasing forward voltage. Until a specific voltage level, called turn-on voltage, the current increment is slow. Then, passing this level, the current starts to increase in an accelerated manner. Finally, after reaching the saturation voltage, the rise in the current stops. The J-V behavior helps to understand the boundaries of the forward bias region, charge injection efficiency, and carrier concentration.

2.3.3.2 Electroluminescence and Luminance

Luminance is a parameter describing the brightness per unit area that is perceived by a human observer. To measure this quantity for an LED, the emission intensity is multiplied by the photopic function, which is the human eye's sensitivity to light with varying wavelengths [37]. The spectrum of photopic function is depicted in Figure 2.15.

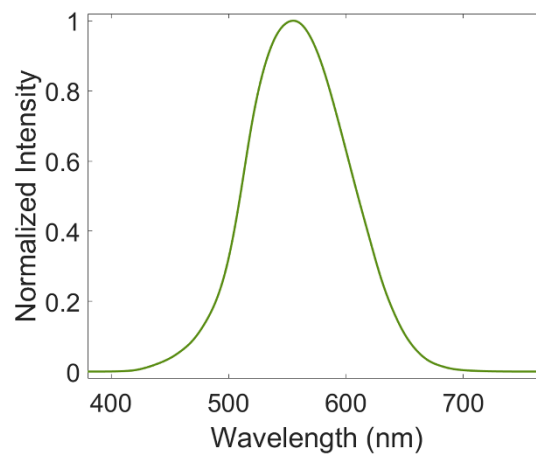


Figure 2.15: Photopic efficiency function.

2.3.3.3 External Quantum Efficiency (EQE)

EQE, defined as the ratio of extracted photons to injected electrons, is a crucial parameter to evaluate the efficiency of an LED. This quantity is calculated by multiplying two parameters, internal quantum efficiency (IQE) and light extraction efficiency. Dividing the number of photons generated inside the emissive layer by the number of electrons injected into the device, the IQE value is obtained. However, a large portion of the generated photons are trapped or lost

inside the LED structure due to the limited optical cone through which photons can be coupled from higher refractive index medium of the LED to air (constrained by the TIR at the device-air interface) and the waveguiding effects (that couple some of the generated photons into the lateral leaky modes) or optical losses, if any, introduced by the substrate and device. The ratio of the photons escaping the structure to the photons generated by the emissive layer determines the light extraction efficiency [38].



Chapter 3

Experimental Methods

3.1 Thin Film Deposition Techniques

3.1.1 Spin-Coating

Spin-coating is a widely used technique for depositing solution-processed materials as thin films onto a substrate. To utilize this method, a small amount of solution is dropped to a substrate rotating at high speed. The fast rotation produces a centrifugal force, which uniformly spreads the solution to the substrate and evaporates it rapidly. Spin-coating can be employed for the deposition of various solutions and dispersions, such as organic polymers, metallic nanoparticles and colloidal nanocrystals. It is also compatible with different substrate types, such as metals, glass, quartz, and silicon.

To control the thickness and uniformity of the films, several parameters are of great significance during the spin-coating process. For instance, the flatness of the substrate has an immediate effect on the film morphology. Nonuniformities or contaminants on the surface of the substrate directly cause defects and undesirable roughness. Another critical parameter is the speed of substrate rotation. Faster spinning speed leads to thinner and more uniform films, whereas decreasing the speed yields thicker and rougher films. However, speed is not the only variable to control the film thickness since the concentration of the solution dispersion also has a major impact. In a fixed rotation speed, the amount of materials coated onto the substrate is determined by the concentration. Therefore, incrementing the concentration increases the film thickness. Apart from the parameters mentioned above, properties of the solvent, such as viscosity, surface density, and molecular weight, must be carefully examined to reach the desired film characteristic.

The spin-coating technique presents significant advantages over alternatives for our devices owing to its simplicity, low cost, and ability to deposit uniform thin films. By adjusting the spin speed and solution concentration, we were able to fine-tune the film thickness to achieve the desired device structure. Thanks to high-speed rotation, the solution evaporates in a short time (<1 min), which reduces the process duration. Also, because the spin-coating process can be conducted at room temperature, previously deposited device layers were safe from heat-induced

damage. Therefore, we frequently preferred spin-coating for our device fabrication processes since it is a versatile technique for depositing layers from solution-processed materials.

3.1.2 Thermal Evaporation

Thermal evaporation is a physical vapor deposition (PVD) technique applied to deposit metals, metal oxides, and organic materials. To apply the technique, the target material is placed inside a conductive boat. Then the chamber is vacuumed, and a power supply is used to apply a voltage to produce current passing through the boat. The current heats the boat to the evaporation temperature of the target. The vaporized material goes up and is deposited on the surface of the substrate. Figure 3.1 depicts the schematic of the thermal evaporation process.

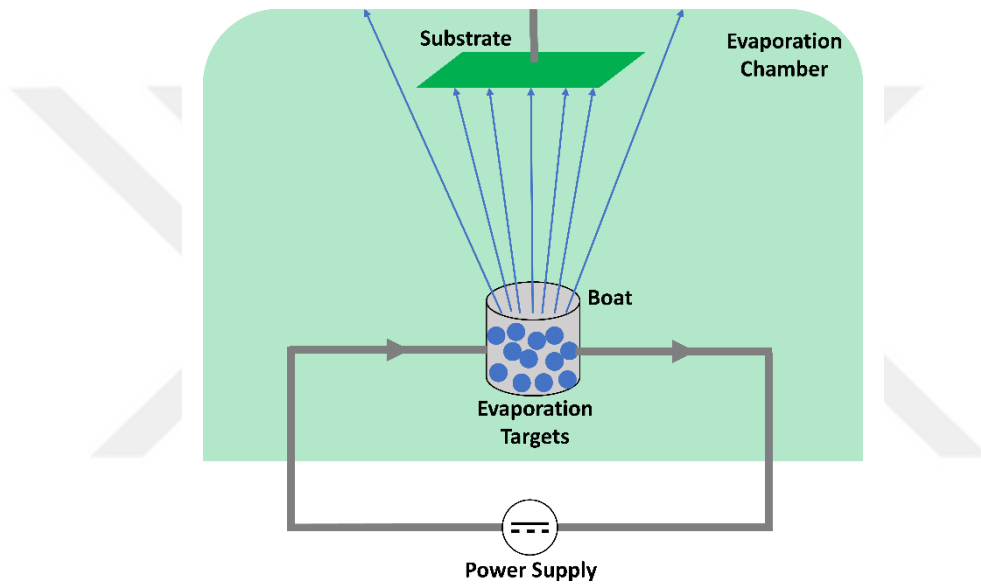


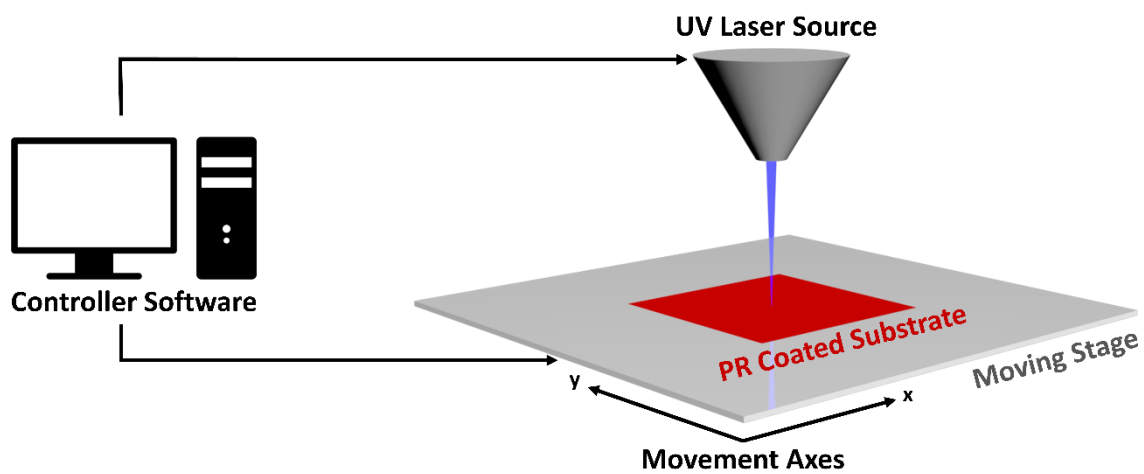
Figure 3.1: Schematical illustration of the thermal evaporation process.

The deposition rate is a critical value during evaporation. A slower process helps to achieve smoother films, while high-speed evaporation increases roughness of the film surface. To control the deposition rate, the key parameter is the target's heat. Higher temperatures cause higher rates. The heat can be tuned by changing the current flowing through the boat. Additionally, the vacuum level of the chamber significantly affects the deposition speed. Since a material's evaporation temperature depends on the chamber pressure, a higher vacuum accelerates the deposition speed. We utilized thermal evaporation (Nanovak NVTO-500) to deposit Ag, Al, MoO₃, and CBP in our work. We kept the deposition speed below 2 Å/s for all the deposition processes.

3.2 Lithography Techniques

3.2.1 Photolithography

Photolithography is a commonly used tool for producing nano/micro-scale patterned structures via UV light. The patterns can be created either by exposing the light through a patterned mask or by a laser scanning the substrate in a predefined pattern. To create patterns on our devices, we preferred the latter, which is called maskless direct laser writing, due to its ability to perform new designs without additional mask cost. For this purpose, as the direct writer, we employed Durham Magneto Optics Microwriter ML3. Figure 3.2 shows the working mechanism of a simple maskless photolithography instrument consisting of a moving stage, a UV laser source,



and controller software.

Figure 3.2: Working mechanism of direct laser writing. The software controls the stage's movement and the power of the UV laser.

The maskless photolithography process is started by spin-coating a photoresist (PR), a photosensitive organic polymer, onto a flat and cleaned substrate. To reach the target thickness, an appropriate PR type is selected, and the speed of rotation is tuned accordingly. After coating the PR, the substrate is baked on a hot plate. Baking time and temperature are determined depending on the specific PR type. The baked PR is exposed by a UV laser having a narrow beam width ($< 1 \mu\text{m}$). By moving the stage through a predefined path, the exposure light scans the substrate and generates the specific pattern on the PR.

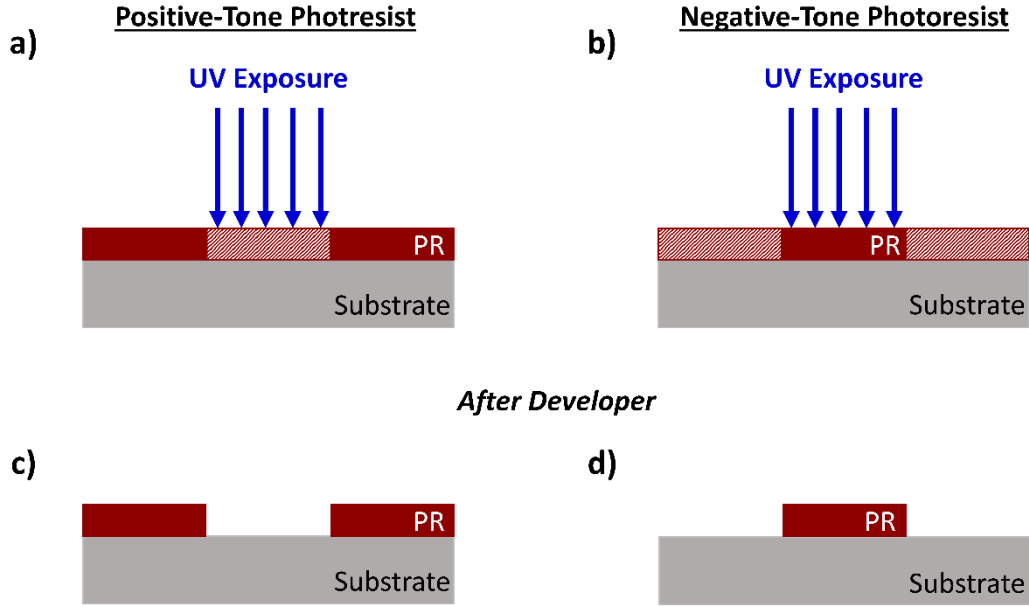


Figure 3.3: Effect of UV illumination and developer solvent on a, c) positive-tone resist, b, d) negative-tone resist.

The effect of UV exposure on the resist alternates by the selected PR type. When UV light illuminates a positive-tone PR, the exposed portion of the resist is softened and becomes soluble in the PR developer solvent. Immersing the positive resist in the developer removes the exposed area, while the unexposed portions remain intact. In the case of a negative-tone PR, which is already soluble in the developer, UV light hardens the resist and eliminates its solubility. Thus, the developer solvent purges all the resist except the exposed region. The optimal immersion time for this process depends on thickness and type of PR, developer concentration, and exposure power. Figure 3.3 depicts the behavior of positive- and negative-tone PRs after UV exposure and immersion into the developer.

3.2.2 Electron-Beam Lithography (EBL)

Similar to direct laser writing, EBL is a lithography technique that does not require a mask for pattern generation. However, unlike optical lithography, EBL utilizes a focused electron beam rather than light. Because the electron's wavelength is much smaller than the wavelength of light, the EBL method offers a greater patterning resolution. Except for the exposure source, the operational structures of the EBL and direct laser writing are analogous.

The lithography process starts with spin-coating the *polymethyl methacrylate* (PMMA), a widely used positive electron beam resist, onto a substrate. The PMMA-coated substrate is baked on a hot plate. Then the desired pattern is produced by the electron beam exposure.

Finally, the exposed regions of the PMMA are removed by its developer solvent. Spin-coating parameters, exposure time and power of electron beam, and development time must be optimized according to the concentration and molecular weight of the selected PMMA type. In this thesis, as the EBL tool, we used Raith Elphy Plus.

3.3 Characterization Techniques

This section explains the techniques that we employed to characterize colloidal quantum wells (CQWs) in various forms, including their dispersions and thin films. Absorption, photoluminescence (PL), and photoluminescence quantum yield (PLQY) spectroscopies optically characterize the in-solution CQWs. To analyze their thin film properties, optical profilometry, spectroscopic ellipsometry, and scanning electron microscopy are implemented. Also, for their light-emitting diode (LED) characterization, we conducted current density – voltage (J-V), luminance, and external quantum efficiency (EQE) measurements.

3.3.1 Absorption Spectroscopy

When incident light interacts with CQWs, a specific amount of photons at different photon energies are absorbed. The absorption rate is determined by the volume of CQWs dispersion, solution concentration, energy bands, sizes, shapes of CQWs, and the photon's wavelength. To quantify the amount of absorption for each specific wavelength, we used absorption spectroscopy. To apply this method, first, a monochromator, which selects a single wavelength from white light using a diffraction grating, directs photons into CQWs solution placed in a quartz cuvette. A photodetector collects the transmitted light and calculates the absorption. The wavelength of the incident photons changes by the rotation of the diffraction grating. Recording the absorption value for each wavelength, spectral data is generated. Figure 3.4 illustrates the schematic of the absorption spectroscopy technique.

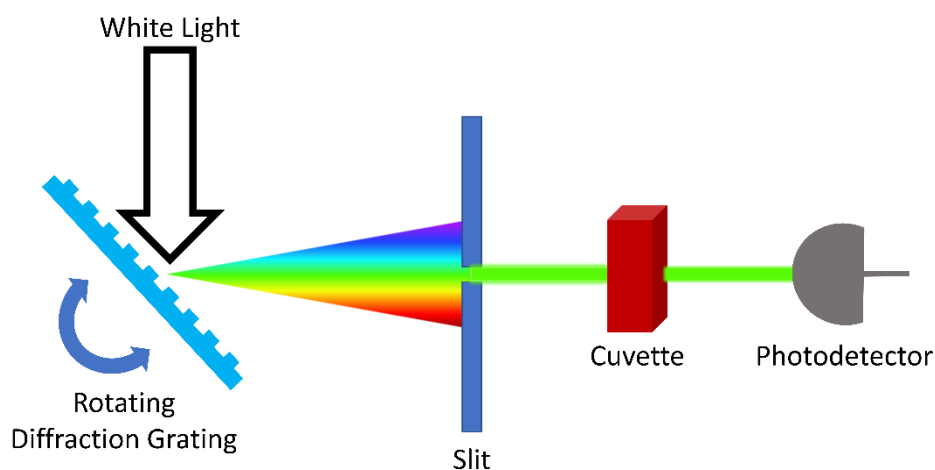


Figure 3.4: Working mechanism of absorption spectroscopy.

The absorption spectrum of CQWs presents sharp peaks at particular wavelengths referring to the electronic transitions within the nanocrystals. The position and shape of the absorption peaks in the spectrum are determined by sizes, geometries, elemental compositions, and the surrounding environment of the CQWs. Also, bandgap energy, exciton dynamics, and dispersion concentration can be understood using the information extracted from the absorption spectrum. Therefore, absorption spectroscopy is one of the most versatile methods to quantify several properties of CQWs. For these measurements, we utilized Agilent Cary 60 UV-Vis Spectrophotometer.

3.3.2 Photoluminescence (PL) Spectroscopy

An optically excited electron within the CQWs absorbs the incident photon's energy and jumps to a higher level. Relaxation of this electron leads to photon emission with various energy levels. To quantify the emission intensities at each energy level, PL spectroscopy, a widely used and well-established characterization technique, is utilized. Applying this technique requires an excitation light source within the absorption range of CQWs, a rotatable diffraction grating, a small slit, and a photodetector. The light source excites the CQWs, in the form of either a solution or thin film, and induces photon emission. The emitted photons are diffracted by the grating with varying angles depending on their wavelengths. The slit prevents all the diffracted photons from passing through except those directed to the slit's hole. Rotation of the grating changes the diffraction angle that the slit allows to pass. Thus, for each grating angle, the photons at a specific wavelength can reach the photodetector, which is located behind the slit. By rotating the grating, the photodetector measures the photon intensities across the spectrum. Quantifying the PL spectrum reveals information about the thickness, shape, and electronic properties of CQWs. Moreover, combining the data from PL spectroscopy with the absorption spectroscopy results offers a more comprehensive picture of the CQWs. To quantify the spectrum, we used Ocean Insight QE Pro spectrometer.

3.3.3 Photoluminescence Quantum Yield (PLQY) Measurements

PLQY is a crucial parameter to characterize the efficiency of CQWs. This attribute is defined as the ratio of the number of photons emitted by the excited CQWs to the number of photons absorbed by CQWs. Higher PLQY indicates efficient radiative recombination dynamics of CQWs as a large number of excited electrons release their energy radiatively. On the contrary, lower PLQY implies the existence of strong nonradiative recombination channels opened by the trap states or surface defects within the CQWs.

We characterized the efficiency of our CQWs by quantifying the absolute PLQY via a measurement setup, which is precisely calibrated based on an emissive material having a well-known PLQY value. To use this setup, first, in-solution CQWs are taken inside a quartz cuvette, and the cuvette is placed in an integrated sphere. An excitation source that emits light at a wavelength, which lies within the absorption range of CQWs but does not coincide with the spectral emission region, illuminates the sphere. The CQWs absorb a certain portion of this light, while the remaining part is collected by a fiber connected to a spectrometer (Ocean Insight QE Pro). Besides, the spectrometer detects the photons emitted by the sample. Comparing the intensities of these photon sets, which are in separate spectral regions, the PLQY value of CQWs is calculated.

3.3.4 Optical Profilometry

Optical profilometry is a convenient technique to characterize the surface topography of a film without any destruction effect. An optical profilometer includes an optical microscope, a light source, and a photodetector. The microscope focuses the light into the area where the surface profile is acquired and captures the light reflected back from the sample. The photodetector measures the intensity of the reflected light.

When a height discontinuity exists on a surface, the reflected light contains different waves with different phase values. These waves overlap with each other and create an interference pattern. Depending on the phase difference, the interference can be constructive or destructive, directly affecting the intensity of light evaluated by the photodetector. According to the detected light intensity, the system determines the phase difference and calculates the thickness difference in the measured area. Therefore, using the optical profilometry technique, step height and roughness of the surface can be assessed with high accuracy and sharp resolution. In these studies we used Zygo NewView 7200 profilometer.

3.3.5 Ellipsometry

Ellipsometry is a non-destructive optical measurement technique to characterize the thin film properties of dielectrics, semiconductors, and metals. The ellipsometer tracks the change in polarization before and after the interaction of light with the film. Since the optical and structural features of the film affect the rate of modification in the polarization, evaluation of the polarization states of transmitted and reflected light in terms of both intensity and phase provides information about the thickness, refractive index, and roughness of the film. Because the tool can precisely measure the intensity and phase in different electric field orientations, the

optical properties can be calculated with high fidelity at the sub-nanometer scale. During this characterization we employed J.A.Woollam V-VASE ellipsometer.

3.3.6 Scanning Electron Microscopy (SEM)

SEM technique is utilized to image the geometries of micro- and nano-scale structures based on electron-atom interactions. Unlike optical microscopy, which limits the imaging resolution due to the long wavelength of light (on the order of a few nanometers), SEM can produce high-resolution images with sub-nanometer precision. The tool generates electrons via an electron gun and accelerates their energies within an electric field. The accelerated electron beam is focused on the surface of a sample, and the response signals are collected by a variety of detectors depending on the types of signals. The beam and detectors scan a specific region of the sample, and an image of the scanned area is produced by analyzing the detected signals.

Different imaging techniques can be conducted through the detection and analysis of different response signal types, such as secondary electrons and backscattered electrons. Secondary electrons are generated by inelastic interaction of the primary electrons with the sample. These electrons are low energy and provide information about the surface topography of the sample. On the contrary, backscattered electrons are high-energy particles produced by elastic scattering that occur after the primary electrons hit the sample. This signal type is used to image the deeper region of the studied sample. As the SEM tool, we utilized FEI Quanta 200F.

3.3.7 Amplified Spontaneous Emission (ASE) and Lasing Characterization

As discussed in Chapter 2, ASE or lasing can be observed from CQWs depending on the optical structure they are laid down or integrated with. To distinguish these properties from the typical PL behavior, the emission of the studied samples should be monitored under the excitation of an ultra-short pulsed laser with a high pump fluence. Due to the fast Auger recombination in CQWs, femtosecond lasers are commonly applied in these characterizations. Hence, we used an advanced optical setup capable of tuning the pump laser's wavelength, amplitude, and beam size.

In the setup, the excitation source is an ultrafast laser with Ti:sapphire amplifier having 120 fs pulse width, 1 kHz repetition rate, and 800 nm operation wavelength (Spectra Physics – Spitfire Pro XP regenerative amplifier system). The intensity of the excitation laser is adjusted via a variable neutral density filter. Because the absorption of 800 nm light in CQWs is near zero and higher energy photons are required for the excitation, a barium borate crystal doubles the frequency of the laser and converts its wavelength to 400 nm. A short-pass filter in front of the BBO crystal avoids residual 800 nm light. For the ASE characterization, a cylindrical lens focuses the pump in lateral dimension to obtain a stripe of $\sim 150\ \mu\text{m}$ in width. The stripe excitation hits the sample at normal incidence, and PL emission of the structure is collected from the edge with the help of an optical fiber connected to a spectrometer. For the lasing measurement, rather than the cylindrical lens, a spherical lens with a focal length of 3 cm is used to focus the light into the laser structure. The focused laser beam must be wider than the structure so that the whole region of the structure is excited. PL emission of the laser is collected from the edge using the same setup with ASE measurements. Figure 3.5 presents the schematic of our optical measurement setup used in the lasing characterization.

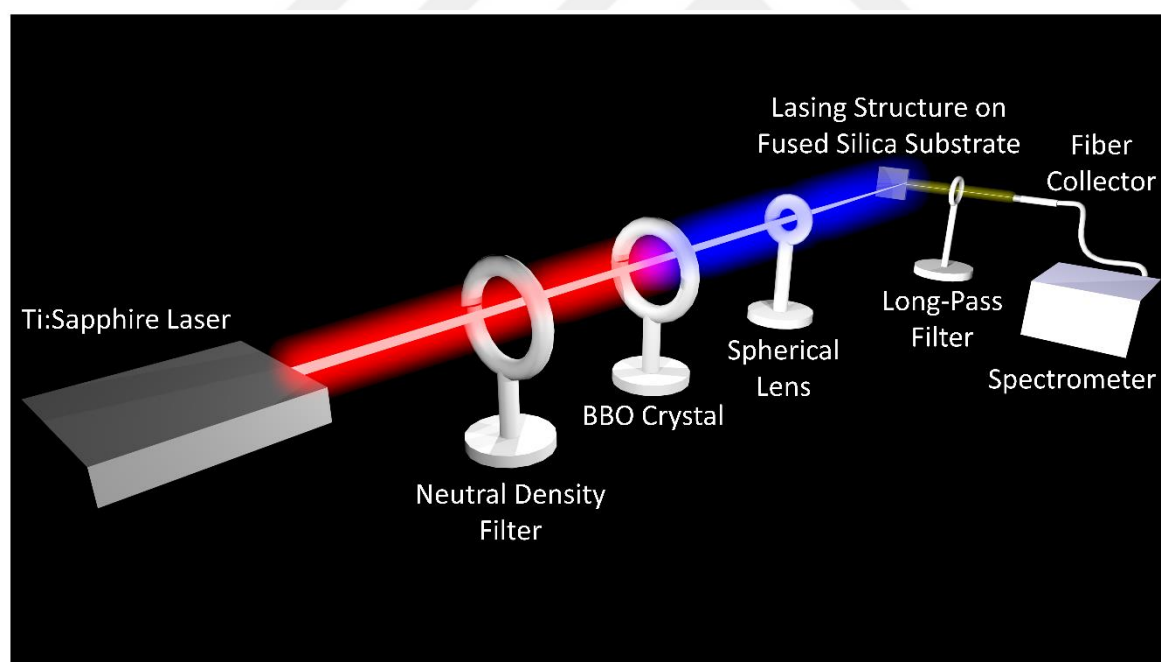


Figure 3.5 Schematic of the optical setup for lasing measurements.

3.3.8 LED Characterizations

3.3.8.1 J-V Characterization

J-V characteristics of LEDs are measured by a voltage source and an amperemeter (Agilent U3606A Multimeter&DC Power Supply). The voltage source applies a gradually increasing voltage across the device under test, and the amperemeter reads the current value at each voltage level. Dividing the current value by the area of the active device gives the current density.

3.3.8.2 Luminance Characterization

The luminance value of the LED is determined with high precision using a spectroradiometer (Konica Minolta CS-2000). First, the device connected to the voltage source is placed in front of the spectroradiometer. The angle between the emission surface and the measurement tool must be carefully aligned since the luminance is highly dependent on the angular emission profile of the LED. Also, the position of the sample should be at the spectroradiometer's focal point so that the measurement is taken from the focused point of the device. Besides the luminance, this measurement also provides the data of the electroluminescence (EL) spectrum and cie coordinates of the LED.

3.3.8.3 EQE Characterization

As discussed in Chapter 2, EQE is the ratio of detected photons to injected electrons. The number of injected electrons can be calculated from the current value passing through the device. In unit time, the following formula gives the number of electrons injected into the LED:

$$N_e = \frac{I}{Q} \quad (3.1)$$

where I is the current value with the unit of amperes (A), and Q is the charge of an electron with the unit of Coloumb (C), which is equal to $1.602 \times 10^{-19} C$. The number of photons is found by measuring the power density of the emitting device located inside an integrated sphere. The sphere is calibrated with a reference sample such that it detects all the emission extracted from the LED. Having the number of photons and electrons, EQE is calculated.

Chapter 4

Dual-Color Amplified Spontaneous Emission and Lasing in Colloidal Quantum Wells

This chapter is based on the publication “Simultaneous dual-color amplified spontaneous emission and lasing from colloidal quantum well gain media in their own layered waveguide and cavity,” **A. T. Isik**, F. Shabani, F. Isik, S. Kumar, S. Delikanli, and H. V. Demir, *Laser & Photonics Reviews*, 2300091 (2023) [39].

4.1 Motivation

For a few decades, intense research efforts have been made on semiconductor micro- and nano-lasers because of their unique advantages including their compactness, fast dynamics, and emission efficiency [40]–[42]. In particular, multicolor lasers have attracted great scientific interest owing to their high demand in many application fields, such as on-chip optical communication [43], integrated photonic circuits [44], [45], biochemical sensing [46], and biological imaging [47], besides their high potential for realizing efficient white light generation [48] and full-color laser display [49], [50]. To date several systems of fluorescent semiconductor nanomaterials and deposition techniques have been investigated as part of the efforts to demonstrate efficient and stable multicolor lasers. Among them, group III-V quantum wells using epitaxial growth [51], group II-VI semiconductors using chemical vapor deposition (CVD) [52], and colloidal perovskites [53] and organic molecules [54] using spin-coating have been employed as the active media for the demonstration of multi-wavelength optical gain and lasing. To achieve the lasing at two or more different wavelengths simultaneously, different types of optical cavities such as distributed-feedback (DFB) cavity [55], whispering-gallery-mode (WGM) cavity [56], and nanowire [57] and nanosheet [58] self-resonant cavities have been implemented.

Solution-processable colloidal semiconductor nanocrystals offer great potential as a gain medium for multicolor laser devices thanks to their low-cost production, easy processability, size-tunable bandgap, strong light absorption, and compatibility with several optical cavity types [20], [59]–[61]. Various studies have been conducted to realize low-threshold single-color optical gain for colloidal quantum dots (CQDs) and their two-dimensional counterparts, colloidal quantum wells (CQWs) [34], [62], [63]. Through the integration of these

nanomaterials into different optical feedback configurations, such as Fabry-Pérot type cavity [64], vertical cavity surface emitting laser (VCSEL) [65], [66], WGM resonator [67], [68], and DFB laser [69], [70], single-color colloidal lasers have been demonstrated. Moreover, broadband multicolor ASE and random lasing have been shown using a particular type of colloidal nanocrystals with large core and shell sizes [71]. However, these “giant” crystalline heterostructures with a highly broadband spectral emission make the sensitive band-gap tuning exceedingly difficult. Also, this approach sacrifices ultra-narrowband ASE, which is one of the most attractive features of colloidal nanocrystals.

Despite all the reports on multicolor lasers, achieving multicolor optical gain and lasing using multiple band-gap tunable narrow-band materials is still challenging because of nonradiative resonance energy transfer (NRET) or, in particular, Förster resonance energy transfer (FRET) [72], [73]. A recent study shows that, when two different colloidal nanocrystals, blue-emitting CQDs and green-emitting CQDs, are in the same medium, energy transfer from the blue CQDs to green CQDs occurs since the absorption bands of the green CQDs and the emission spectrum of the blue QDs overlap [74]. This phenomenon prevents blue CQDs from lasing while increasing the lasing performance of green CQDs. To overcome this challenge and avoid the negative effect of FRET, a new design strategy must be developed.

Here, for the first time, we demonstrate dual-color optical gain and lasing using two active media of separate CQW thin films emitting in the green and red spectral regions. For this purpose, we proposed a structure that distances the gain media with a tunable spacing layer. In this structure, we employed green-emitting core/hot-injection-shell grown CQWs and red-emitting core/crown@shell CQWs as active materials and solution-processed SiO₂ nanoparticles (NPs) as the spacing layer. The red CQWs, SiO₂ NPs, and green CQWs were stacked by sequentially spin-coating on a fused silica substrate. Such structures with tuned thicknesses allowed us to obtain a multicolor ASE at a low threshold of $\sim 17 \mu\text{J}/\text{cm}^2$. The results confirmed the achievement of two narrowband ASE peaks in two colors, green and red. The configuration was combined with a WGM cavity to convert the ASE response into a lasing action. Our proposed multicolor microdisk cavity structure excited by femtosecond laser pulses simultaneously lases in green and red.

4.2 Single-Color ASE in CQWs

To produce a dual-color lasing device, the initial step is to carefully choose two gain materials that exhibit high optical gain performance. This is essential to ensuring efficient and reliable laser operation. Additionally, the ASE thresholds of these selected gain media should be comparable to each other since the dual-color device needs to initiate lasing in both colors at approximately the same pump intensities. Here, starting with 4 monolayers (MLs) CdSe CQWs emitting at 512 nm, the emission wavelength was tuned through alloying the core and deposition of the shell. In particular, increasing the thickness of the CQWs relaxes the quantum confinement effect and results in a red shift of the original emission peak [75]. In this work, two different heterostructures were employed to ensure the highest PLQY and chemical stability. We utilized CdSeS/Cd_{0.1}Zn_{0.9}S alloyed core/hot-injection shell and CdSe/CdS@CdZnS core/crown@shell CQWs as green and red emitters, respectively [76].

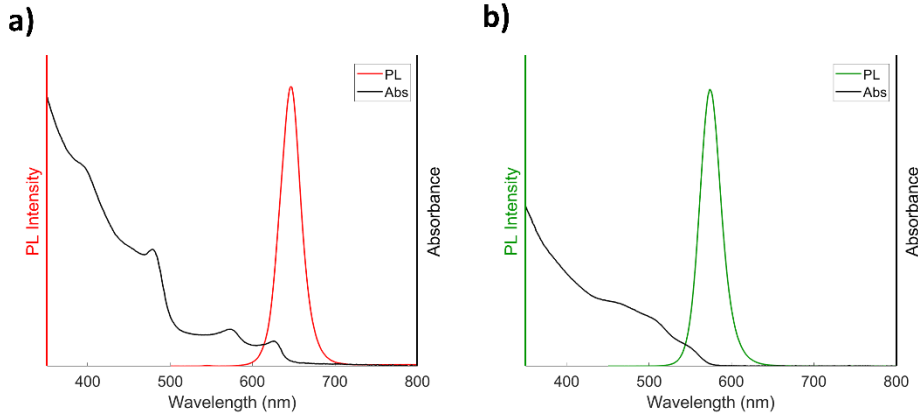


Figure 4.1: PL and absorption spectra of a) the red-emitting CdSe/CdS@CdZnS core/crown@shell CQWs and b) the green-emitting CdSeS/Cd_{0.1}Zn_{0.9}S core/hot-injection-shell CQWs.

Both sets of CQWs have a quasi type-II band structure, in which the electrons with a lower effective mass can leak into the shell more than holes, and also the inorganic crown and shell reduce the hole surface traps and increase the optical performance [77]. PL and absorption spectra of the employed CQWs are presented in Figure 4.1. PL measurements showed that peak emission wavelengths of green and red CQWs are 575 and 642 nm, respectively. We also note that red CQWs absorb strongly in the spectral region where the green CQWs emit. This spectral overlap causes nominally strong energy transfer from green CQWs to red CQWs in close proximity and prevents us from reaching the dual-color optical gain conditions by using our in-solution CQWs in the blended form [78].

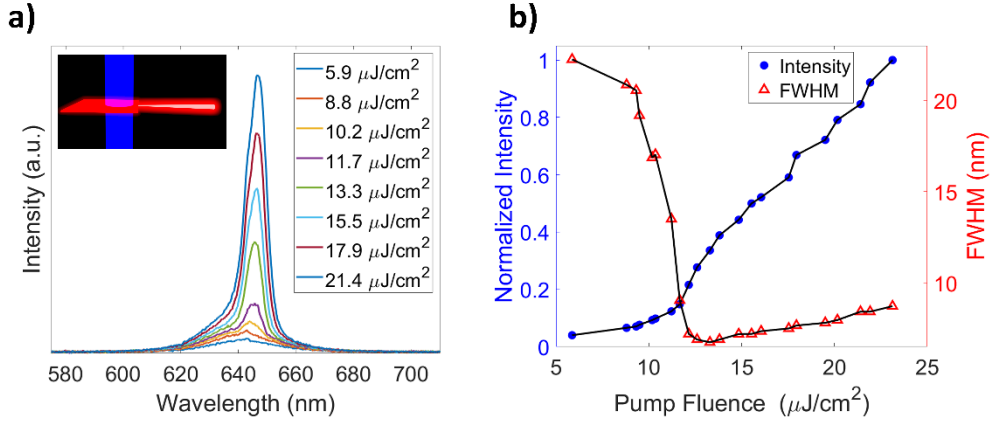


Figure 4.2: a) PL spectra and b) normalized peak intensities along with FWHMs of the red CQWs film under various pump intensities. ASE threshold of this film is $\sim 12 \mu\text{J}/\text{cm}^2$. The inset displays the schematic of the structure under test.

To elaborate on the optical gain behavior of our nanocrystals, we explored each CQWs by coating them on top of fused silica substrates as single thin films. For the optical gain measurements, we excited our device with a pulsed laser beam pulse width of 110 fs and 1 kHz repetition rate at the wavelength of 400 nm. During the characterization, we pumped the device by gradually increasing the excitation energy until the spectrometer was saturated. Figure 4.2b shows the intensity and FWHM of red-emitting CQWs thin film under varying pump fluence. The uptrend of PL intensity starts to accelerate when the excitation energy is higher than $\sim 12 \mu\text{J}/\text{cm}^2$, which is the ASE threshold of the gain medium. After the threshold level, the full-width-at-half-maximum (FWHM) of the emission decreases from 22 to 7 nm. As seen in Figure 4.2a, the film exhibits red-shifted ASE spectra, with the emission peak sliding from 643 to 646 nm. This red shift experienced in the optical gain regime is a characteristic feature of quasi type-II heterostructures [79]. Figure 4.3b depicts the intensity and FWHM values of the green CQWs thin film excited by a pump laser at varying intensity levels. Above the ASE threshold of $\sim 9 \mu\text{J}/\text{cm}^2$, the increment of emission intensity accelerates and the FWHM reduces from 23 to 5 nm.

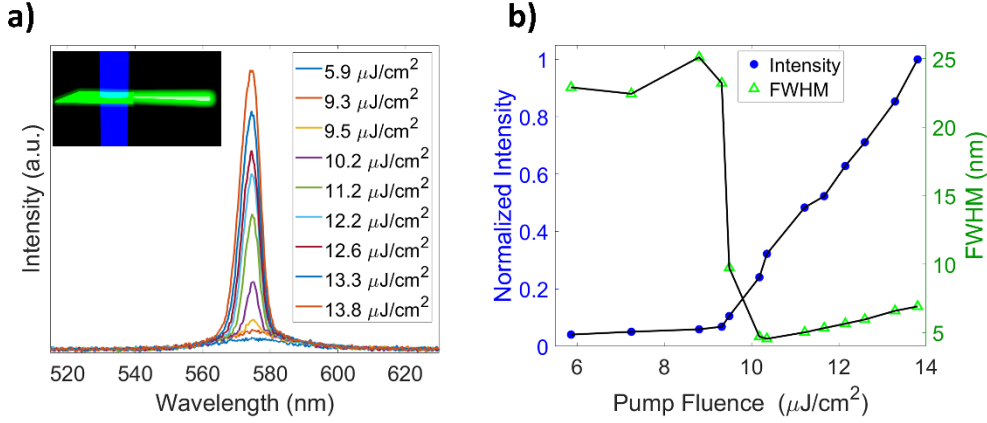


Figure 4.3: a) PL spectra and b) normalized peak intensities along with FWHMs of the green CQWs film under various pump intensities. ASE threshold of this film is $\sim 9 \mu\text{J}/\text{cm}^2$. The inset displays the schematic of the structure under test.

4.3 Dual-Color ASE in CQWs

Avoiding energy transfer is crucial for constructing a device exhibit an efficient multi-color optical gain. In such a device, when the distance between two gain materials is short enough, the emergence of FRET is inevitable [80]. Therefore, separating the CQWs from each other necessitates the selection of a spacing layer with a reasonable thickness that does not allow energy transfer. One of the most critical concerns in choosing the ideal material for the spacing layer is the proportion of generated photons that couple into the active media [35]. The lower the confinement in gain media is, the higher pump energy is required to achieve optical gain. Selecting a low refractive index material for the passive spacing layer will minimize the reduction of photons coupled to active media. Hence, as the spacing layer, we decide to use SiO_2 with a refractive index of 1.48, which is smaller than the refractive index of the films of active layers. The complex refractive indices of the green and red CQW thin films are found as $1.74 + i0.03$ and $1.76 + i0.04$ at their emission wavelength.

We measured the thicknesses and refractive indices of the materials used in the proposed architecture by a variable angle spectroscopic ellipsometer. We took the measurements from the materials coated on a silicon substrate separately. Ψ and Δ data were collected from incidence angles between 55° and 65° . The wavelength range of this measurement was 500 – 1000 nm. We obtained the refractive index data by fitting the measured curve using a Fresnel-based mathematical model. Figure 4.4 shows the real and imaginary parts of the complex refractive indices of all three materials.

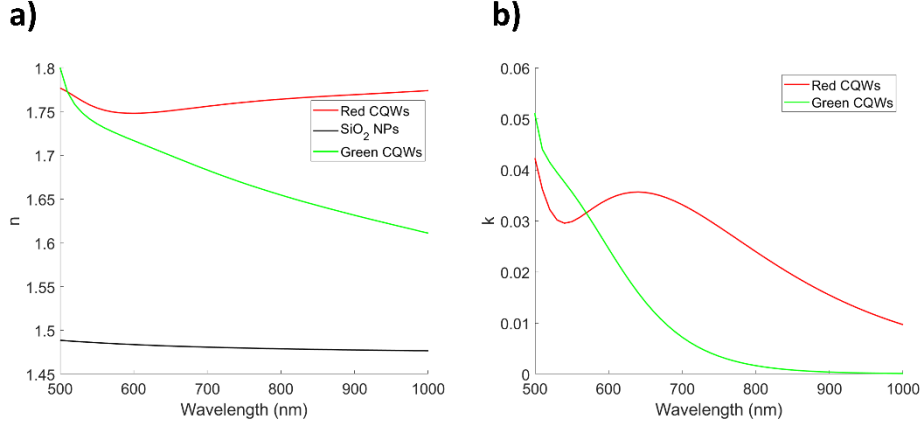


Figure 4.4: a) Real and b) imaginary parts of the complex refractive indices of red-emitting CQWs, SiO₂ NPs, and green-emitting CQWs.

In our architecture, energy transfer occurs from green-emitting CQWs (donor) to red-emitting CQWs (acceptor) via FRET. Through numerical calculations, we determined the total rate of FRET for various thicknesses of the spacing layer. We modeled our proposed waveguide architecture using the finite-difference time-domain (FDTD) model (Lumerical Inc.). The green-emitting CQWs layer in the waveguide was represented as a single radiating dipole located on top of the spacing layer, whereas the red-emitting CQWs layer was modeled as a lossy medium. For different spacing layer thicknesses, we performed numerical analyses to find the electric field inside the acceptor region, and the total rate of FRET was computed by using the following formula [72]:

$$\Gamma_{FRET} = \frac{Im(\epsilon_{acceptor}(\omega))}{2\pi\hbar} \int \mathbf{E} \cdot \mathbf{E}^* d\mathbf{v} \quad (4.1)$$

Figure 4.5 depicts the total FRET rate for various spacer thicknesses, and Figure 4.6 shows the electric field distributions inside the acceptor region.

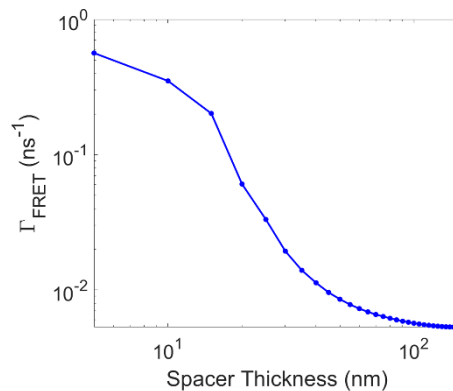


Figure 4.5: Total FRET rate of multilayered waveguide structure for various thicknesses of spacing layer.

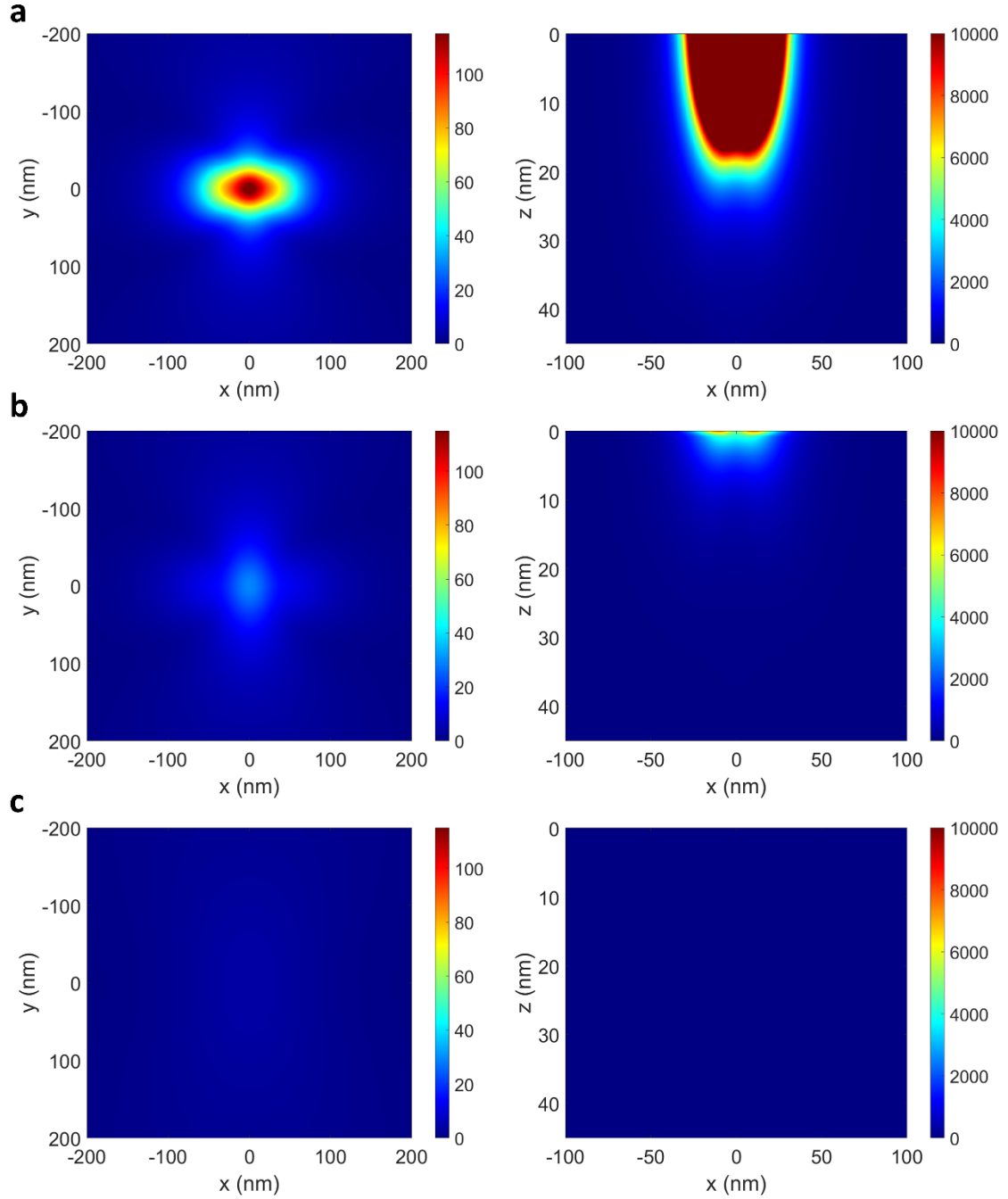


Figure 4.6: Electric field intensity distributions on xy-plane (left column) and xz-plane (right column) within the red-emitting active layer for (a) 5 nm, (b) 25 nm, and (c) 125 nm spacing layer thicknesses.

As the thickness of this layer increased, we observed a profound decrease in the FRET rate. To achieve a FRET rate that is close to zero (practically zero), it is necessary to select a spacing layer thickness that is greater than ~ 100 nm. Choosing a spacing layer thickness below ~ 100

nm leads to a non-negligible amount of nonradiative energy transfer, resulting in increased optical loss and elevating the ASE and lasing threshold values.

Though it is possible to deposit SiO₂ thin film using intricate techniques such as plasma-enhanced chemical vapor deposition (PECVD), thermal evaporation, and sputtering, spin-coating solution-processable SiO₂ NPs is the most versatile method to implement a separating layer without any requirement of using high-temperature process. The solvent to disperse the SiO₂ NPs must also be carefully selected, which should be orthogonal to the solvent of the underlying film to avoid complications during the spin-coating procedure. The freedom of choice in the stabilizing ligands of the NPs allows us to use the same material in different solvent environments. We chose SiO₂ NPs dispersed in water, an orthogonal solvent for the underlying emissive layer.

Fabrication of the proposed structure started by cleaning the fused silica wafer, which is a suitable choice as the substrate since it has a low refractive index and is transparent in the visible range. Then, by employing only the spin-coating technique, we built an active-passive-active film architecture in the following order: red CQWs (50 g L⁻¹, 45 nm)/SiO₂ NPs (85 g L⁻¹, 120 nm)/green CQWs (50 g L⁻¹, 45 nm). Optical characterization of these films was conducted following the same procedure explained above. Figure 4.7 depicts the schematic of our dual-color device under optical excitation.

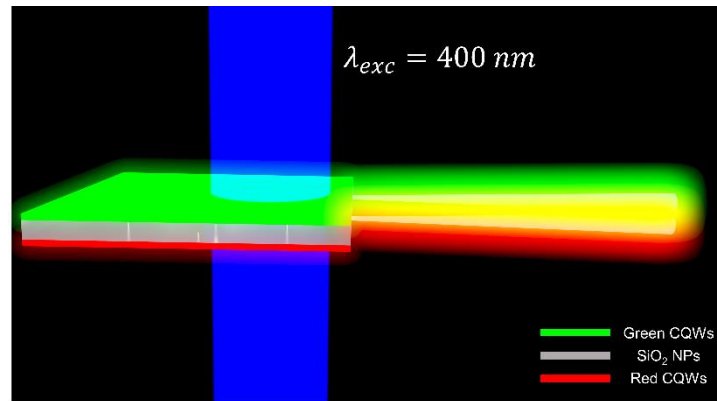


Figure 4.7: a) Schematic view of the multilayered film under the 400 nm pump excitation illustrating dual-color ASE behavior.

The evolution of the PL spectra with pump energy is displayed in Figure 4.8. In the beginning, with the low pump levels, the PL spectra present two peaks at 643 and 569 nm and relatively broadband spectra with FWHM of 26 nm for red and 30 nm for green emissions. Intensities of green and red emissions increase linearly until the excitation energy reaches a threshold, ~16 μJ/cm² for the red emission and ~17 μJ/cm² for the green emission (Figure 4.9a, b). This finding

suggests that, compared to the ASE thresholds of single-layered films, the multicolor configuration shows higher threshold levels individually for both the red and green.

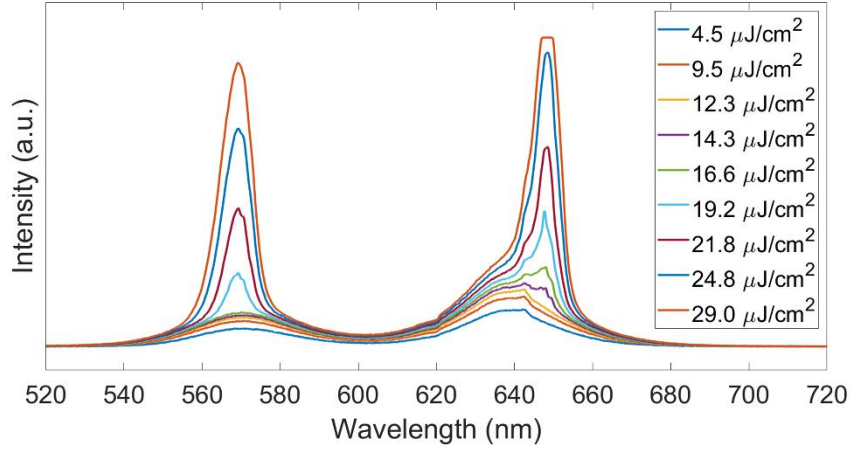


Figure 4.8: PL spectra of the multicolor structure under various pump intensities.

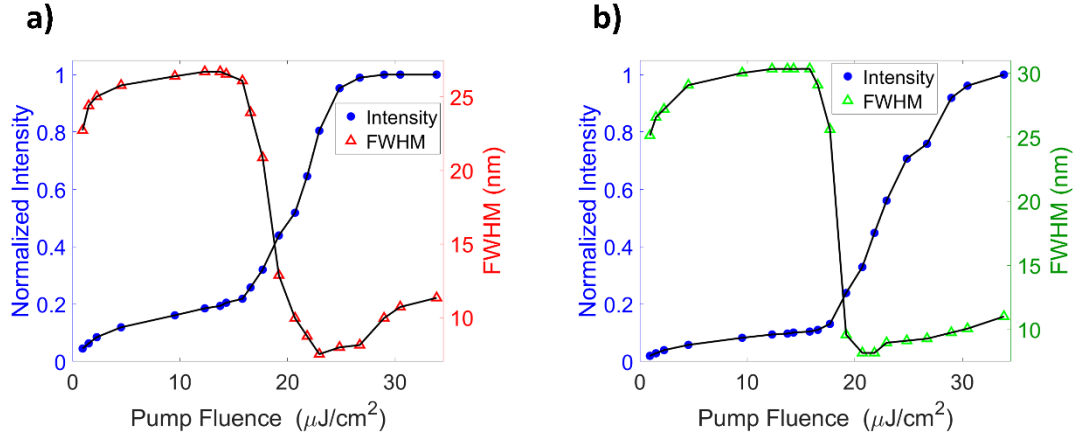


Figure 4.9: Normalized peak intensity and FWHM in the a) red and b) green spectral emission regions. ASE thresholds in the red and green regions are ~ 16 and ~ 17 $\mu\text{J}/\text{cm}^2$, respectively.

For the red emission, the superiority of a single thin film over the multicolor configuration in terms of optical performance is due to the absorption and reflection that the pump laser experiences in the first and second layers (spacing layer and green CQWs) of the multilayer structure, which causes partial loss of excitation energy. However, unlike the red CQWs, the green CQWs are in the first layer of the multicolor stack, and hence pump laser interacts with it before the other layers. Therefore, green CQWs do not suffer from insufficient excitation power since they are not affected as much by the absorption and reflection of pump light inside the spacing layer and red gain medium. However, in a multilayer structure, each medium introduces a certain amount of roughness due to imperfections in the coating process. The roughness accumulates with the deposition of each new layer. Since the green gain medium is the last coated layer in our multicolor configuration, it has the highest roughness. As the high

optical gain requires a smooth and uniform waveguide structure, using a rough medium in ASE measurements decreases the performance [81]. This can explain the higher ASE threshold in the multicolor structure compared to the single green thin film.

In addition to the impact of roughness-induced scattering losses and decreased pump fluence, the optical performance of multilayer structures is also influenced by a reduced confinement factor as a result of incorporating another high refractive index gain medium into the structure, which decreases the overlap between each of the gain media and the associated optical modes at their respective emission wavelengths. This reduction in the confinement factor should in turn increase the ASE threshold in our multilayer structure.

Above the thresholds, we observed a slight red shift in the peak red emission wavelength from 643 to 648 nm, while the peak green emission wavelength remained the same. The amount of change is larger by 2 nm compared to the multicolor configuration. This difference is due to the size increase in the waveguide since this spectrally determines the supported mode and the modal distribution affects the peak wavelength of the ASE spectra. Besides the spectral shift, the increase in PL intensities of green and red emissions stepped up. Also, FWHM values in two distinct spectral regions decreased drastically. PL spectra show narrowband emission with FWHMs of 8 nm for both colors, indicating that the single film exhibits slightly narrower ASE spectra than that of the multicolor layer stack. This small change can also be attributed to the evolution of modal distribution inside the waveguide. Because the multicolor structure introduces a larger waveguide size, broader ASE spectra are expected. The sharp changes in intensities and FWHMs prove that by exciting our multicolor structure with a pump fluence level above the threshold of $17 \mu\text{J}/\text{cm}^2$, dual-wavelength ASE behavior emerges.

4.4 Single-Color Lasing in CQWs

After showing a multicolor ASE behavior, we designed single-layer architectures by integrating our CQWs with a cavity. WGM resonator is a very suitable candidate to make the cavity out of our thin films. To make a WGM cavity, we designed a single-layered microdisk structure that includes a CQW layer. Although different types of WGM resonators, such as microspheres [50] and fiber lasers [82], could be employed to show single-color lasing, we selected the microdisk structure due to its compatibility with our multicolor ASE configuration. The proposed microdisk laser allows for modification of the lateral axes of the ASE structure by patterning without changing the vertical axis.

We examined the lasing behavior of our CQWs by simulation, fabrication, and characterization of two different single-color microdisk structures. First, we performed a simulation utilizing FDTD based full-wave electromagnetic solutions. The material of the microdisk is assumed to be lossless dielectric with a refractive index of 1.8. In the model, the disc is surrounded by air ($n=1$). Dipole sources with three fundamental orientations at the microdisk's center initiate the signal. We obtained the optical response of the cavity by running two-dimensional FDTD simulations. The simulations determined the electric-field distribution and the spectral modes by a field monitor encompassing the microdisk. Figure 4.10 displays the layout used in the FDTD simulation setup. Figures 4.11a and 4.12a show the cavity modes of green and red microdisks along with the measurement results.

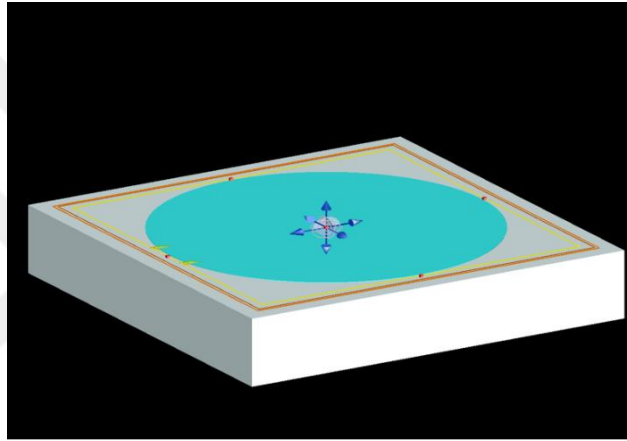


Figure 4.10: Layout of the optical cavity architecture for FDTD simulation recording the electric field distribution.

Figures 4.11b and 4.12b display the electric-field distribution on a single-layer microdisk structure. The field is concentrated at the sides of the microdisk thanks to the resonance presented by WGM. Based on our simulation, we fabricated and characterized two separate single-color microdisk lasers with the active media of red CQWs and green CQWs individually. To characterize the lasing behavior of our microdisks, we excited the structures with the same laser beam used for ASE characterization. Since microdisk-type WGM resonators lase from the edges, we collected the emission from the side of each structure.

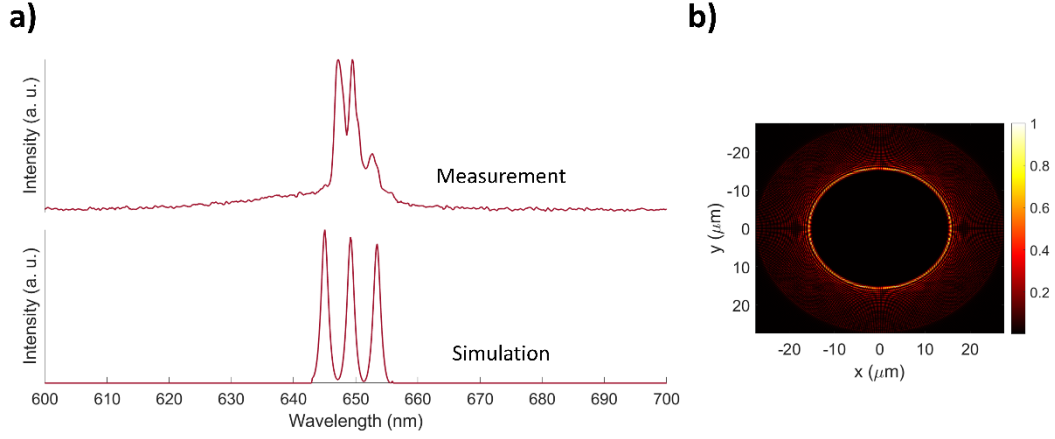


Figure 4.11: a) Experimental and numerical results of lasing modes and b) electric-field distribution of red microdisk resonator.

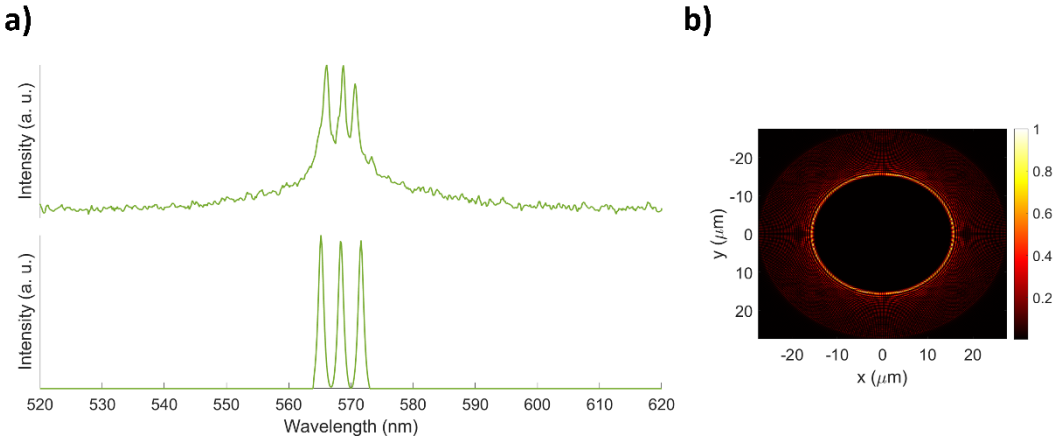


Figure 4.12: a) Experimental and numerical results of lasing modes and b) electric-field distribution of green microdisk resonator.

Pump-dependent PL intensities of red and green lasers are presented in Figures 4.13b and 4.14b. The threshold level of the red laser is $96 \mu\text{J}/\text{cm}^2$, whereas the green laser has a threshold of $94 \mu\text{J}/\text{cm}^2$. We note that, compared to the ASE of the red and green CQWs, with thresholds of $12 \mu\text{J}/\text{cm}^2$ and $9 \mu\text{J}/\text{cm}^2$, respectively, lasing actions in microdisk structures require much higher pump energy. This rise in the threshold can be explained by the optical loss presented in the system by the WGM cavity. Although an ideal microdisk resonator does not have any cavity loss except for the outcoupling, in practice, irregularities of the optical structure introduce additional loss to the system. As can be seen in the insets of Figures 4.13b and 4.14b, our microdisk lasers contain imperfections around their edges. Due to the optical losses imposed by these imperfections, higher pump energy is necessary to observe lasing. Figures 4.13a and 4.14a present the spectral response of PLs measured from the red and green single-color microdisk lasers.

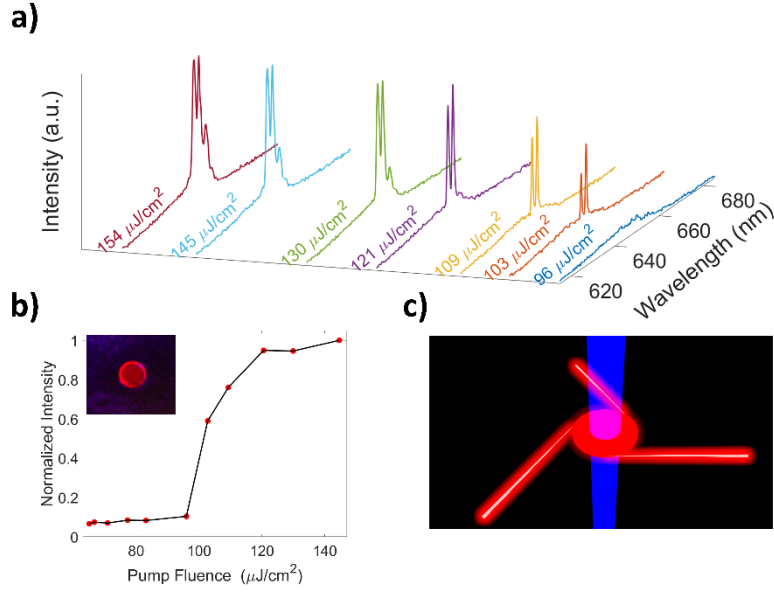


Figure 4.13: a) PL spectra of the single-layer red CQWs microdisk under various pump intensities. b) Normalized peak intensity of the red CQWs microdisk under various pump intensities. Lasing threshold of the structure is $\sim 96 \mu\text{J}/\text{cm}^2$. The inset shows the PL image of the microdisk. c) Schematic of the structure under test.

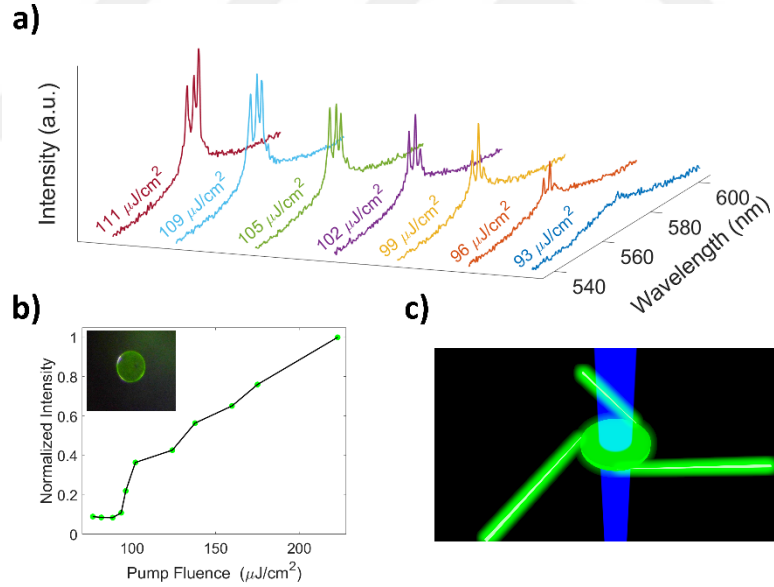


Figure 4.14: a) PL spectra of the single-layer green CQWs microdisk under various pump intensities. b) Normalized peak intensity of the green CQWs microdisk under various pump intensities. Lasing threshold of the structure is $\sim 94 \mu\text{J}/\text{cm}^2$. The inset shows the PL image of the microdisk. c) Schematic of the structure under test.

From Figures 4.13a and 4.14a, we find the FWHMs structures to be $\sim 0.9 \text{ nm}$ for the red microdisk and $\sim 1.2 \text{ nm}$ for the green microdisk. Besides, FSRs are measured to be 2.15 and 2.05 nm for the red and green lasers, respectively. As can be seen in Figures 4.11a and 4.12a,

we compared the measured spectral response with the results of numerical simulations and found that there was a high degree of concordance between the measured and simulated data, despite some minor discrepancies observed when comparing the two graphs, which may be attributed to variations in the sample or measurement conditions.

4.5 Dual-Color Lasing in CQWs

The achievements in demonstrating multicolor optical gain from a well-designed multilayered structure and single-color lasing inside a WGM cavity proved that a multicolor laser can be realized via our CQWs. For this purpose, we integrated our multicolor ASE structure with a WGM cavity to concurrently observe lasing action in two separate wavelengths. To obtain a multicolor WGM cavity, we designed a multilayered microdisk structure that includes two active media and the spacing layer.

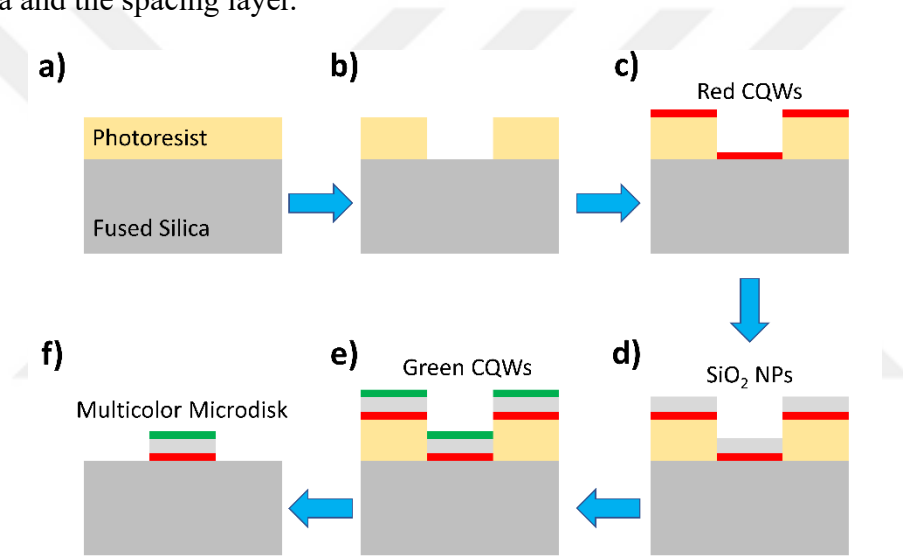


Figure 4.15: Schematics of the multicolor microdisk's process flow. (a) Photoresist coating on a fused silica substrate. (b) Patterning of the resist by photolithography. Coating of (c) red-emitting CQWs, (d) SiO₂ NPs, and (e) green-emitting CQWs. (f) Lift-off of the photoresist.

Figure 4.15 explains the fabrication steps of our multicolor laser. First, we spin-coated AZ5214 photoresist on a fused silica substrate at 5,000 rpm for 75 s and kept it on a hot plate at 110 °C for 1 min. Then, we patterned a circular shape with a 50 μm diameter on the baked film via the maskless photolithography method using a commercial product, MicroWriter ML3 (Durham Magneto Optics Ltd.). The tool used a laser beam with a beam radius of 1 μm and an intensity level of 300 mJ/cm^2 at 405 nm in the lithography process. Afterward, we post-baked the patterned photoresist film on a hot plate at 110 °C for 1 min. For the development, we used AZ351B developer by mixing it with DI water at a ratio of 1:4. We dipped the film in the developer-DI water solution and waited for 140 s. Then, we rinsed the film in DI water for 30

s to clean off the developer and stop the development. We applied oxygen plasma treatment to the film's surface for 1.5 min to remove the remaining photoresist residue on the patterned area. After the patterning process, we built the layers of the proposed design on the substrate. First, we spin-coated the red CQWs solution dispersed in hexane with a concentration of 50 g/L at 1,500 rpm for 60 s. The sample dried on a hotplate at 60 °C for 10 min. Then, we spin-coated the SiO₂ NPs solution in water with a concentration of 85 g/L at 3,500 rpm for 75 s and kept the sample on the hotplate at 60 °C for 30 min. We coated the green CQWs and dried them using the same spin-coating and drying parameters as the red CQWs. Finally, to lift off the photoresist, we kept the sample in acetone for 20 s and rinsed it with isopropyl alcohol.

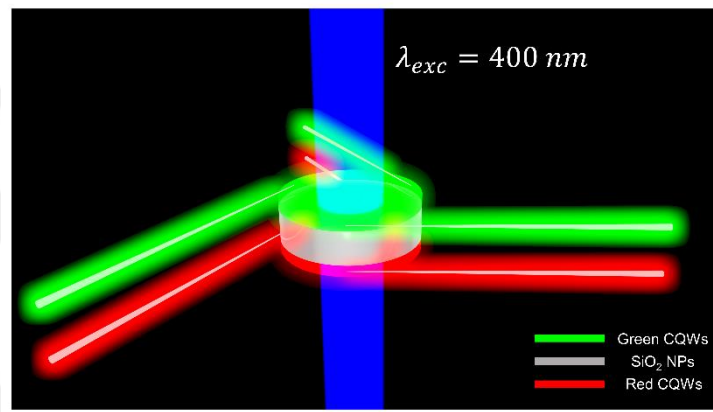


Figure 4.16: Schematic view of the proposed multicolor microdisk architecture under the 400 nm pump excitation showing dual-color lasing behavior.

This multilayer architecture is the same as the multicolor ASE configuration to preserve the optical gain properties. A schematic of the microdisk laser under the optical excitation is presented in Figure 4.16. Figure 4.17 shows the multilayer microdisk's tilted and non-tilted scanning electron microscopy images.

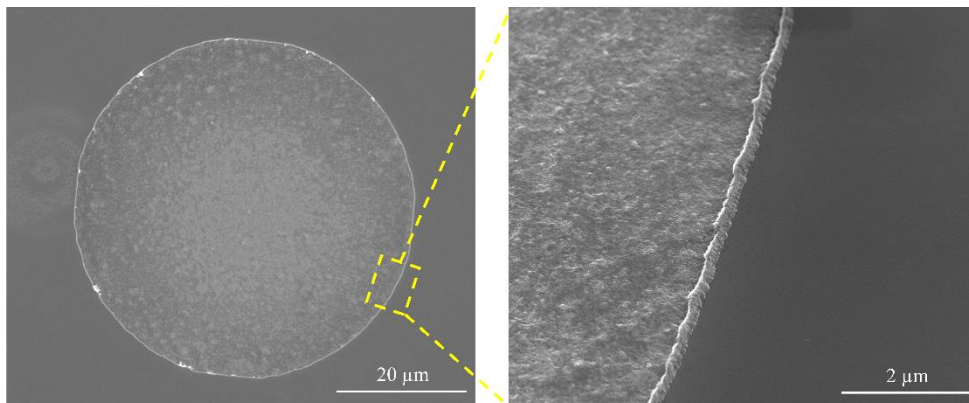


Figure 4.17: SEM images of the multilayer microdisk tilted at 0° (left) and 52° (right).

The emission spectra of the multicolor microdisk WGM laser at different pump fluences are given in Figure 4.18a. The spectral response shows the signature features of a multi-mode dual-color lasing behavior since multiple lasing peaks appear in both emission regions at the pump fluences above the threshold. These lasing spectra present narrow spectral features with FWHMs of ~ 2 and ~ 1.5 nm for the green and red emissions, respectively. As shown in Figure 4.18b, the lasing threshold for both the green and red regions is observed at the pump fluence of $\sim 106 \mu\text{J}/\text{cm}^2$ with the appearance of a narrow peak and superlinear increase of the intensity. Thus, our dual-color laser has a single threshold that is valid for both colors.

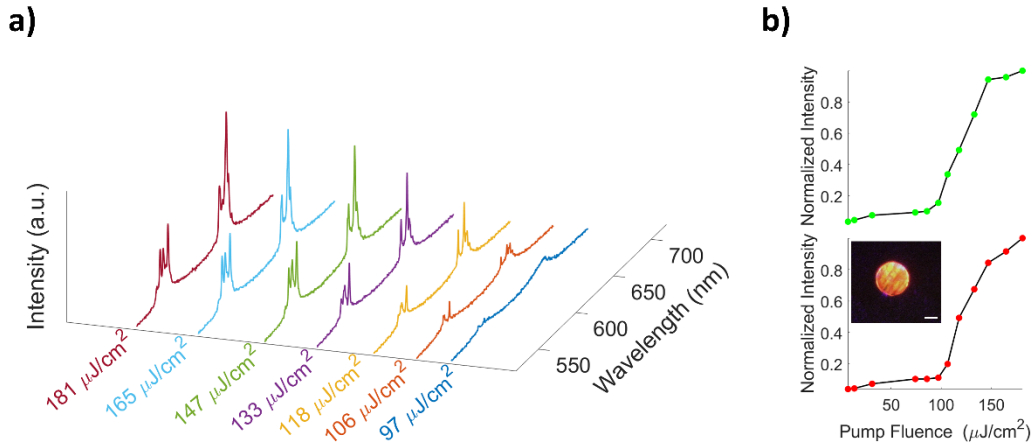


Figure 4.18: a) PL spectra of the multicolor microdisk laser under various pump intensities. b) Normalized peak intensity in green and red spectral emission regions showing that the lasing threshold is $106 \mu\text{J}/\text{cm}^2$ for both colors. The inset shows the PL image of the microdisk excited by a 405 nm continuous-wave laser. The scale bar is $20 \mu\text{m}$.

We realized that the threshold level of our multicolor laser is slightly higher than the thresholds of the single-color lasers ($\sim 96 \mu\text{J}/\text{cm}^2$). Because each layer comes with an additional loss due to scattering and residual absorption, a multilayer structure is expected to include more losses. Indeed, a spacing layer of nanoparticles showing a vital scattering feature and the second active medium introduce a significant amount of optical loss to the system. This additional loss further increments the lasing threshold. Furthermore, FWHM values indicate that the multilayer architecture decreases the Q-factors from 722 to 433 for the red lasing and 467 to 280 for the green lasing. Similar to the rise in the threshold level, the fall of the Q-factors stems from by the higher cavity losses of the multicolor structure. In future investigations, the quality of lasing can potentially be enhanced through a variety of approaches including utilization of an etching process leaving a smoother edge, incorporation of a flatter spacer layer between the active

layers, and adaptation of other optical cavity architectures that would possibly be less subject to the edge effects and thus suffer less from the edge roughness.

4.6 Summary

In summary, we demonstrated multicolor lasing in the visible region using a novel colloidal layered architecture having all-solution-processed cavity composed of two separated CQW layers for green and red emissions and a dielectric spacing layer of silica nanoparticles between these gain films, which effectively prevents degradation of the green emission by eliminating otherwise strong nonradiative energy transfer from the green to the red CQWs. Here, we showed the achievement of multicolor ASE by exploiting these designed optical structures with a low threshold of $\sim 17 \mu\text{J}/\text{cm}^2$. The configuration was also integrated into a multilayered microdisk cavity acting as a WGM resonator. This architecture exhibits multi-mode lasing behavior in two colors simultaneously, with a lasing threshold of $\sim 106 \mu\text{J}/\text{cm}^2$ both in the red and green spectral emission regions. Our findings reveal the promising potential of CQWs as the active media for multicolor optical gain applications. Moreover, relying on this low-cost and easy-fabrication method, such an all-colloidal layered laser structure is an excellent candidate for many optoelectronic applications including integrated photonic circuits and multicolor laser displays.

Chapter 5

Light-Emitting Diodes of Colloidal Quantum Wells with Directed Bright Edge Emission

5.1 Motivation

After the first demonstration of electroluminescent devices in 1928 [83], progress made in the research of light-emitting diodes (LEDs) has opened a new era for indoor and outdoor lighting, advanced displays, and consumer electronics. Owing to their superiority in low power consumption, long lifetime, directional emission (relative to blackbody emitters), compact size, and integration capability with semiconductor driving circuitry, LEDs have prevailed quickly as promising alternatives to conventional light sources such as incandescent bulbs and fluorescent lamps and have become the dominating technology today. To further decrease the manufacturing costs, various semiconductor materials, such as group III-V compounds [84], organic polymers [85], perovskites [86], and colloidal quantum dots (CQDs) [87], have been utilized as emissive layers for LED structures.

Along with the emissive materials mentioned above, colloidal quantum wells (CQWs), 2D counterparts of CQDs, have recently been attracting great interest in LED research [88]. These nanocrystals' near-unity photoluminescence quantum yield (PLQY) enables high external quantum efficiency (EQE) devices. Also, enhanced color purity LEDs are achieved thanks to the narrow emission bandwidth of CQWs. By tuning the size of these crystals during the colloidal synthesis, the peak electroluminescence wavelength of the device can be adjusted precisely. Because of these unique optoelectronic properties, CQWs offer a bright future for the next generation of LED applications.

Over the past few years, significant breakthroughs have been demonstrated in the research of colloidal quantum well light-emitting diodes (CQW-LEDs). For example, in the past years, our group has reported red-emitting CQW-LEDs with a record-high EQE value of 19.2% by enhancing the PLQY of the CQWs into near-unity values [89]. In addition, Kelestemur et al. have shown ultra-bright electroluminescence (EL) from CQWs of graded shell composition with a luminance of $\sim 46,000$ cd/m² [90]. Though these record-high values have been achieved by engineering the shape and composition of the nanocrystals, it is also possible to increase the outcoupling efficiency by controlling the orientation of the CQWs taking advantage of their

anisotropy. In a previous work of our group, we reported a significant improvement in the outcoupling efficiency of CQW-LEDs from 22% to 34%, thanks to the all-face-down orientation of self-assembled CQWs [91]. Besides the research efforts on high-performance devices, our group demonstrated LEDs utilizing various heterostructures of CQWs such as CdSe/CdZnS core/hot-injection-shell (HIS) [91], (CdSe/CdS)@(1-4 CdS/CdZnS) (core/crown)@(colloidal atomic layer deposition shell/HIS) [77], and CdSe/CdSeTeCdSe/CdS core/multicrown CQWs [92]. However, it should be noted that the works cited above all investigate the surface-emitting CQW-LED architecture, referring to the group of devices that extract this emission surface-normal to their substrate.

In surface-emitting devices, the outcoupling efficiency is limited because a high portion of the generated light leaks to the substrate and cannot be extracted. On the contrary, edge-emitting LEDs typically have a much higher level of outcoupling efficiency and hence present a great potential for a significant increment in output power. Also, edge-emitting devices can be fabricated in more compact sizes, if designed right, providing better design flexibility and the ability to control the directivity of the emission. These advantages make such edge-emitting LEDs commonly preferred in specific applications such as optical communications [93], stereoscopic displays [94], and photonic integrated circuits [95]. Although several studies have been conducted to demonstrate edge-emission from electroluminescent devices with various emissive layers, such as GaN-based quantum wells [96], organic emitters [97], [98], perovskites [99], and CQDs [100], to the best of our knowledge, to date, there has been no report on CQW-LEDs with the emission from the device edge.

In this thesis study, we demonstrated edge-emitting CQW-LED with high luminance and high current density. For this purpose, we proposed an architecture consisting of two reflective metal electrodes that confine the light inside the active device, electron and hole transport layers to increase the charge injection efficiency, and a CQW layer for light emission. We integrated this architecture into a thin waveguide slit that directs the emitted photons to the edge of the device. Guiding the light into the side of the LED, we obtained electroluminescence with a luminance level of $\sim 20,400 \text{ cd/m}^2$. Moreover, owing to the micro-scale current injection area of our LED structure, we reached a current density value of up to $\sim 5.6 \text{ A/cm}^2$.

5.2 Surface-Emitting LEDs of CQWs

To obtain high-performance edge-emitting LEDs, we need an electroluminescent device structure with the highest possible efficiency in charge injection and brightness. Integrating this

structure into a systematically designed optical architecture, the emission direction can be modified. Therefore, we started our experiments by optimizing a conventional surface-emitting CQW-LED. Optimizations on this device were conducted to achieve a high luminance value that indicates high extracted photon number through the LED structure.

To minimize the loss of generated photons during the light extraction, we selected glass as the substrate due to its transparency in optical wavelengths. Also, we utilized indium tin oxide (ITO), a transparent conductive material, as the cathode for the same reason. On top of the ITO, we coated zinc oxide (ZnO) with a thickness of 40 nm as the electron transporting layer. For the emissive layer, we employed CdSe/Cd_{0.25}Zn_{0.75}S core/hot-injection-shell grown CQWs, which has an in-solution photoluminescence quantum yield (PLQY) of ~85%. As the hole transporting and hole injection layers, we used 4,4-N,N-dicarbazolebiphenyl (CBP) and molybdenum trioxide (MoO₃) with 60 and 10 nm thicknesses. Lastly, an 100 nm aluminum (Al) layer serves as the anode of the LED. Figure 5.1a illustrates the schematic of our surface-emitting CQW-LED structure. This architecture presents a stepwise energy band alignment to achieve a high charge injection efficiency (see Figure 5.1b).

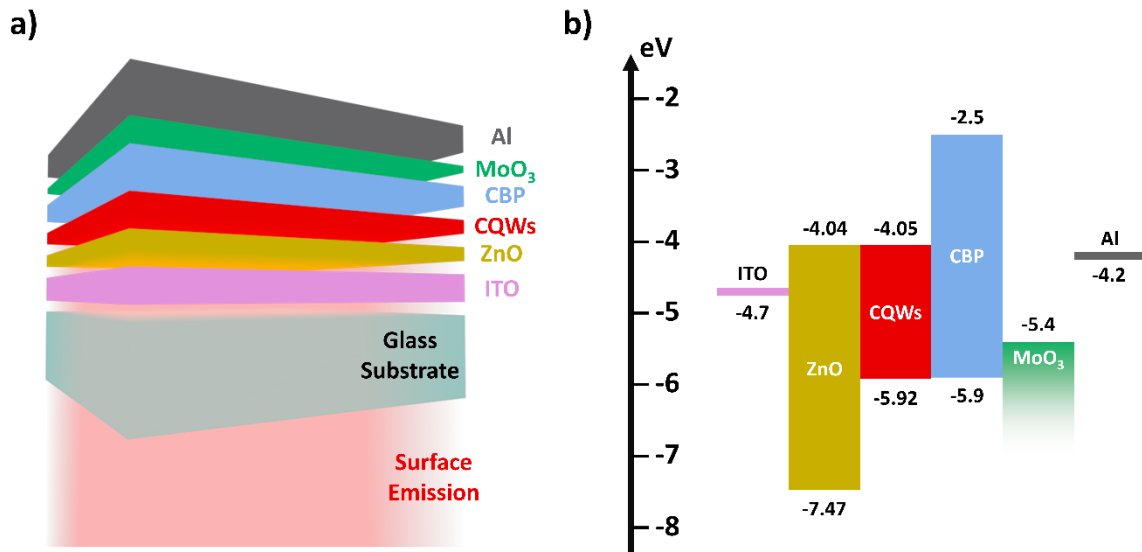


Figure 5.1: a) Schematic view and b) energy band diagram of the surface-emitting CQW-LED architecture.

We carried out the fabrication process by employing different deposition techniques and parameters for each material. For instance, ZnO (dispersed in ethanol with a concentration of 25 g/L) and CQWs (dispersed in toluene with a concentration of 12 g/L) layers were dynamically spin-coated at a rotation speed of 2,000 rpm on top of an ITO-coated glass substrate, and kept on a hotplate at 110 °C for 40 min and 75 °C for 10 min, respectively. CBP, MoO₃, and Al layers were deposited by thermal evaporation technique at a deposition rate of

$\sim 2\text{\AA/s}$. Because these layers are highly sensitive to oxygen and moisture, we performed the fabrication in a nitrogen (N_2) filled glove box system. Finally, we encapsulated the device with ultraviolet-curable epoxy resin to avoid performance degradation in the ambient atmosphere.

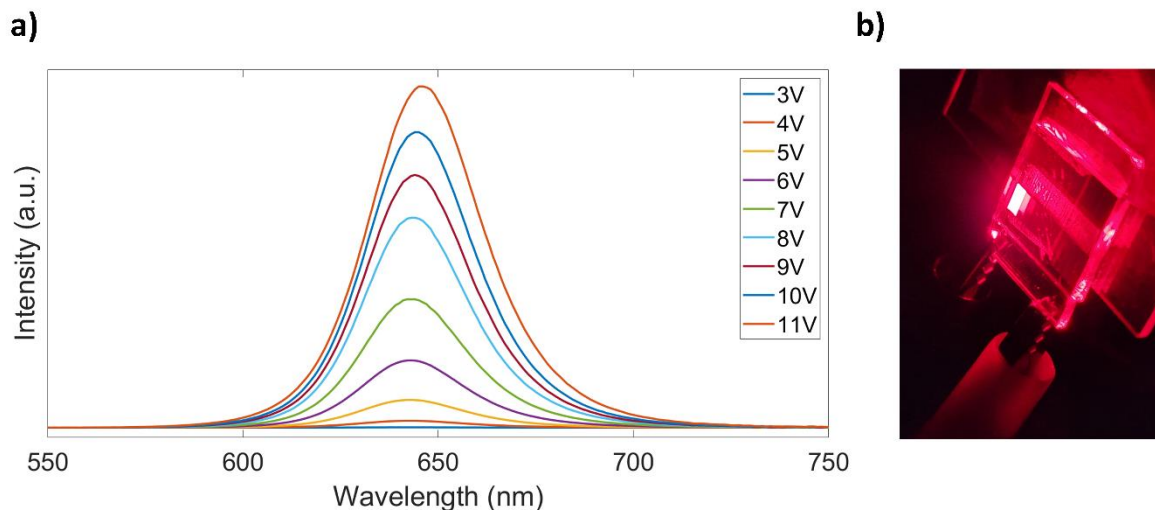


Figure 5.2: a) Evolution of EL spectrum of the surface-emitting CQW-LED in response to varying voltage levels. b) Photograph of the surface-emitting LED under a voltage bias.

After encapsulating the device, we took the LED into the air environment for electrical and optical characterizations. First, we determined the EL spectrum of the device for varying voltage levels by a spectrometer. As seen in Figure 5.2a, the emission intensity peak is located at the wavelength of 645 nm with a full-width-half-maximum (FWHM) of ~ 35 nm. Figure 5.2b shows a photograph of our CQW-LED presenting a bright surface emission under a bias voltage of 7 V.

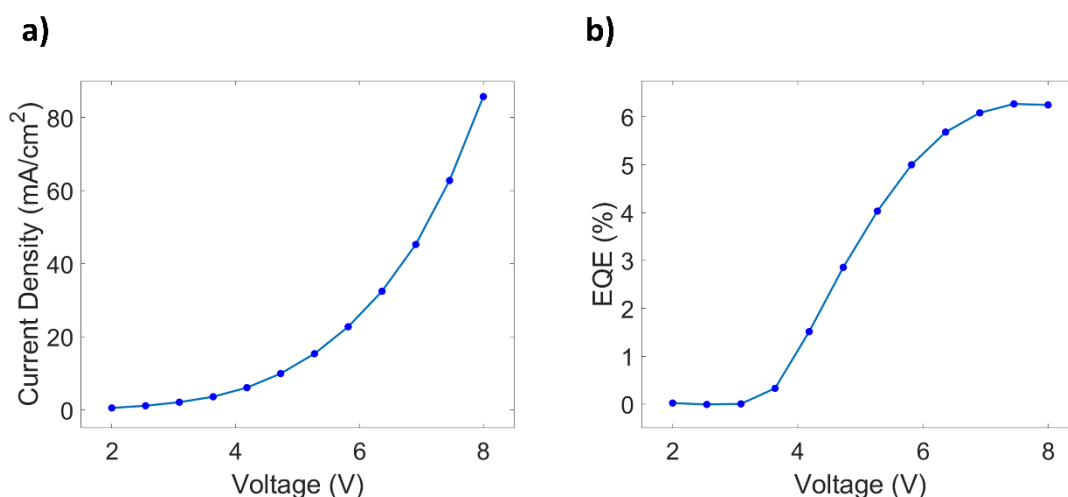


Figure 5.3: a) Voltage vs. current density and b) voltage vs. EQE graphs of the surface-emitting CQW-LED.

To obtain the EQE of our LED, the number of injected electrons and generated photons must be calculated. For the injected electrons, we measured the current density passing through the active area of the device (4.5 mm^2). Figure 5.3a displays the graph of current density values at varying voltage levels. The diode behavior of our device with a turn-on voltage of $\sim 3.5 \text{ V}$ can be observed from this plot. Also, we noted that our CQW-LED reaches the current density level of 85 mA/cm^2 . We calculated the total extracted photon number by placing the LED into an integrated sphere calibrated according to a reference sample with precisely known radiation flux. Having these two quantities, EQE is computed for different bias voltages. As depicted in Figure 5.3b, the peak EQE value of our device is 6.2%.

To measure the maximum luminance value of the CQW-LED, we located the device in front of a spectroradiometer. Because the EL intensity of the surface emission varies with the observation angle, we aligned the spectroradiometer to make a 90° angle with the device surface and collected the maximum possible emission. Figure 5.4 depicts the change in the luminance with the varying voltage levels applied across the LED. The increase in the luminance starts to accelerate at $\sim 4 \text{ V}$ and reaches its peak value of $14,765 \text{ cd/m}^2$ at 12 V .

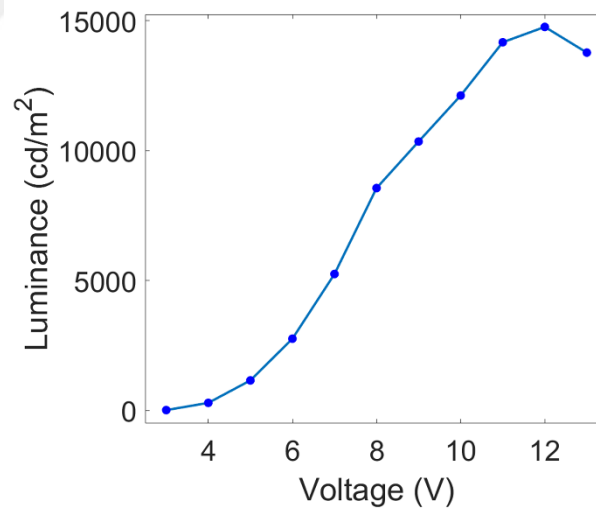


Figure 5.4: Voltage vs. luminance graph of the surface-emitting CQW-LED.

5.3 Edge-Emitting LEDs of CQWs

After successfully fabricating and characterizing a surface-emitting CQW-LED, we designed an optical device architecture that suppresses the surface emission and enhances the edge emission. Since we proved that our surface-emitting device exhibits high efficiency and great brightness, there is no need to utilize a new charge injection structure. Instead, we made modifications to the surface-emitting LED and built a passive optical structure around it.

Besides the electrical and optical properties of the CQW-LED, we also engineered its thermal features since it significantly affects the performance of our device. During the LED operation, an increment in the current density leads to a rise in brightness up to a specific level. Then, the applied electrical power causes excess heating, and the luminance degrades. In an LED, dissipating the heat by surrounding the active device with thermally conductive materials prevents overheating even in high current densities.

In the surface-emitting architecture, to achieve optical transparency, glass and ITO were selected as the substrate and the cathode, respectively. However, since the edge-emitting device requires to block the emission extracted from the surface, we used non-transparent materials for these layers. As the substrate, rather than glass, we preferred silicon owing to its high thermal conductivity. In the cathode, we replaced the ITO with a silver (Ag) layer because the high reflectivity of Ag avoids the optical losses inside the device. Moreover, its consistency with the electrical structure and superior thermal conductivity make Ag an excellent contact layer (cathode).

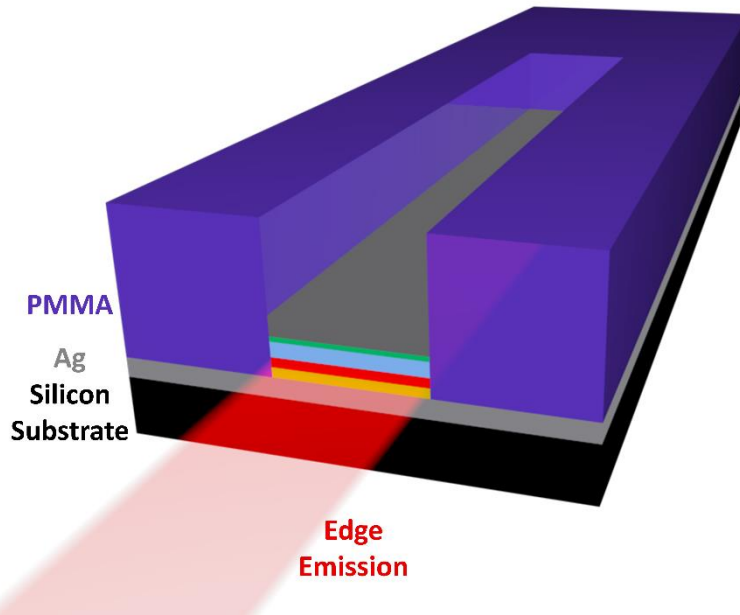


Figure 5.5: Schematic of edge-emitting CQW-LED architecture.

The remaining layers of the edge-emitting CQW-LED are the same as the ones used in the surface-emitting CQW-LED, although the sizes of the active device were modified. To direct the emission to the edge of the structure, we designed an LED in the shape of a thin strip with the sizes of $150 \times 900 \mu\text{m}^2$. We completed the waveguide by surrounding the active region with a polymethylmethacrylate (PMMA) layer that helps to trap the light inside the strip and dissipate the heat generated by the injected current. This waveguide architecture that confines the light

vertically by metal layers and laterally by the PMMA leads to the propagation of generated photons into the device edge. Figure 5.5 sketches the schematic of edge-emitting CQW-LED architecture.

We started the device fabrication by depositing an Ag layer on the silicon substrate using thermal evaporation at a deposition rate of $\sim 0.7 \text{ \AA/s}$. Then, we spin-coated 400 nm PMMA with a speed of 5,000 rpm. The sample was baked on a hot plate at 180 °C for 10 min. We patterned the thin strip on the PMMA layer utilizing the electron-beam lithography (EBL) technique. Since PMMA is also used as a positive electron-beam resist, the exposed regions were removed after dipping the sample into the developer solvent. Subsequently, we followed the same fabrication flow with the surface-emitting CQW-LED. Although the device layers are coated over the entire surface, PMMA blocks the current injection, and the device operates only in the strip-shaped active region.

After the fabrication, we measured the EL spectrum collected from the device edge using a spectrometer. As shown in Figure 5.6a, we collected the spectrum data for the voltage levels from 7 to 17 V. The peak electroluminescence intensity wavelength is 645 nm and FWHM is $\sim 32 \text{ nm}$. Comparing the peak wavelength and FWHM values to the ones in the surface-emitting device, we did not observe any significant change. This stability indicates that the waveguide architecture does not have a major effect on emission modes.

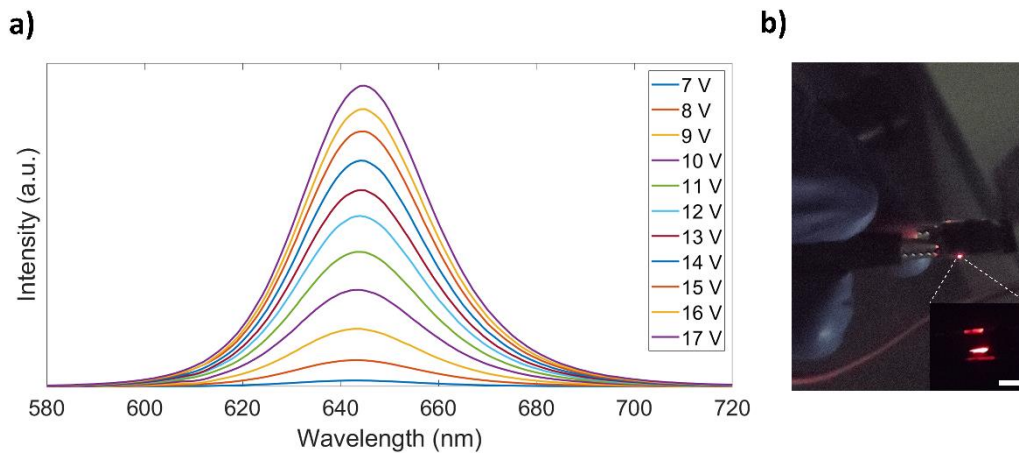


Figure 5.6: a) Evolution of EL spectrum of the edge-emitting CQW-LED with varying voltage levels. b) Photograph of the edge-emitting LED under a voltage bias. The inset is the optical microscopy image of the device edge with a 90° angle. The scale bar is 200 μm .

Figure 5.6b displays the photograph of our edge-emitting CQW-LED under a bias voltage of 10 V. Emission extracted from the edge of the device is clearly seen when there is a slight tilt between the sample and the observation point. The inset of this figure demonstrates the optical

microscopy image focused on the device edge with a precisely aligned perpendicular angle. Because the edge emission is observed only in the region where the PMMA layer is removed, this microscope image proves that the generated photons propagate inside the active device, and the penetration into the PMMA layer is negligible.

We measured the total current passing through the LED with an amperemeter. Dividing the total current value by the area of the active region, we calculated the current density of our strip-shaped CQW-LED. Figure 5.7a depicts the behavior of current density with rising bias voltages. Owing to the heat-dissipating features of our design, the current density reaches 5.6 A/cm^2 at 17 V without causing emission degradation, exhibiting a ~ 60 -fold increase compared to the surface-emitting CQW-LED.

We determined the luminance of the edge-emitting CQW-LED using a spectroradiometer following the same protocol that we applied for the luminance measurements of the surface-emitting device. The only difference during the measurement is the sample's orientation angle since its electroluminescence must be collected from the edge rather than the surface. As seen in Figure 5.7b, the emission intensity of our device starts to increase after the voltage level of $\sim 6.5 \text{ V}$. At 17 V, the LED demonstrates a luminance value of $\sim 20,400 \text{ cd/m}^2$.

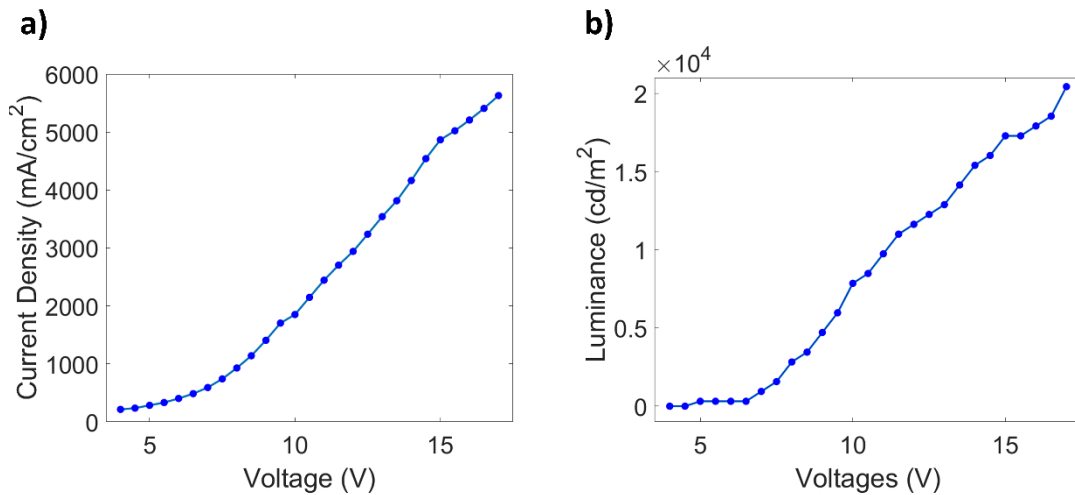


Figure 5.7: a) Voltage vs. current density and b) voltage vs. luminance graphs of the edge-emitting CQW-LED.

5.4 Summary

In summary, for the first time, we demonstrated an LED of CQWs with an emission directed into the edge of the device structure. We integrated an efficient conventional surface-emitting LED into a novel architecture, which avoids light extraction from the surface and directs all the generated photons into the device edge. By transforming the active device into an optical waveguide structure, we achieved collecting the emission with high brightness from the side of our device. Owing to the superior thermal conductivity of this architecture, we prevented the excess heating in the active region and enabled the high current density values up to 5.6 A/cm^2 , which is ~ 60 times more than the current density values of the surface-emitting device. This study exhibits that the CQWs are excellent materials for the emissive layers of edge-emitting LEDs, which can be utilized in several applications, such as photonic integrated circuits and new-generation displays. Additionally, the proposed architecture presents a great potential to demonstrate electrically driven amplified spontaneous emission (ASE) in CQWs thanks to its waveguide feature and ability to achieve high current densities.

Chapter 6

Conclusion

The utilization of semiconductors in light generation applications as the active layer paved the way for the large-scale manufacturing of light-emitting devices with low cost and high efficiency. Colloidal quantum wells (CQWs), a recently emerged quasi-2D subfamily of semiconductor nanocrystals, offer a significant potential to serve as an emissive layer to various light-emitting devices owing to their distinctive optical features. Combining CQWs with carefully designed optical waveguiding architectures within the light-emitting device structures, their optical characteristics can be tailored.

In this thesis, first, we demonstrated an all-solution-processed optically-pumped dual-color laser waveguide using CQWs as the gain media. Selecting red- and green-emitting CQWs with low threshold amplified spontaneous emission (ASE) and separating them by an optically transparent dielectric layer in their waveguide to suppress the otherwise detrimental nonradiative energy transfer, we showed dual-color ASE in CQWs. Then, placing this multilayered waveguiding structure into a microdisk cavity, lasing in red and green spectral regions was observed. This study indicates that CQWs are versatile and efficient materials for multicolor lasing applications.

The second study included in this thesis exhibits a directed edge emission from a slit-shaped light-emitting diode (LED) by building an active waveguide architecture. First, we fabricated an efficient conventional surface-emitting LED structure. Then, by altering the substrate and bottom electrode with non-transparent materials, we prevented surface emission and confined the emitted light in the active region. Finally, by modifying the lateral shape of the diode structure as a micro-scale thin optical waveguide, the emitted light is directed into the edge of the device. Therefore, for the first time, we demonstrated edge-emitting waveguide LED based on CQWs.

Despite all the achievements shown in this thesis, further studies are required to develop more advanced devices. For instance, our dual-color lasing device can be improved by adding a third color which lases in blue. Having lasing behavior in red, green, and blue spectral regions simultaneously, a white laser, which is highly desired for many applications such as laser displays, optical communication, and biomedical imaging, can be obtained. Additionally, the

fabrication parameters of our edge-emitting waveguide LED can be further optimized to reach ultra-high luminance values. Moreover, eliminating the optical losses inside the waveguide structure and increasing current density passing through the active region can enable the first electrically driven ASE based on CQWs.

In this thesis, Chapter 4 is based on the following publication:

- (1) **A. T. Isik**, F. Shabani, F. Isik, S. Kumar, S. Delikanli, H. V. Demir, “Simultaneous Dual-Color Amplified Spontaneous Emission and Lasing from Colloidal Quantum Well Gain Media in their Own Layered Waveguide and Cavity,” *Laser Photonics Rev.*, 2300091 (2023) [39].

During this thesis study, besides the works explained in Chapters 4 and 5, we contributed to the several publications listed below:

- (2) S. Foroutan-Barenji, F. Shabani, **A. T. Isik**, Z. Dikmen, H. V. Demir, “All-colloidal parity–time-symmetric microfiber lasers balanced between the gain of colloidal quantum wells and the loss of colloidal metal nanoparticles,” *Nanoscale* 14, 13755 (2022) [82].
- (3) H. D. Baruj, I. Bozkaya, B. Canimkurbey, **A. T. Isik**, F. Shabani, S. Delikanli, S. Shendre, O. Erdem, F. Isik, H. V. Demir, “Highly-Directional, Highly-Efficient Solution-Processed Light-Emitting Diodes of All-Face-Down Oriented Colloidal Quantum Well Self-Assembly,” *Small*, 2206582 (2023) [91].
- (4) S. Delikanli, B. Canimkurbey, P. L. Hernández-Martínez, F. Shabani, **A. T. Isik**, I. Ozkan, I. Bozkaya, T. Bozkaya, F. Isik, E. G. Durmusoglu, M. Izmir, H. Akgun, and H. V. Demir, “On the Rational Design of Core/(Multi)-Crown Type-II Heteronanoplatelets,” *J. Am. Chem. Soc.*, 145, 22, (2023) [92].
- (5) Z. Dikmen, **A. T. Isik**, I. Bozkaya, H. D. Baruj, B. Canimkurbey, F. Shabani, M. Ahmad, H. V. Demir, “Vertically oriented self-assembly of colloidal CdSe/CdZnS quantum wells controlled via hydrophilicity/lipophilicity balance: optical gain of quantum well stacks for amplified spontaneous emission and random lasing,” *Nanoscale*, 15, 9745-9751, (2023) [101].
- (6) F. Isik, S. Delikanli, E. G. Durmusoglu, **A. T. Isik**, F. Shabani, Hamed Dehghanpour Baruj, H. V. Demir, “"Giant" Colloidal Quantum Well Heterostructures of CdSe@CdS Core@Shell Nanoplatelets from 9.5 to 17.5 Monolayers in Thickness Enabling Ultra-High Gain Lasing,” in submission (2023) [102].

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