High Q factor modes in semiconductor nanoantennas for active nanophotonics and lasing

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HIGH Q FACTOR MODES IN SEMICONDUCTOR
NANOANTENNAS FOR ACTIVE NANOPHOTONICS
AND LASING

VASILII MYLNIKOV

School of Electrical & Electronic Engineering

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AND LASING**

VASILII MYLNIKOV

School of Electrical & Electronic Engineering

A thesis submitted to the Nanyang Technological University
in partial fulfillment of the requirement for the degree of
Doctor of Philosophy

2021

Statement of Originality

I hereby certify that the work embodied in this thesis is the result of original research, is free of plagiarised materials, and has not been submitted for a higher degree to any other University or Institution.

03.01.2021

Date

Vasilii Mylnikov

Supervisor Declaration Statement

I have reviewed the content and presentation style of this thesis and declare it is free of plagiarism and of sufficient grammatical clarity to be examined. To the best of my knowledge, the research and writing are those of the candidate except as acknowledged in the Author Attribution Statement. I confirm that the investigations were conducted in accord with the ethics policies and integrity standards of Nanyang Technological University and that the research data are presented honestly and without prejudice.

08/01/2021

Date

Prof. Hilmi Volkan Demir

Authorship Attribution Statement

This thesis contains material from 1 paper published in the following peer-reviewed journal in which I am listed as an author.

Chapter 2 (plus partially other sections of the thesis) is published as:

Mylnikov, V.; Ha, S. T.; Pan, Z.; Valuckas, V.; Paniagua-Domínguez, R.; Demir, H. V.; Kuznetsov, A. I. Lasing Action in Single Subwavelength Particles Supporting Supercavity Modes. *ACS Nano* **2020**, *14*, 7338–7346. © 2020 American Chemical Society.

The contributions of the co-authors are as follows:

R.P.-D. and A.I.K. conceived the idea. V.M. performed numerical simulations. Z.P. performed epitaxial lift-off of the GaAs film, its bonding to the quartz substrate, and etching. V.M. performed etching, electron-beam lithography, and etching after EBL. V.V. performed SEM measurements. S.T.H. constructed the optical setup, performed all optical measurements and their data processing. All authors discussed the results. V.M. wrote the first draft of the manuscript and all authors worked on it. R.P.-D., H.V.D., and A.I.K. coordinated and supervised the work.

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Date

Vasili Mylnikov

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Summary, Motivations, and Objectives

In the past decade, resonant dielectric nanoantennas have emerged as a promising platform for nanophotonic applications. Low Ohmic losses and CMOS compatibility, accompanying small form factors, are the key advantages of these systems that make them particularly attractive for industrial applications compared to prior designs based on plasmonic metals.¹ In recent years, significant efforts are being made in combining dielectric nanoantenna concepts with active material platforms² to extend this promising approach toward efficient directional light sources. Dielectric-based nanocavity lasers are promising devices for sensing in biomedical applications and on-chip light sources in the future photonic circuits, which can be used in the next-generation high-performance computers (see [Figure 0.1](#)).

Figure 0.1. Semiconductor microlasers used for ultrasensitive detection (A) and photonic circuit (B). These are some of the most attractive motivations for dielectric-based nanocavity lasers. Panels are adapted with permission from (A) ref 3, © 2011 Springer Nature; (B) ref 4, © 2009 IEEE.

By now, the use of dielectric nanoantennas in emission processes has been mainly focused on enhancing fluorescence from molecules,⁵⁻¹⁰ enhancing the Raman signal¹¹⁻¹⁴ and enhancing photoluminescence (PL) from the nanoantenna material itself (see [Figures 0.2A–C](#)).¹⁵⁻¹⁸ Due to the limited quality factors (Q factors) of low-order modes excited in dielectric

nanoantennas, lasing has not been demonstrated yet for particle sizes significantly smaller than the wavelength, with the recent exceptions of micrometer-sized systems in which subwavelength nanoantennas are arranged in lattices (see Figure 0.2D).^{19, 20} Hence, besides strongly dissipative systems involving metals,^{2, 21} on-chip lasers have been mostly restricted to spatially extended systems with sizes on the order of a few microns.

Figure 0.2. Prior art. The use of dielectric nanoantennas in emission processes. (A) Enhancing fluorescence from molecules. (B) Enhancing the Raman signal. (C) Enhancing photoluminescence (PL) from the nanoantenna material. (D) Lasing in nanoparticle array. Panels are adapted with permission from (A) ref 5, © 2018 John Wiley & Sons; (B) ref 14, © 2020 American Chemical Society; (C) ref 15, © 2018 Royal Society of Chemistry; (D) ref 19, © 2018 Springer Nature.

The smallest fully dielectric lasers were experimentally demonstrated using suspended disk structures.^{22, 23} High-order Mie-type modes (*i.e.*, whispering gallery modes) were used in those designs to achieve lasing. In this regard, the smallest nanolaser reported consisted of a disk on top of a pedestal with a diameter of 627 nm, operating in the fourth-order Mie resonance and emitting at a wavelength of 870 nm, which corresponds to a size/wavelength

ratio $\zeta = 0.72$.²³ Very recently, one of the smallest, fully dielectric single-particle-based laser has been demonstrated using perovskites in a nanocube configuration.²⁴ This design also uses the fourth-order Mie-type mode and has a slightly higher size/wavelength ratio of $\zeta = 0.79$ but operates at room temperature. Further reduction of size (*i.e.*, in disk diameter) is challenging for two different reasons. First, using smaller resonators while keeping the same operating wavelength implies using lower order modes. Those present higher radiative losses, which leads to a drop in the Q factor. Second, smaller particles have larger surface-to-volume ratios, which increases the effect of surface recombination and roughness, resulting in higher losses that make the gain-loss compensation even more challenging. While the second is more difficult to overcome without improving the fabrication processes and/or including some surface passivation methods, the first still allows some improvement from the design perspective. Thus, different approaches need to be implemented to improve the quality of the cavities to achieve lasing in smaller resonators.

A recent promising approach to boost the Q factor of a system is engineering it to support a special type of resonant mode called bound state in the continuum (BIC). BICs are nonradiative states, thus perfectly spatially confined, which nevertheless coexist with the continuum of radiative states (*i.e.*, despite the presence of available radiation channels).²⁵ BICs are a general wave phenomenon, first reported in electronic systems and subsequently extended to photonic ones,²⁵ and are true eigenmodes of the system (in fact, often referred to as embedded eigenstates). Strictly speaking, for any system comprising regular materials with $\epsilon \neq 0$, $\mu \neq 0$, $\epsilon \neq \pm \infty$, or $\mu \neq \pm \infty$, BICs can only be supported if the system is infinite, and their Q factors are infinite.²⁵ However, parameters of a finite system can be tuned close to the condition of BICs in the corresponding infinite system.²⁶ The obtained quasi-BICs are often referred to in the literature as supercavity modes and possess finite but high Q factors.²⁶ This makes supercavity modes or quasi-BICs useful for many photonic applications,^{25, 27} including, for example, sensing^{28, 29} and high-harmonic generation.³⁰⁻³⁴

Regarding the use of BICs for lasing, one of the first quasi-BIC lasers was demonstrated in 1985. It supported a symmetry-protected quasi-BIC by suppressing radiation into the transverse directions in a 1D distributed feedback laser cavity.³⁵ After that, similar quasi-BIC principles were demonstrated in many photonic crystal surface-emitting laser devices.²⁵ In recent years, quasi-BIC-based lasing results have been shown for on-chip systems based on nanoparticle arrays.^{19, 20} Going beyond the BIC supported by spatially extended systems toward single-particle configurations, it should be mentioned that a spatially confined system can only support a true BIC if it contains some region in which the permittivity tends to zero.^{36, 37} Nevertheless, several recent reports have theoretically predicted that it should, still, be possible to obtain, if not a true BIC, but a quasi-BIC or a supercavity mode in such a simple system as a single dielectric nanocylinder.^{31, 38, 39} This quasi-BIC has a Friedrich–Wintgen nature and allows some degree of radiation to escape the system. Thus, by reciprocity, it is also possible to excite it by external sources. Recent experimental studies have, indeed, demonstrated the excitation of these quasi-BICs in a single cylinder at GHz⁴⁰ and optical³³ frequencies. However, no experimental results of lasing in a supercavity regime for a single semiconductor nanoparticle have been shown so far.

That motivated me to investigate supercavity lasing possibilities in a single semiconductor nanocylinder and demonstrate the smallest fully dielectric nanolaser. After the lower size limit for the lasing realization in a single nanoparticle was reached in my research, I was motivated to investigate that limit for nanolasers consisting of several nanoparticles using lower-order azimuthal modes for lasing. Coupled particles give an opportunity for directivity control. Thus, the following objectives can be outlined for my thesis:

Numerical optimization, fabrication, and optical characterization of

1. Single-particle-based lasers
2. Nanoparticle structures-based lasers

The numerical optimization or design part focuses on obtaining optimal

geometrical parameters of the nanostructures of interest, *i.e.*, achieving the maximum Q factor (minimal radiative losses) in the smallest possible nanostructure size. High Q factor is the characteristic that is critical for gain-loss compensation in the subsequent lasing experiment. At the same time, the optimization procedure must take into account the actual experimental constraints, as detailed later.

Fabrication of the nanostructures involves developing and optimizing the nanofabrication processes to realize semiconductor nanoantennas on a low-index substrate. It combines the techniques of epitaxial lift-off, electron-beam lithography, and reactive ion etching.

The optical characterization includes dark-field scattering measurements of the fabricated nanoantennas and optically pumped lasing experiments at cryogenic temperatures.

Major contribution of the Thesis

In this thesis, elaborating on the BIC concept, I report the first single nanoparticle laser working in a supercavity regime. This also enables the realization of the smallest fully dielectric nanolaser with a record-low size/wavelength ratio $\zeta = 0.6$. It is the first fully semiconductor laser using the third azimuthal order mode. Moreover, we have demonstrated lasing for a particle on a substrate. As compared to previous approaches, the present design does not need the particle to be suspended in the air using a pedestal and does not rely on modes of high azimuthal orders, which increases the robustness of the device.

Along with other theoretical and experimental accomplishments reported in this thesis, this is the most important contribution of my thesis, the realization of lasing action in the smallest fully dielectric nanoparticle, as characterized by means of size/wavelength ratio. This significant accomplishment opens a way for the realization of smaller low-loss dielectric nanolasers that might find applications in future photonic circuitry, among others.

Organization of the Thesis

In accordance with the thesis's major contribution, I find it essential to survey the prior art regarding micro- and nano-sized lasers' development and their recent advances and applications. Secondly, bound states in the continuum, along with their types, nature, and applications, are reviewed.

The second and third chapters contain the actual work, performed either solely by me or together with collaborators from my research group (as detailed in the corresponding section above, where their names and contributions can be found and acknowledged by the use of "we," "our," *etc.* throughout the thesis).

The second chapter contains our research about lasing action in a single dielectric nanoparticle of a record-low size/wavelength ratio enabled by the concept of quasi-bound states in the continuum. Along with numerical simulations and optical experiments, I report the outlook for further studies about light outcoupling from the nanolaser.

The third chapter comprises the research about obtaining the lower size limit for lasing in multiple particles. There, 2-nanoparticle interactions are studied in detail.

The fourth chapter of the thesis highlights its major conclusions from each chapter and propositions for future research. The fifth chapter contains the Methods.

Chapter 1

1 Introduction

1.1 Micro- and Nano-Sized Lasers

1.1.1 Evolution in Development of Small Lasers

In this part of the thesis, I discuss the types of micro- and nanoscale lasers, their operation principles, and their limitations. The story of microscale lasers started from the invention of vertical-cavity surface-emitting laser (VCSEL) around four decades ago.⁴¹ VCSELs have found many applications, ranging from optical interconnections to laser printing, data storage, and more. A typical VCSEL has two distributed Bragg reflector (DBR) mirrors with the gain medium between them⁴² (see [Figure 1.1A](#)). DBR mirrors are designed in a way that the reflectivity to the lasing wavelength is very high (~ 0.99). Upper and lower Bragg mirrors usually are *p*-type and *n*-type semiconductors, respectively. Highly reflective mirrors lead to high quality factor cavities, having a large photon lifetime (on the order of several picoseconds). In essence, a VCSEL is a Fabry–Perot type laser. It is well known that Fabry–Perot modes follow the equation $L = m\lambda/2n$, where L is the cavity length in the longitudinal direction, λ is the wavelength in vacuum, m is an integer number and n is the refractive index of the medium. Consequently, the size of the active region needs to be longer than $\lambda/2n$ to support at least one mode.

It could, of course, be bigger as long as the undesired modes do not overlap, spectrally, with the gain. The laser size in the transverse direction is several wavelengths in order to keep the modal gain high enough for compensation of the losses caused by surface roughness. The modal gain G_m is directly proportional to material gain G_a :⁴¹ $G_m = \Gamma G_a$, where Γ is the confinement factor (which is the fractional overlap of the lasing mode volume and the volume of the active medium).⁴³ The laser's size in the longitudinal direction usually reaches several microns because of the thick Bragg reflectors required for having a high reflection coefficient.

The first demonstration of a VCSEL was at cryogenic temperatures (liquid nitrogen) and used GaInAsP as an active layer.⁴⁴ The emission wavelength of the laser was 1.18 μm , and the laser was pumped by 400 nanoseconds current pulses having a pulse repetition frequency of 1000 pulses per second. Au-based mirrors were used in that setup.

It is important to note that commercial applications require a small laser operation under continuous-wave (CW) electrical pumping at room temperature (RT). A decade after the first VCSEL was made, one of the first CW electrically pumped RT VCSEL was demonstrated. That laser was based on GaAs as active material⁴⁵ and used Bragg mirrors and better fabrication techniques to realize the active layer. The laser had a room-temperature operation and a threshold current of a few dozen mA, which was nearly two orders lower than in the first VCSEL demonstration. The lasing wavelength was 895 nm. Later on, the threshold current was lowered down, first to the order of hundreds of μA ,⁴⁶ and then to several μA ,⁴⁷ with a simultaneous decrease of the transverse size of the active region in VCSEL from 10 to 3 μm . Currently, VCSELs are widely used in high-speed optical fiber communications.⁴⁸ VCSELs is one of the best solutions for short-distance optical links.⁴⁹ Compared to edge-emitting lasers (which are hundreds of micrometers in size and have an energy consumption of $\sim$ hundreds of pJ per bit), VCSELs have small sizes and consume less energy. They are typically several micrometers in size with an energy consumption of $\sim$ hundreds of fJ per bit.⁴⁹

The development of micro and nanoscale lasers, with potential applications in chip-to-chip and on-chip data transmission in photonic circuits, is reviewed in refs 41, 49. The major types of microlasers are presented in Figure 1.1. In the laser miniaturization process, several issues and challenges arise. Among the main ones is the deterioration of the device lifetime,⁴¹ affected by the strong surface recombination arising from surface roughness induced by etching during the fabrication. That is a particularly acute issue for small lasers because the surface-to-volume ratio is high in these devices.

The next milestone in the way of laser miniaturization was arguably the invention of microdisk lasers (see Figure 1.1B), which have already found their first applications in sensing.^{3, 50} Light confinement in the microdisk cavity is based on whispering gallery modes (WGM). Electromagnetic waves travel along the circumference (more generally the perimeter) of the structure and experience total internal reflections, which leads to very high Q factors (up to 10^7 , as obtained for toroidal microlasers based on silicon).⁵¹ The size of these lasers in the vertical direction can be smaller than the $\lambda/2n$ necessary for VCSELs because the nature of the modes in microdisk laser is different. In the latter case, the critical dimension defining the optical mode is rather the diameter.

One of the first microdisk lasers was demonstrated almost three decades ago. In that original design, the disk was 5 μm in diameter and 150 nm in thickness.⁵² It was made of InGaAs multiple quantum wells separated by InGaAsP barriers. The laser was optically pumped and had a 1.5 μm laser emission wavelength. The microdisk cavity was standing on an InP pedestal, closely mimicking a disk suspended in the air. Lasing was achieved using a whispering gallery mode of high azimuthal order. Smaller microdisk lasers with continuous-wave electrical pumping at room temperature were subsequently demonstrated nearly a decade later. For example, in ref 53, such microdisk laser was based on a multi-quantum well structure made of GaInAsP/InP and had a lasing wavelength of 1.56 μm . The laser diameter was 2.7 μm , and the threshold current was 40 μA . Decreasing the transverse size of the microdisk cavity leads to higher radiative losses and, therefore,

higher lasing thresholds, making a demonstration of lasing extremely difficult. Nevertheless, submicron-sized disk lasers based on semiconductors have also been demonstrated, as discussed below.

Figure 1.1. Microlasers. (A) Vertical-cavity surface-emitting laser. (B) Microdisk laser. (C) Photonic crystal laser. Light confinement by the total internal reflection and distributed Bragg reflection is highlighted by blue and green arrows, respectively. Panels are adapted with permission from (A) ref [54](#), © 2017 Chinese Laser Press; (B) ref [55](#), © 2019 Optical Society of America; (C) ref [56](#), © 1999 The American Association for the Advancement of Science.

A different class of microlasers is based on photonic crystals (PhCs), thus known as photonic crystal lasers. One of the first experimental demonstrations of this type of lasers was made around two decades ago.⁵⁶ Typical photonic crystal laser has a two-dimensional structure (as shown, for example, in [Figure 1.1C](#)). Laser thickness can be as small as $\lambda/2n$ for light confinement in the transverse direction *via* total internal reflection (see [Figure 1.1C](#)). Modes are localized in the longitudinal direction using the distributed Bragg reflection. A laser cavity is usually formed by introducing a defect in the photonic crystal (for example, by removing one of the holes, as shown in [Figure 1.1C](#)) to create a discrete state in the otherwise forbidden bandgap of the PhC.

Using these concepts, prior art in the literature⁵⁶ reported PhC lasers with thickness $\lambda/2$ and formed by a hexagonal array of air holes etched in the slab (see [Figure 1.1C](#)). One of the limitations of PhC is that the extent (size) of the photonic crystal must be large, on the order of several micrometers, to ensure

a proper formation of the bandgap *via* Bragg scattering.⁴¹ The gain in that photonic crystal laser⁵⁶ was provided by four layers of InGaAsP quantum wells separated by 1.22 μm bandgap material barriers. The laser had a pulsed pumping and operated at a temperature 143 K. Q factors on the order of 10^4 were obtained at $\lambda = 1.5 \mu\text{m}$, which is the order of magnitude of Q factors obtained for typical photonic crystal lasers. Pinning these results, some of the first continuous-wave electrically pumped room-temperature photonic crystal lasers were subsequently demonstrated.^{57, 58}

Another breakthrough in the field was the invention of metallo-dielectric nanolasers. The modes supported by these structures can be divided into two main groups: plasmonic modes and conventional resonant modes confined in the nanocavity due to reflections from the metal surface.^{59, 60} Plasmonic nanolasers have a higher mode overlap with metal, which leads to high Joule losses compared to conventional-mode-based metallo-dielectric nanolasers. Thus, in general, high Q factors and low threshold operation are achievable in the conventional metallo-dielectric lasers based on reflections compared to plasmonic nanolasers.⁵⁹ The main advantage of using the latter is subwavelength confinement of the lasing mode (a tight concentration of the electromagnetic energy in a low modal volume). This means smaller laser sizes, which will be discussed further in the text.

Non-plasmon metallo-dielectric nanolasers use metal as mirrors. Compared to fully dielectric nanolasers, that allows to increase the confinement factor Γ , which results in a higher modal gain.⁴¹ The higher gain allows for compensating high cavity losses, which arise when decreasing the laser size. All in all, by the use of metal cladding, it is possible to realize smaller lasers.

The first electrically pumped conventional-mode-based metallo-dielectric nanolaser was demonstrated by Hill *et al.* in 2007.⁴³ In that work, a semiconductor heterostructure was covered by a layer of gold. InGaAs was used as the active material, and the emission wavelength was 1.4 μm . Q factors on the order of 140 were obtained at 77 K in that study. Lasing was achieved by continuous-wave electrical pumping with a threshold of around

10 μA . The laser cavity consisted of a cylindrical pillar of height of $\sim 2\text{ }\mu\text{m}$ and diameter as low as $\sim 260\text{ nm}$. Similar concepts were implemented to obtain lasing using other cavity configurations. As an example, a metallo-dielectric laser was demonstrated using whispering gallery modes.⁵⁹ In that study, InGaAsP nanodisk was used as an active medium (see Figure 1.2A). One of the strategies for reducing Joule losses in metallo-dielectric nanolaser is using a dielectric spacer to isolate the supported mode from the metal. Indeed, a silica layer between the metal (aluminium) and the active material was used to reduce the overlap of the mode with the metal for reduction of the Joule losses. In that way, the gain threshold could be reduced, allowing lasing at room temperature.^{60, 61} The transverse and longitudinal dimensions of the laser⁵⁹ were 490 and 480 nm, respectively. In that research, the laser was excited by a pulsed optical pump and had a gain threshold power on the order of 70 kW/cm^2 . The laser operated at $1.43\text{ }\mu\text{m}$ wavelength and had a Q factor on the order of 10^3 . One of the first room-temperature continuous-wave electrically pumped metallo-dielectric non-plasmon nanolasers was finally demonstrated in 2013.⁶²

Figure 1.2. Metallo-dielectric nanolasers. (A) Non-plasmon-based nanodisk laser. (B) Plasmonic nanowire laser. (C) Spaser. Panels are adapted with permission from (A) ref 59, © 2010 Springer Nature; (B, C) refs 63, 64, © 2009 Springer Nature.

As mentioned earlier, plasmonic nanolasers allow better confinement than the above-discussed conventional metallo-dielectric nanolasers, which operate far from the plasmon resonance using metals as mere mirrors. Instead, plasmonic nanolasers use free electrons to confine the field tightly. This

implies better confinement and a higher mode overlap with the metal and, therefore, higher Joule dissipation and, ultimately, higher losses. Broadly speaking, there are two types of plasmonic nanolasers. The first kind uses propagating surface plasmon polariton (SPP) modes, which are surface waves that propagate along the interface between a dielectric and a metal. They are confined to the interface, *i.e.*, they decay evanescently away from it.⁶⁵ The mode size in this direction can be more than 100 times smaller than the diffraction limit,⁶⁶ which allows a significant decrease of nanolaser's transverse dimension based on SPPs compared to the setups discussed above.

The first demonstration of this kind of plasmonic nanolaser is attributed to Oulton *et al.* in 2009 (see [Figure 1.2B](#)).⁶³ The lasing mode in that device was formed by hybridizing the surface plasmon polariton and the Fabry–Perot mode of the semiconductor nanowire. That allowed to obtain subwavelength confinement of the mode in the vertical direction (diameter ~ 100 nm) and long-distance propagation in the longitudinal direction (length ~ 10 μm).⁶⁶ The device in that work consisted of a CdS semiconductor nanowire on an Ag film having an interlayer of MgF_2 between them (see [Figure 1.2B](#)). Lasing was obtained at a wavelength of 489 nm, with a threshold pump intensity on the order of 100 MW/cm^2 . The device was pumped in a pulsed regime using a femtosecond laser and operated at low temperatures (below 10 K). A good example of the implementation of somehow analogous lasing concept but using hybridization of modes of a different nature was demonstrated by Perahia *et al.* the same year.⁶⁷ In that study, a lasing mode was obtained by hybridizing a surface plasmon polariton and a WGM mode supported by a dielectric cylinder.

The second kind of plasmonic nanolasers is based on localized surface plasmon resonance (LSPR) modes. These devices are sometimes referred to as spasers (surface plasmon amplification by stimulated emission of radiation),⁶⁸ although sometimes the same name is also coined for those lasers using the SPP modes. Contrary to SPP-based, in LSPR-based nanolasers, the plasmon modes are collective oscillations of electrons bound

to a closed geometry, typically of subwavelength size in all dimensions. Their operation is also based on having the active material close to the region of surface plasmon oscillations.

Arguably, the first experimental demonstration of an LSPR-based nanolaser was carried out by Noginov *et al.* in 2009.⁶⁴ In that work, the spaser had a core-shell structure (gold core and silica shell which was doped by dye and acted as an active medium of the laser, see Figure 1.2C). The spaser diameter was only ~ 45 nm, which makes it one of the smallest nanolasers ever made. A spaser of a smaller size (diameter ~ 20 nm) was demonstrated in 2017.⁶⁹ The emission wavelength of the spaser from ref 64 was 530 nm. The laser exhibited a Q factor of around 15. Due to the strong Ohmic and radiation losses in this kind of LSPR-based lasers, the active material's gain must be very high to compensate the losses.

Figure 1.3. The smallest fully dielectric nanolasers in terms of size-to-wavelength ratio $\zeta = D/\lambda$. Panels are adapted with permission from (A) ref 23, © 2009 American Institute of Physics; (B) ref 22, © 2007 American Institute of Physics.

In parallel to the studies on metallo-dielectric lasers, and due to the large Joule losses of these devices, a significant effort to reduce the size of purely dielectric lasers has been made. To date, one of the smallest dielectric nanolasers compared to the operation wavelength was demonstrated by Song *et al.*²³ The size-to-wavelength ratio of the laser was $\zeta = 0.72$. The laser cavity in that work consisted of a GaAs disk (see Figure 1.3A). Again, the disk was supported by a thin pedestal, made of AlGaAs in that case. The pedestal

was used in order to lower the radiation leakage into the GaAs substrate, thus increasing the quality factor of the cavity by closely mimicking the case of a free-standing disk. Inside the GaAs disk, there were 6 multi-layers of quantum dots made of InAs and spaced equally, having 25 nm of gallium arsenide barriers in between. The smallest laser demonstrated in that study had a diameter of 627 nm and a height of 265 nm. The lasing experiments were carried out at a low temperature of 10 K (a liquid helium cryostat was used). The laser was operating under pulsed optical pumping at 800 nm wavelength, having a pulse width of 200 fs and a pulse repetition frequency of 76 MHz. The laser had a threshold power $\sim 200 \mu\text{W}$. Experimentally shown Q factor was around 500 for an operating wavelength of 870 nm. The azimuthal order of the mode used was $i = 4$, being one of the lowest reported so far. Simulation results obtained in that work showed a significant decrease of the Q factor from 10^4 to 10^2 when the azimuthal mode number went from 6 to 3. A limited Q factor associated with low-order azimuthal modes is the main reason why in most results reported to date, lasers still operate using modes of orders larger than 4. That drop in the Q factor is due to a significant increase in the radiation losses (an increase of the light leakage through the disk boundary).

Several other studies reported similar results. In particular, the work by Zhang *et al.* had demonstrated small disk lasers as well.²² The size-to-wavelength ratio of the laser was $\zeta = 1$. It had a similar structure as the nanolaser mentioned above (a disk was on top of a pedestal, see Figure 1.3B). The laser's active medium consisted of two InGaP quantum wells and three InGaAlP barriers. The laser operated under pulsed pump excitation at room temperature. The lasing threshold was $\sim 50 \mu\text{W}$. The smallest laser demonstrated in that work had a diameter of 645 nm. In that case, however, the emission wavelength was $\sim 645 \text{ nm}$ (similar to the size). That wavelength was smaller than that of the laser mentioned above. Thus, the reported Q factor was larger, on the order of 1000.

To finalize the description of current approaches to obtain micro- and nano-scaled lasers based on purely dielectric structures, it is worth mentioning

those based on Fabry–Perot resonances in vertically standing dielectric nanowires and nanopillars. A good example of these is the laser demonstrated by Chen *et al.* in 2011.⁷⁰ In that work, the authors produced a nanolaser based on a vertically standing hexagonal nanopillar grown on a silicon substrate. Specifically, the nanopillar had a core-shell structure with an InGaAs core (active medium) and a GaAs shell (passivation layer). The pillar diameter was close to 600 nm, and the height was on the order of a few microns. The laser worked under pulsed optical pumping at room temperature. And the experimentally shown Q factor was around 200 for an operational wavelength near 950 nm.

In conclusion, vertical-cavity surface-emitting lasers, microdisk lasers, and photonic crystal lasers have relatively big sizes (on the micrometer scale). Non-plasmonic metallo-dielectric lasers, plasmonic lasers, and spasers are lossy. Hence, in this thesis, I focus on fully dielectric nanolasers, which have lower Joule losses and relatively small sizes, which makes them promising candidates for on-chip light sources.

1.1.2 Challenges of Small Laser Integration

It is important to note that commercial applications (where a small laser is integrated into a photonic circuit) require the laser operation under continuous-wave electrical pumping at room temperature. In general, optical pumping is replaced by electrical current injection using the concept of a laser diode. The laser diode is a $p-i-n$ junction or a $p-n$ junction combined with a laser resonator.⁷¹ Its structure consists of the gain medium (intrinsic region) sandwiched between p - and n -type semiconductors. This structure is embedded between metal contacts used for the current injection. The electrons are injected from the n -doped semiconductor, while the holes are injected from the p -doped semiconductor into the gain medium. A potential barrier on the interface between the active layer and the p -doped heterolayer prevents the electrons from moving into the p -doped semiconductor. The same mechanism prohibits the holes from passing into the n -doped semiconductor. Thus, the electrons and the holes injected recombine primarily in the active layer (intrinsic region), resulting in the emission of radiation.⁷¹

By the use of that concept and its slight variations, small laser operation under continuous-wave electrical pumping at room temperature was realized in many studies for vertical-cavity surface-emitting lasers,⁷²⁻⁷⁵ microdisk lasers,^{4, 53, 76-78} photonic crystal lasers,^{57, 58, 79} and conventional-mode-based metallo-dielectric lasers.^{62, 80}

A single nanodisk laser is studied in my research. Thus, I discuss the concepts and challenges of electrical current injection primarily for disk lasers and other small lasers. Current injection is carried out using the $p-i-n$ junction concept mentioned above (e.g., a disk laser in ref [53](#)). Acute challenges of the continuous-wave (CW) electrical injection at room temperature (RT) in small lasers are about low thermal conductance, high electrical resistance, optical absorption losses, and poor gain properties at room temperature.^{76, 81} It is easier to obtain lasing in a pulsed regime or at low temperatures because these challenges subside. That is why lasing under these conditions is carried

out in many studies for the proof of concepts. One of the most important things for small laser integration is cost-effectiveness. For that, energy per bit must be minimized to get a low power consumption (see ref 49 for more details).

A good thermal conductance is necessary for good heat dissipation (which helps to avoid overheating). That is important for increasing laser lifetime. Some of the reasons for low thermal conductance are thick contact p - and n -layers and a thick dielectric spacer layer. Thermal conductance can be increased by reducing the thickness of layers mentioned above or by introducing a thermal *via*.⁷⁶ Straightforward thickness reduction of the p -type heterolayer cannot be carried out for thin-film lasers. That is due to significant optical absorption losses in the p -doped semiconductors.⁸² Thus, other approaches need to be implemented for increasing the thermal conductance, as discussed further in the text. In addition, metal contacts in small electrically driven lasers act like heat sinks, reducing the heating of the laser cavity, which increases its lifetime.⁴⁹ It is also important to obtain low electrical resistance in the p -type heterolayer.⁷⁶

The above-mentioned challenges of current injection and operating temperature can be overcome using a tunnel junction on the p -side of the p - i - n junction (see Figure 1.4A).⁷⁶ The tunnel junction is used in the reversed-biased configuration. Holes are generated in the junction's p -region due to electrons tunneling from the valence band of the p -region to the n -region of the tunnel junction.⁸² This approach allows to reduce the optical loss, lower the electrical resistance while having a thin p -side of the p - i - n junction with sufficient thermal conductance for stable device operation at RT CW electrical pumping.^{4, 76}

A typical structure of optical interconnect using a disk laser was demonstrated in ref 4. The microdisk laser used InAsP multi-quantum-well-based active medium. The laser was vertically coupled to a silicon-on-insulator waveguide. The radiation from the waveguide was received by a membrane photodetector (see Figure 1.4B). Coupling efficiency can be increased by reducing the thickness of the bonding layer.⁷⁶

Another promising way to outcouple the radiation from the disk laser into the waveguide is by using a radial coupling.⁸³ The radial coupling can be advantageous compared to the above-discussed vertical coupling and regular azimuthal coupling (e.g., ref 84). During the azimuthal coupling between a disk laser and a waveguide, the coupling directions of the clockwise and counterclockwise modes of the cylinder are different. In the case of an ideal cylinder, these two modes are degenerate.⁷⁶ This means that the detector, placed at one of the waveguide's ends, receives only half of the outcoupled power. Radial coupling of radiation from a disk laser operating at room temperature under electrical pumping was successfully demonstrated in ref 83. Besides the above-discussed outcoupling strategies, interconnect characteristics can be improved using a design from ref 77, where a microspiral disk laser is gaplessly coupled into a waveguide.

It is important to highlight the electromagnetic interference issue arising when chip-scale integration of the photonic link mentioned above⁴ is considered. Electromagnetic interference or inter-waveguide crosstalk can damage the performance of densely packed optical interconnects. Approaches like nanophotonic cloaking obtained from the inverse design⁸⁵ (see Figure 1.4C) or periodic nano-strip array inserted in between the adjacent waveguides⁸⁶ can be utilized to reduce the effect of signal crosstalk.⁸⁷ This will allow to increase the density of photonic integrated circuit and reduce its area.

Figure 1.4. Challenges of small laser integration. (A) Typical structure of continuous-wave electrically driven microdisk laser operating at room temperature. (B) Photonic link using a disk laser. (C) Nanophotonic cloaking allowing to reduce the effects of signal crosstalk or electromagnetic interference. (D) One of the smallest lasers operating at room temperature using continuous-wave electrical pumping. (E) Device platform implemented in nanoLED using electrical pumping. Panels are adapted with permission from (A) ref 76, © 2007 Optical Society of America; (B) ref 4, © 2009 IEEE; (C) ref 85, Creative Commons; (D) ref 62, © 2013 Optical Society of America; (E) ref 88, Creative Commons.

It is essential to note that the above-mentioned concepts for electrical injection, room-temperature operation, and light outcoupling were applied for

microdisk lasers, which have a relatively big size. However, these concepts can be useful for lasing in smaller cavities. If such a submicron fully semiconductor disk laser operation is too challenging to obtain in further research, conventional-mode-based metallo-dielectric nanolasers can be considered as a promising solution. Moreover, one of the smallest disk lasers (cavity volume $\sim 3 \mu\text{m}^3$) using CW electrical pumping made so far is based on the metallo-dielectric structure utilizing non-plasmonic modes.⁸¹ The laser made use of InP/InGaAs/InP disk core and amorphous Al_2O_3 spacer between the core and the metal shield. By the use of a silver metal shield as a mirror, it was possible to reduce the gain volume (size of the laser). Joule losses were reduced by introducing a dielectric spacer (amorphous Al_2O_3) between the active medium and the metal shield. This spacer can also act as a passivation layer to enhance the gain properties.⁸¹ The laser demonstrated in this study had a stable operation at cryogenic temperature 77 K. At greater temperatures, the material gain decreases, making gain-loss compensation challenging. Because of that, the device did not reach the lasing threshold at room temperature. The gain broadened and blue-shifted at high pumping levels, resulting in parasitic mode competition, which did not allow to obtain lasing at room temperature and lasing for high pumping levels at cryogenic temperatures. Self-heating was not the obstacle for obtaining lasing at room temperatures in this device. Lasing at room temperature was not achieved due to insufficient material gain.⁸¹

The smallest lasers operating at room temperature by using CW electrical pumping were obtained in metal-clad cavities.⁴⁹ One of the lasers of this kind had a cavity volume $\sim 2.7 \mu\text{m}^3$ and used InP/InGaAs/InP core and SiN spacer between the core and a silver shell (as shown in [Figure 1.4D](#)).⁶² The design was optimized by adjusting the spacer's thickness and improving the fabrication process, which allowed to obtain lasing at RT using CW electrical driving. The etching was optimized to reduce the pillar tilting. HSQ resist was used instead of PMMA resist to decrease the edge roughness and associated cavity losses. The silver deposition technique was improved to achieve near single-crystal quality, which was essential to reduce Joule losses. Surface

treatment using oxygen plasma and dilute H_3PO_4 was carried out to reduce surface recombination for enhancing gain properties. The thickness of the SiN spacer was optimized, as well. The too thick spacer ~ 120 nm resulted in low thermal conductance, which was not sufficient for good heat dissipation, not allowing RT lasing. Too thin spacer ~ 10 nm significantly increased Joule losses, which lowered the Q factor making gain-loss compensation more challenging. A good trade-off between those two effects was obtained at moderate spacer thickness ~ 30 nm, which permitted lasing at RT under CW electrical pumping. The lasing threshold was ~ 1 mA. Lasing was also obtained at high pumping powers up to ~ 2 mA. At pumping powers greater than that lasing was not possible due to overheating. Dielectric spacer breakdown at higher voltages is another obstacle that has to be taken into account for such lasers. Using these techniques for improving the nanofabrication combined with higher Q nanocavities will allow achieving lasing at RT for CW electrical pumping in submicron lasers.

Not so long ago, an electrically pumped nanopillar light-emitting diode (LED) coupled to a waveguide was demonstrated using a similar InP/InGaAs/InP structure discussed above.⁸⁸ Although LEDs use spontaneous emission for operation, the platform of the device (shown in [Figure 1.4E](#)) can be utilized to obtain lasing in smaller cavities with effective radiation outcoupling useful for optical interconnects.⁴⁹

In summary, continuous-wave electrically pumped lasers operating at room temperatures and elevated temperatures can be used for practical applications in the future photonic integrated circuits. The above-discussed device platforms and concepts can be implemented to get such laser operation. Semiconductor laser structure is based on $p-i-n$ junction. Contacts p - and n - of the junction need to be made of CMOS compatible metal. For obtaining small gain volumes and good heat dissipation, a CMOS-compatible metal shell can be implemented. The metal deposition must be optimized for having near single-crystal quality to reduce Joule losses. Introducing a dielectric resist spacer like silicon nitride (SiN) of a proper thickness is

essential for lowering Joule losses and having good heat dissipation. The thickness of the p -type layer can be reduced using the tunnel junction concept, allowing to increase thermal conductance and decrease electrical resistance with associated Joule losses. Fabrication needs to be optimized at all steps for obtaining minimal surface roughness to have a sufficient Q factor for gain-loss compensation. Passivation of the device must be carried out to improve gain properties that suffer at high temperatures. High gain materials like InGaAs can be implemented for CW electrically pumped RT laser operation. Approaches such as nanophotonic cloaking should be used for reducing signal crosstalk between adjacent photonic links to increase the integration density.

1.2 Bound States in the Continuum (BICs)

Conventional lasers trap the light in the cavity by the use of mirrors, total internal reflection, or Bragg scattering in periodically extended structures, such as photonic crystals, as discussed above. In these setups, the quality factor is mainly determined by the efficiency of the reflective parts of the laser cavity; this is, by how well radiation (and dissipative) losses can be avoided. At the same time, the Q factor of a conventional mode gradually increases as so does the cavity size.²⁶ In recent years, a different way of trapping light in a system without relying on reflective surfaces has been actively used. It refers to using a special class of modes supported by certain material/geometrical configurations, the so-called bound states in the continuum (BICs).

Bound states in the continuum are a general wave phenomenon. As the name suggests, they are non-radiative, confined states above the continuum threshold, *i.e.*, they coexist with the continuum of radiative states of the system (see Figure 1.5).^{25, 89} This somehow surprising property can be understood in terms of full scattering cancellation between different radiative channels, as well as in terms of the symmetries of the system.²⁵

Figure 1.5. BIC schematics. Figure is reproduced with permission from ref [25](#), © 2016 Springer Nature.

Strictly speaking, true BIC can only exist in infinitely extended systems. In fact, it was theoretically shown that BICs do not exist in compact structures (unless exotic materials such as epsilon-zero or mu-zero ones are considered).²⁵ For those infinite systems, when the BIC condition is met, the Q factor of the mode diverges to infinity, and that mode cannot be excited by external sources. It occurs due to the fact that the BIC is decoupled from the continuum of radiative modes. Light coupled to the BIC mode would be trapped forever. Of course, in reality, such infinite systems do not exist. However, the parameters of a finite system can be optimized in a way that it will be very close to the condition of BIC of the corresponding infinite system. This kind of quasi-BIC is often referred to in the literature as a supercavity mode.²⁶ Supercavity modes have finite but extremely high Q factors compared to regular resonant modes (see Figure 1.6). The Q factor of these quasi-BICs is ultimately determined by the size of the fabricated sample, by fabrication imperfections (*i.e.*, losses from scattering), and by absorption, if any. However, depending on the particular supercavity mode in consideration, extremely large Q-factors can be obtained even for a small number of particles.⁹⁰

Figure 1.6. Q factor comparison between a supercavity mode and a regular resonant cavity mode. Figure is adapted with permission from ref [26](#), © 2017 Springer Nature.

The work presented in this thesis is about obtaining single-particle dielectric

nanolasers based on the quasi-BIC that originates from full scattering cancellation of 2 radiative channels, specifically, a Friedrich–Wintgen BIC.²⁵ It should be noted that this is only one out of several existing types of BICs. Since the concept of BICs is central in the realization of a significant amount of work performed for this thesis, next, I present an overview of known BIC types and their applications.

Historically, BICs were introduced, for the first time, as a mere theoretical prediction in an artificial electronic system in 1929.²⁵ Years later, they were extended to photonic systems, where they have found the vast majority of applications. Generally, BICs can be classified into three types:²⁵ symmetry-protected BICs and separability-protected BICs; parametric BICs based on coupled resonances and a single resonance; and inverse construction-based BICs. In my review, the focus is on BICs obtained in optics only.

The physics of symmetry-protected BICs consists in decoupling between a leaky mode (see [Figure 1.5](#)) and a radiative wave due to their symmetry mismatch when the system has a particular symmetry with respect to rotation or reflection that the radiation channel does not have.^{25, 91} It was theoretically shown that, in such a situation, the coupling amplitude between those modes equals zero, ensuring that the state is non-radiative, despite being embedded in the continuum of radiative states (*i.e.*, having a dispersion relation that does not fulfill the waveguiding condition).⁹² Arguably, one of the first experimental demonstrations of a symmetry-protected BIC was performed using periodic metal mesh in [ref 93](#). Very recently, such BIC has been used in a vortex microlaser.⁹⁴ Symmetry-protected BICs were demonstrated experimentally in many systems with 2D and 1D periodicity^{25, 27} such as photonic crystals, gratings, arrays of dielectric particles, metasurfaces...

I will discuss the symmetry-protected BIC formation mechanism using one particular example from literature, which focuses on a one-dimensional photonic crystal (PhC) slab.⁹⁵ In that theoretical work, a PhC slab embedded in a periodic dielectric background was studied, as shown in [Figure 1.7B](#). The band structure of the photonic crystal is depicted in [Figure 1.7A](#). Radiative

modes with one leaky channel were considered in that work (existing in the yellow shaded area in [Figure 1.7A](#)). Let us consider a point 1 in [Figure 1.7A](#), corresponding to the Γ point of the reciprocal lattice, $k_y = 0$. At that point, the system has 180° rotational symmetry with respect to the x-axis; thus, odd and even modes with respect to this symmetry are decoupled.²⁵ In particular, E_z is an odd function in the y-coordinate for the slab mode while E_z is even in y for the radiative wave (solution of Maxwell's equations outside of the slab).⁹⁵ Hence, these two modes decouple due to the symmetry mismatch,⁹² resulting in the vanishing of the amplitude of the radiative wave and the (otherwise radiative) mode of the slab is being bound to the surface. Thus, the symmetry-protected BIC is formed.

Here, BICs are supported not only in the gamma point of the reciprocal lattice ($k_y = 0$) but at the edge of the first Brillouin zone as well ($k_y = \pi/a$), where a is the distance between the layers of the photonic crystal (see [Figure 1.7B](#)). The Q factors of the modes are shown in [Figure 1.7C](#). As can be seen, for the wavevectors corresponding to BICs, the Q factor diverges to infinity (see, for example, the modes 1 and 4 at $k_y = 0$). When the wavevector k_y is gradually increased, high but finite Q factors are observed, a situation in which the mode is usually referred to as a quasi-BIC. The near field patterns corresponding to symmetry-protected and topologically-protected BICs are depicted in [Figure 1.7D](#), demonstrating how the modes are trapped in the photonic crystal slab.

Figure 1.7. Symmetry-protected BIC. (A, C, D) Simulated characteristics of one-dimensional zinc oxide photonic crystal slab embedded in a periodic dielectric background shown in panel (B). (A) Band structure. The area marked by yellow is where only one leaky channel in the surrounding medium is present. The green and blue lines show the dispersion of the leaky modes of the PhC slab that are odd or even in x , respectively. The grey line depicts the only true guided mode supported by the structure (located below the light line). (C) Q factors of the guided modes and bound states in the continuum. States denoted by numbers 1, 4, 3, 6 are symmetry-protected BICs. States 2 and 5 are topologically-protected BICs. (D) Near field patterns. Dotted lines show the boundaries of the simulated nanostructure. Figure is adapted with permission from ref 95.

A quasi-BIC allows some degree of radiation to escape the system. Thus, by reciprocity, it is also possible to excite the quasi-BIC by external sources. In

the example above, the radiative channel was open by breaking the symmetry in the reciprocal lattice. Logically, breaking a system's structural symmetry is another way to open a radiative channel for obtaining a quasi-BIC.⁹⁶ An illustrative example of this approach is shown in a study about asymmetrical metasurfaces.⁹²

Another way of BIC formation is based on separability, which can be explained by an example of a two-dimensional Hamiltonian for a quantum system.^{25, 97} If the Hamiltonian can be expressed as a superposition of two summands as $H = H_1(x_1) + H_2(x_2)$, each component of this Hamiltonian H_i refers to an eigenfunction $\Psi_i(x_i)$ and eigenenergy E_i . Due to the separability of the coordinates in the Hamiltonian, the product $\Psi_{12} = \Psi_1 \cdot \Psi_2$ is an eigenfunction with energy $E_{12} = E_1 + E_2$. If E_1 and E_2 lay below the continuum edge ($\Psi_{1,2}$ are bound states), Ψ_{12} is also a bound state ($|\Psi_{12}| \rightarrow 0$ when $|x_i| \rightarrow +\infty$ for $i = 1, 2$). However, E_{12} can be greater than the continuum edge, meaning that for this case, Ψ_{12} is a bound state in the continuum. By now, separable BICs were demonstrated theoretically for several optical systems.²⁵

The second major BIC group is called BICs from parameter tuning or accidental BICs.^{25, 27} The first BIC of this type is the Fabry–Perot BIC. In photonics, these BICs can be described using an example of photonic crystals. Let us consider a single 1D photonic crystal slab made of a square array of silicon rods (see the inset in Figure 1.8A). Let us assume that there is a guided-mode resonance present in the slab. The guided resonance (or the leaky resonance) is strongly confined by the slab and placed above the light line, allowing the radiation outcoupling (see Figure 1.5). It is well-known that the presence of a leaky resonance in the slab results in a Fano line-shape observed in the transmission spectrum.⁹⁸ If the slab is irradiated by a plane wave, light can partially reflect from the slab surface and partially propagate through the slab, creating a so-called direct pathway.⁹⁹ At the same time, the guided resonance will be excited by the incident radiation. This resonance will then couple to radiative waves, which will propagate on both sides from the photonic crystal, resulting in a so-called resonant pathway.⁹⁹ As a result, the interference effect between these two channels may happen. In particular, in

the vicinity of the guided resonance, the transmission spectrum reaches zero and unity (see [Figure 1.8A](#)). Unity transmission corresponds to the case of constructive interference of the direct and resonant pathways in transmission and destructive interference in reflection. The opposite situation occurs for zero transmission. In the latter case, when there is unity reflection, the PhC slab acts as a perfect mirror. Thus, by the use of two identical PhC slabs, it is possible to achieve a dark state (a BIC) when the radiation cannot escape the system. The slabs must be displaced by an appropriate distance at which the Fabry–Perot resonance, formed between the slabs, occurs exactly at the frequency ω_0 where a zero reflectivity is achieved for each slab separately (see [Figure 1.8A](#)).⁹⁸ That is how the Fabry–Perot BIC is formed in the system of double PhC slab.

Let us consider a double 1D photonic crystal slab shown in the inset of [Figure 1.8B](#). If the double PhC slab is irradiated by a plane wave at normal incidence, there is a unity transmission peak with diverging Q factor for optimal distance d_0 between the slabs corresponding to the Fabry–Perot BIC (see [Figure 1.8B](#)). The transmission reaches unity because this FP BIC originates from a guided resonance (see ref [98](#) for more details). For the Fabry–Perot BIC, the Q factor is infinite, and the mode is perfectly spatially confined (see the field profile in [Figure 1.8C](#)). The Q factor of the system can be tuned by altering the distance between the slabs.⁹⁸ When the distance is slightly off the optimum d_0 , the mode has a finite Q factor corresponding to a quasi-BIC (see the field profile in [Figure 1.8D](#)). The quasi-BIC is a leaky resonance (see [Figure 1.5](#)). In that case, the radiation can escape the photonic crystal slab (unlike in the BIC case).

In optics, Fabry–Perot BICs were demonstrated for several systems, such as double PhC slabs,^{98, 100} stacked gratings, waveguide arrays.²⁵

Figure 1.8. Fabry–Perot BIC. Simulated transmission through the one-dimensional single (A) and double (B) silicon photonic crystal slab shown in the inset. The red arrows highlight the direction of irradiation. The black arrow indicates the Fano resonance (A) and a sharp resonance in the transmission corresponding to the Fabry–Perot BIC (B), whose field profile is shown in the panel (C). (D) Field profile corresponding to a quasi-BIC. Figure is adapted with permission from ref 98, © 2009 Optical Society of America.

Another interesting accidental BIC is based on a single resonance, the so-called single-resonance parametric BIC or topologically-protected BIC.²⁵ Such BIC is realizable if a sufficient number of parameters of a system is tuned. In optics, BICs of that type were investigated in various systems,²⁵ such

as periodic arrays of rods,¹⁰¹ arrays of spheres,¹⁰² single dielectric ridges,¹⁰³ and photonic crystals.¹⁰⁴⁻¹⁰⁹

In one of the pioneering studies that experimentally demonstrated such BIC,¹⁰⁴ the photonic crystal consisted of a square array of circular holes etched in the dielectric slab (see Figure 1.9B). The band structure of the slab is displayed in Figure 1.9A. The mode TM_1 undergoes the condition of BIC for several points in the reciprocal lattice. In this case, two BICs exist in the slab – a symmetry-protected one (at the Γ point) and a topologically-protected BIC (at a regular k point, between Γ and X points, see Figure 1.9C showing the Q factors of the guided modes and BICs). The field profile of the topologically-protected BIC is displayed in Figure 1.9D.

This BIC origin can be explained by the simultaneous accidental vanishing of radiative wave amplitudes for both s and p polarizations (namely c_s and c_p , respectively) at optimal parameters of the system.¹⁰⁴ This allows a single resonance to evolve into a BIC. In general, the radiative amplitude for each polarization is a complex number (it has two degrees of freedom, real and complex components). Thus, in total, we have four degrees of freedom for two polarizations while two variables are tuned – components of in-plane wavevector k_x and k_y . Thus, the simultaneous vanishing of the complex amplitudes can occur at a very low probability.¹⁰⁴ There are ways to improve the chances of such accidental BIC to emerge. The logical way consists of decreasing the degrees of freedom for both radiative amplitudes, which can be made by implying special symmetry conditions to the system of interest.¹⁰⁴

In ref 104, the total number of degrees of freedom is two, which allows matching the BIC condition by altering k_x and k_y . Specifically, there are only four points in the reciprocal lattice where the radiative wave amplitudes are equal to zero simultaneously (these points are highlighted by black circles in Figure 1.9E). Thus, there is only one such point in the Γ –X direction (depicted by a green line in the right panel of Figure 1.9E), which corresponds to the topologically-protected BIC at the k point highlighted in Figure 1.9C. Accidental vanishing of the radiative amplitude c_p is illustrated in Figure 1.9F.

An interesting feature of topologically-protected BICs is that they are robust against small tuning of geometrical parameters, which will tune them in k -space. For instance, such evolution of these BICs was demonstrated in theory in ref 95.

Figure 1.9. Topologically-protected BIC. (A) Computed band structure of silicon nitride photonic crystal slab displayed in (B). (C) Q factor of the guided modes and bound states in the continuum for the TM_1 band, shown along the Γ – X direction of the reciprocal lattice. The infinite Q factor at the Γ point refers to the symmetry-protected BIC. The infinite Q factor at the k point (between the Γ and X points) corresponds to the topologically-protected BIC. (D) Field profile of the topologically-protected BIC. (E) Colormaps of radiative wave amplitudes c_s and c_p (which refer to s - and p - polarizations, respectively) for the band TM_1 . Black circles indicate the parameters of the system at which simultaneous vanishing of both radiative wave amplitudes is achieved, which results in the topologically-protected BIC. The green line segment highlights the Γ – X direction. (F) c_p displayed along the Γ – X direction. Figure is adapted with permission from ref 104, © 2013 Springer Nature.

The next type of accidental BICs is the so-called Friedrich–Wintgen BICs.²⁵ Such BIC is achieved by destructive interference of resonances in the far field. For example, in a system studied in ref 110 consisting of a metal grating coupled to a dielectric slab, the destructive interference between a photonic mode of the slab and a surface plasmon polariton of the grating resulted in a

formation of the quasi-BIC.

In optics, Friedrich–Wintgen BICs have been studied in 1D dielectric gratings,¹¹¹ metallic gratings,¹¹² photonic crystal slabs.¹¹³ In addition to that, BICs of this type were found in waveguides and lasers.²⁵ Friedrich–Wintgen BICs have also been studied theoretically in a single dielectric cylinder geometry, as introduced here earlier. Since the major contribution of my thesis is the demonstration of lasing in a small dielectric nanoparticle enabled precisely by Friedrich–Wintgen quasi-BICs for that system, I will discuss their formation in detail here.

Quasi BIC in a single dielectric cylinder originates from the strong interaction of two resonant modes: a Mie-like mode (which strongly depends on the diameter of the cylinder and is largely insensitive to its height) and a Fabry–Perot (FP)-like mode (which is formed by the multiple reflections of the guided mode supported by the cylinder at its top and bottom interfaces and therefore highly sensitive to the cylinder height).³⁸ The Mie-FP mode formulation is addressed in more detail in the [Methods](#). The strong interaction between these two resonant modes manifests itself as an avoided resonance crossing in the scattering spectrum of the cylinder (see [Figures 1.10A,B](#) and [Figure 1.11A](#)).^{38, 40} The near field patterns at points *A* and *F* of [Figure 1.10A](#) refer to a Mie-like mode. The near field patterns at points *C* and *D* of [Figure 1.10A](#) refer to a Fabry–Perot-like mode.

Figure 1.10. Supercavity mode formation in a dielectric cylinder of a high permittivity

$\epsilon = 80$. A distilled water at room temperature and microwave frequencies can have a dielectric permittivity $\epsilon \approx 80$. (A) Illustration of the frequencies of the coupled modes together with their near-field patterns shown in the insets. Curve thickness corresponds to linewidth in the scattering spectra. White lines in the near-field patterns correspond to the cylinder boundaries. The quasi-BIC originates from the strong interaction between two resonant modes in the nanocylinder. (B) Frequencies of the two modes TE_{012} and TE_{020} which experience avoided resonance crossing. (C) Q factor in dependence on the aspect ratio. The Q factor reaches a maximum at the supercavity mode point. Figure is adapted with permission from ref [38](#), © 2017 American Physical Society.

Along with the avoided crossing, one of the modes exhibits linewidth narrowing (increase in the Q factor) and a decrease in the scattering efficiency in the anticrossing region. Specifically, the quasi-BIC (the supercavity mode point)^{[38](#)} corresponds to geometrical parameters of the system where the maximum Q factor (minimum radiation losses) is reached (see [Figure 1.10C](#) and [Figure 1.11B](#)). The formation of the dark mode, which refers to the minimal scattering, occurs at a different point (the dark mode point shown in [Figure 1.11C](#)).^{[40](#)} These two points match exactly for the case of an ideal BIC.^{[40](#)} Thus, there are three main signatures of the emergence of a supercavity mode of this type: anticrossing between Mie-like and FP-like modes, increase in the Q factor for one of the modes, and decrease in its scattering efficiency in the anticrossing region. The use of a supercavity mode reduces the system's radiative losses (by increasing its Q factor several times with respect to a regular Mie mode of the same order),^{[38](#)} enabling gain/loss compensation in nanoparticles of a smaller size.

A more formal approach to the concept of supercavity modes in a single cylinder consists of using the so-called multipole expansion. In general, a far field can be expanded into a multipole series, and each multipole can be referred to as a radiation channel.^{[40](#)} Let us consider an aspect ratio corresponding to a point α in [Figure 1.11D](#). Here the dominant radiation channel is an electric dipole (ED, see [Figure 1.11E](#)). We can refer to it as the initially dominant radiation channel. The radiated power through other

channels is negligible. For example, the radiated power through the multipole of the next order of approximation, namely magnetic quadrupole (MQ), is suppressed (see Figure 1.11D). After we increase the aspect ratio from the point α to the point β in Figure 1.11D, the magnetic quadrupole becomes the dominant radiation channel. At the same time, radiation of the initially dominant channel is suppressed (see the point β in Figure 1.11D). This can be interpreted as a decoupling of a leaky mode from the radiation continuum, which creates a quasi-bound state in the continuum. The same thing occurs when decreasing the aspect ratio from the point γ to the point β . When the dominant radiation channel changes from the lower-order multipole to the higher-order multipole, the Q factor reaches a maximum (at the quasi-BIC point β). Due to that multipole conversion, the far-field radiation pattern drastically reshapes, as shown in Figure 1.11E.

Figure 1.11. Quasi-BIC and dark mode in a dielectric cylinder of a high permittivity $\epsilon = 80$. A distilled water at room temperature and microwave frequencies can have a dielectric permittivity $\epsilon \approx 80$. (A) Frequencies of the two modes TE_{110} and TM_{111} which experience avoided resonance crossing. The anticrossing branches can be referred to as higher and lower frequency modes. (B) Q factor of the higher frequency mode *versus* the aspect ratio. The Q factor reaches a maximum at the supercavity mode point. (C) Scattering peak amplitude of the higher frequency mode in dependence on the aspect ratio. It reaches a minimum having a negligible value at the dark mode

point. This value is non-zero, meaning that the mode excitation from the far field is highly suppressed but still possible (the mode is almost dark). (D) Radiated power of the components (electric dipole and magnetic quadrupole) of the multipole decomposition of the higher frequency mode. (E) 3D far-field radiation patterns of the higher frequency mode at the aspect ratios corresponding to the points α , β , and γ . Figure is adapted with permission from ref 40.

It is important to note that a dielectric cylinder of a high permittivity $\epsilon = 80$ was studied in refs 38, 40. That was carried out to demonstrate the concept of quasi-BICs. The higher the permittivity, the closer the system is to an ideal BIC, allowing to highlight the main signatures of the supercavity mode formation – avoided resonance crossing (Figure 1.11A), maximum Q factor at optimal parameters (the quasi-BIC or supercavity mode point, see Figure 1.11B), and minimal scattering near optimal parameters (the dark mode point, see Figure 1.11C). In ref 40, a big (centimeter-sized) plastic cylinder filled with distilled water was studied at microwave frequencies (in a range from 0.8 to 5 GHz) at room temperature. The dielectric material (*i.e.*, distilled water) under these conditions has a permittivity $\epsilon \approx 80$ as highlighted in refs 40, 114.

The third BIC major category comprises BICs from inverse construction.²⁵ In this approach, first, a specific BIC is constructed. Second, a system that can support that BIC is found. One example of such BICs is designing a potential in the Schrödinger equation that can support a BIC.¹¹⁵ The other examples of such BICs are discussed in ref 25 (together with optical experiments where those BICs were realized).

Along with discussed BICs of major categories, new types of BICs have been studied in recent years, such as anisotropy-induced BICs and BICs enabled by optical bistability.²⁷

In conclusion, high Q factors and significant near-field enhancement make BIC concepts applicable for many uses in optics such as lasers, sensors, filters, photonic crystal fibers, high-harmonic generation.^{25, 27} In recent years, several studies used BIC concepts for the experimental demonstration of lasing,^{19, 20, 94} sensing,^{28, 29, 116} filters,¹¹⁷ high-harmonic generation.^{33, 118, 119} In

this thesis, a quasi-BIC supported by a single particle will be used, for the first time, to achieve lasing action in subwavelength-sized resonators.

Chapter 2

2 Lasing Action in Single Subwavelength Semiconductor Cylinder Supporting Supercavity Modes

2.1 Concept

On-chip light sources are critical for the realization of fully integrated photonic circuitry. So far, semiconductor miniaturized lasers have been mainly limited to sizes on the order of a few microns. Further reduction of sizes is challenging fundamentally due to the associated radiative losses. While using plasmonic metals helps to reduce radiative losses and sizes, they also introduce Ohmic losses hindering real improvements. In this work, we show that by using quasi-bound states in the continuum, or supercavity modes, we circumvent these fundamental issues and realize one of the smallest purely semiconductor nanolasers thus far. Here, the nanolaser structure is based on a single semiconductor nanocylinder that intentionally takes advantage of the destructive interference between two supported optical modes, namely Fabry–Perot and Mie modes, to obtain a significant enhancement in the quality factor of the cavity. We experimentally demonstrate the concept and obtain optically pumped lasing action using GaAs at cryogenic temperatures. The optimal nanocylinder size is as small as 500 nm in diameter and only 330

nm in height with a lasing wavelength of around 825 nm, corresponding to a size-to-wavelength ratio as low as 0.6.

We designed our cylindrical nanoresonator to support a quasi-BIC that originates from the strong interaction of two resonant modes: a Mie-like mode (which strongly depends on the diameter of the cylinder and is largely insensitive to its height) and a Fabry–Perot (FP)-like mode (which is formed by the multiple reflections of the guided mode supported by the cylinder at its top and bottom interfaces and therefore highly sensitive to the cylinder height).³⁸ As discussed earlier in the text, there are three main signatures of the emergence of a supercavity mode of this type: anticrossing between Mie-like and FP-like modes, increase in the Q factor for one of the modes, and decrease in its scattering efficiency in the anticrossing region. The use of a supercavity mode reduces the radiative losses of the system (increasing its Q factor several times with respect to a regular Mie mode of the same order)³⁸ enabling gain/loss compensation in nanoparticles of a smaller size.

We focus our attention on obtaining lasing from a supercavity mode of azimuthal order $i = 3$. In particular, we study a single GaAs cylinder standing on a quartz substrate. Due to the relatively low gain of GaAs at room temperature, which is further reduced by the large surface recombination present in small particles, lasing experiments are performed at the liquid nitrogen temperature (*i.e.*, 77 K) to increase the quantum yield of GaAs (note that no surface passivation or any chemical treatment is applied to the nanostructures). Surface passivation or chemical treatment of the nanolasers would improve the emission quantum efficiency of GaAs. Meaning, it would increase the gain of the active medium, allowing the lower Q factor modes to be used in laser operation and, potentially, obtaining room-temperature lasing. We work with unpassivated material to demonstrate that supercavity modes can be used to achieve lasing in a single semiconductor nanoparticle of relatively low gain and small size. We do not use any passivation. This decision is not constrained by experimental conditions. Besides, we have decided to use unpassivated GaAs because it is simpler (it eliminates complex fabrication steps for passivation of the GaAs surface).

When tuning the temperature from 300 to 77 K, the emission peak of GaAs shifts from 870 to 830 nm.¹⁹ Thus, in my work, the geometrical parameters of the cylinder (diameter D and height H) are optimized to achieve high Q factors at $\lambda = 830$ nm.

2.2 Numerical Simulations

To illustrate the physical mechanism behind the supercavity mode formation in a simple way, let us first consider a cylinder with a constant relative permittivity $\varepsilon = 13$ (corresponding, approximately, to GaAs at 830 nm wavelength and 77 K) in vacuum. We perform two types of three-dimensional electromagnetic simulations – scattering simulations and Q factor calculations (illustrated in parts A and B, respectively, of [Figure 2.1](#)). We study the illumination of the cylinder by a transverse electric (TE) wave in the scattering simulations. The TE wave is defined as the one having an incident magnetic field polarized along the axis of the cylinder and the electric field and incident wavevector perpendicular to it, as shown in [Figure 2.1A](#). In the Q factor simulations, we excite TE modes by a pointlike dipole highlighted in [Figure 2.1B](#) (see the [Methods](#) for more details). We compute the scattering efficiency and the eigenmodes supported by this cylinder as a function of its geometrical parameters using full-wave simulations based on the finite-difference time-domain method (see the [Methods](#) for the detailed description). The results are shown in [Figure 2.1C](#). There, we plot the scattering efficiency as a function of the dimensionless parameters $x_H = \pi H/\lambda$ (*i.e.*, the size parameter, where the height of the cylinder is used as a characteristic dimension, λ is the wavelength in vacuum) and $\rho = D/H$ (the cylinder aspect ratio). Overlaid with the scattering map, we plot the result of the Q factor simulations: quasi-TE eigenmodes (quasi-normal modes) corresponding to the azimuthal order $i = 3$ are shown by a set of black circles; their area is directly proportional to the value of the Q factor. Here, we use a standard mode notation TE_{ijl} , where indices i , j , and l are the azimuthal, radial, and axial indices of the mode, respectively.³⁸ “TE” or “TM” denotes the dominant polarization of the mode.⁴⁰ As seen in the plot, two modes of the third azimuthal order coexist in the range of size parameters studied (denoted as TE_{320} and TE_{312}). TE_{320} is the Mie-like mode, and TE_{312} is the FP-like mode. More important is the clear indication of their strong coupling, evidenced through an avoided spectral crossing. In the anticrossing region, the modes are hybrid; thus, the use of the standard

notation might be misleading when addressing the modes in that region. To get around that, we use a simplified notation to denote the branches (modes) TE_3 (higher frequency branch) and $'TE_3$ (lower frequency branch) that originate from strong coupling of the modes TE_{320} and TE_{312} . Along with avoided spectral crossing, a dramatic increase in the Q factor, leading to a maximum (at the supercavity mode point denoted as β in [Figure 2.1C](#)) is observed for the mode TE_3 near the anticrossing region, indicating that a supercavity mode has been formed. The reduction of the scattering efficiency at slightly higher diameters, with respect to the supercavity mode point of the mode TE_3 , is less noticeable due to relatively low permittivity.³⁸ The optimum values of dimensionless parameters for this third azimuthal order mode are $x_H \sim 1.30$ and $\rho \sim 1.47$ (the supercavity mode point). For an operational wavelength $\lambda = 830$ nm, these correspond to a cylinder with height $H \sim 344$ nm and diameter $D \sim 504$ nm. The corresponding Q factor reaches a maximum value of $Q = 970$. It should be noted that it has been theoretically shown³⁸ that an infinite permittivity is formally equivalent to a spatially infinite system in which an ideal BIC may exist. Practically, this means that the higher the permittivity is, the closer to an ideal BIC the supercavity mode is. This explains why in the case of moderate permittivity of GaAs, the supercavity formation leads to a somehow moderate 5-fold increase in the Q factor. This, nevertheless, turns out to be crucial to reduce the lasing threshold for the third azimuthal order mode, as will be shown in the experiment below.

Figure 2.1. Simulated characteristics of a single GaAs nanocylinder ($\epsilon = 13$) in vacuum. (A, B) Illustration of the dielectric cylinder in vacuum during scattering simulations (A) and Q factor (near-field, far-field) simulations (B). (C) Scattering efficiency map Q_{sca} as a function of the dimensionless parameters $x_H = \pi H/\lambda$ (the size parameter) and $\rho = D/H$ (the cylinder aspect ratio). The alternative x- and y-axis correspond to the wavelength and diameter obtained when the height of the cylinder is fixed at $H = 345$ nm. The peak positions and the Q factors of the modes obtained from the Q factor simulations are shown by black circles; their area is directly proportional to the value of the Q factor. The results of the Q factor simulations are shown for the quasi-TE modes of azimuthal order $i = 3$ (labeled TE_3 and ${}^{\prime}\text{TE}_3$). Only these two modes are displayed in order to highlight the anticrossing of the modes TE_{320} and TE_{312} and Q factor evolution as a function of the diameter and wavelength (the Q factor reaches its maximum value for the optimal parameters indicated by a point β , which is the supercavity mode point). The insets show the electric near-field maps in the YZ-plane passing through the center of the cylinder for different regimes of the mode TE_3 . The white lines show the cylinder boundaries.

To complete the characterization of the supercavity mode, the near fields and the far-field radiation pattern in the supercavity regime are depicted in [Figure 2.2](#). From the near-field maps ([Figures 2.2B,C](#)), the TE_{312} character of the mode becomes apparent. On the other hand, the far-field pattern ([Figure 2.2D](#)) shows that, along with the regular in-plane lobes, common for Mie-like

modes, out-of-plane lobes are present as well. This can be explained by the fact that supercavity mode is a hybrid FP-Mie mode and, thus, it cannot be identified as an FP-like mode or a Mie-like mode (the Mie-FP mode formulation is addressed in more detail in the [Methods](#)). A detailed analysis of the evolution of the near-field distribution around the supercavity regime confirms that the mode TE_3 has three different regimes indeed, evolving from a Mie-like one ([Figure 2.1C](#) at point α) to the Supercavity one ([Figure 2.1C](#) at point β) and finally to a Fabry–Perot-like one ([Figure 2.1C](#) at point γ). Note that we collect the very edge of the emission side lobes of the supercavity mode in optical experiments, as expected from the simulated radiation pattern in [Figure 2.2E](#).

Figure 2.2. Simulated characteristics of the supercavity mode (or the supercavity mode point β). (A) Illustration of the dielectric cylinder ($\epsilon = 13$) in vacuum during the Q factor (near-field, far-field) simulations. (B, C) Simulated near fields in the XY (B) and YZ (C) planes of the cylinder, respectively. The white lines in (B, C) show the cylinder boundaries. (D) Simulated 3D far-field radiation pattern. (E) 2D projection of the simulated far-field radiation pattern. The white dashed circle shows the range of wavevectors collected by a 0.55 NA objective (similar to the one used to collect the emission in the lasing experiment).

2.3 Nanofabrication and Characterization

To experimentally demonstrate lasing for the analyzed third-order supercavity mode, GaAs nanocylinders standing on a quartz substrate are fabricated using the method reported in ref 19. First, a GaAs film grown on top of a commercially available GaAs wafer with a sacrificial layer of AIAs (see Figure 2.3A) is transferred to a quartz substrate using the technique known as epitaxial lift-off,¹²⁰ followed by a direct bonding process developed in-house using a wafer bonding system. The transferred film is then etched to the desired thickness using the inductively coupled plasma (ICP) method. The nanocylinder pattern is then defined using a standard e-beam lithography with hydrogen silsesquioxane (HSQ) as a negative resist. The final structure is formed by an additional dry-etching step (see the Methods for more detailed information). The SEM image of one of the fabricated nanolasers is shown in Figure 2.3B.

Figure 2.3. (A) Nanofabrication process flow consists of first, epitaxial lift-off of the GaAs film, its bonding to the quartz substrate, and etching to the required thickness, second, electron-beam lithography, third, etching. (B) Exemplary SEM image of the fabricated nanostructure.

Note that, despite their low index, the presence of the quartz substrate and

the remaining capping HSQ layer affects the spectral position and the Q factor of the supercavity mode. Thus, we reoptimize the parameters of the cylinder to account for them, obtaining a maximum Q factor of 280 for optimum values of the dimensionless parameters $x_H \sim 1.29$ and $\rho \sim 1.47$, corresponding to the cylinder height $H \sim 340$ nm and diameter $D \sim 500$ nm at $\lambda = 830$ nm (the complete analysis of the modes of the reoptimized geometry will be presented in detail later in the text). The height of the GaAs cylinders obtained in the fabricated samples is close to the targeted one, around 330 nm (measured using reflectometer and tilted SEM images). The sample is then cooled down to 77 K using liquid nitrogen for further optical characterization (see the [Methods](#) for more details).

Figure 2.4. Measured absorption of the GaAs film of 350 nm in thickness at 77 K.

Before characterizing the fabricated nanostructures, we first investigate the absorption and PL spectra of GaAs films with a thickness of 350 nm. The absorption spectrum, recorded at 77 K, is presented in [Figure 2.4](#). The spectrum clearly shows the absorption edge at ~ 827 nm, which is in good correspondence with values reported in the literature for GaAs at this temperature.¹⁹ The narrow absorption peak at ~ 830 nm can be attributed to the excitonic resonance of the material.

For the spontaneous emission spectrum, large square areas (of $50 \mu\text{m} \times 50 \mu\text{m}$) patterned in the film using EBL are analyzed. We do so to characterize, together, the emission of GaAs and that of HSQ resist, used to pattern the

nanostructures in our experiments and known to have non-negligible fluorescence when treated with e-beam and high temperatures.^{121, 122} Indeed, for GaAs film with HSQ resist on top, the measured photoluminescence (Figure 2.5A) has a broad emission spectrum from 700 to 850 nm (a long pass filter is used to cut the pump at 700 nm), beyond that for GaAs alone which has a relatively narrow PL spectrum from 800 to 850 nm. The above-mentioned broad spectrum is attributed to the photoluminescence of GaAs combined with that of silicon nanocrystals formed in the silica matrix of cured HSQ (whose PL can range from 650 to 800 nm),¹²² which, in our case, arises due to the high-energy electron beam exposure, in a chemical reaction analogous to that happening during thermal annealing.¹²³ Thus, in our experiments, the photoluminescence of both Si nanocrystals in cured HSQ¹²⁴ and GaAs itself can provide gain for our nanolasers with different resonance wavelengths.

In general, after the etching process, HSQ can be removed using hydrofluoric acid (HF). However, for our system, this approach is not applicable. That is because HF will etch not only HSQ but also the quartz substrate. This will result in the detachment of the cylinders from the substrate. Other ways of HSQ removal, such as using chrome sacrificial layer or resist sacrificial layer, can be utilized.¹²⁵ However, these approaches demand additional fabrication steps that require optimization. For our nanostructures, HSQ was not removed.

Figure 2.5. (A) Normalized photoluminescence spectrum measured from GaAs film

without HSQ (red) and with 150 nm layer of HSQ on top (black). (B) Normalized experimental dark-field scattering spectrum of a single GaAs nanocylinder with parameters $D = 500$ nm and $H = 330$ nm. Inset: exemplary SEM image of the fabricated nanostructure.

With this information in mind, we now move to the optical characterization of the nanocylinders at cryogenic temperatures, first studying their scattering spectra to identify the formation of the supercavity mode and then their emission under optical pumping. Figure 2.5B shows the characteristic dark-field scattering spectrum of a single GaAs nanocylinder with $D = 500$ nm and $H = 330$ nm (as measured using SEM). From our theoretical analysis, we expect the cylinder to support the supercavity mode at around 830 nm for these dimensions. Indeed, one can observe the emergence of a very narrow peak in the experimental spectrum at ~ 828 nm. This is in very good agreement with what is expected from the simulations, thus confirming the presence of the quasi-BIC mode.

To demonstrate lasing action, we optically pump the nanocylinders using a femtosecond laser (with a wavelength of ~ 530 nm, ~ 200 fs pulse duration, and 20 kHz repetition rate) and collect the PL using a microspectrometer setup (see Figure 2.6 and detailed information in the Methods).

Figure 2.6. Optical setup used in the lasing experiment. The green arrows indicate the pumping laser beam. The red arrows highlight the emission from the sample. The

laser beam passes through the beam splitter (BS), the lens system, and is focused on the sample. The sample is located on top of a cold finger of a cryostat placed upside down. The emission from the sample is collected by the same objective lens and imaged onto the spectrometer system. The latter consists of a slit, grating, and electron-multiplying charge-coupled device. The longpass filter (LP filter) is used to cut the pump at 700 nm.

Nanocylinders have been fabricated with a 20 μm spacing, and the pumping laser spot size is $\sim 5 \mu\text{m}$ in diameter, ensuring that each cylinder is individually irradiated during optical measurements. Figure 2.7A shows the PL spectrum evolution of the GaAs nanocylinder with diameter $D = 500 \text{ nm}$ and height $H = 330 \text{ nm}$, supporting the supercavity mode when pumped under different laser fluences. It can be seen that when the pumping fluence is larger than $260 \mu\text{J}/\text{cm}^2$, a pronounced, narrow peak appears at $\sim 825 \text{ nm}$. By plotting the integrated emission intensity *versus* the pumping fluence in log-log scale (Figure 2.7B), one can clearly see the “S” shape that indicates the transition from spontaneous emission (SE) to amplified spontaneous emission (ASE) and finally to lasing, with a lasing threshold of $\sim 300 \mu\text{J}/\text{cm}^2$.

The PL peak position and full-width-at-half-maximum (fwhm) as a function of the pumping fluence, extracted from the spectra, are shown in Figure 2.7C. Based on the fwhm, we observe a maximum Q factor of 235 in the lasing regime (in good agreement with the theoretically calculated value of 280, as discussed above). From the analysis of the lasing peak position, a clear blue-shift is observed when the pumping fluence is increased, which can be attributed to the Burstein–Moss effect associated with the band filling.^{126, 127} The lasing peak broadening that can also be observed when increasing the fluence can be attributed to two effects: band filling and thermal broadening. In our case, we observe the blue-shift of the lasing peak when increasing the pumping fluence, which is strong evidence of the band-filling effect. Given the high pumping fluence (few hundreds $\mu\text{J}/\text{cm}^2$) and the high repetition rate (20 kHz) of the femtosecond laser, in addition to the relatively bad thermal-dissipation setup (single GaAs nanopillar on quartz) used in our lasing experiment, we believe that thermal broadening effect also contributes to the

broadening of the lasing peak. This explains the drop in the Q factor from 500 to 235, as shown in Figure 2.7C. Such linewidth behavior has been observed in small lasers before.¹²⁸

Figure 2.7. Lasing action in a GaAs nanocylinder with diameter $D = 500$ and height $H = 330$ nm. (A) Evolution of the emission spectrum at different pumping fluences. (B) Log-log plot of output emission intensity *versus* pumping laser fluence showing the transitions from spontaneous emission (SE) to amplified spontaneous emission (ASE) and finally to lasing. (C) Peak position and linewidth extracted from the emission spectra.

To further confirm the coherent nature of the lasing emission, autocorrelation measurements using a Hanbury–Brown–Twiss setup^{19, 129} are performed. Figure 2.8 shows coincidence counts at different time delays for three regimes: SE (Figure 2.8A), ASE (Figure 2.8B), and lasing (Figure 2.8C). The normalized second-order correlation function at zero time delay $g_2(0)$ is defined as the ratio between the coincidence count number of the pulse at zero time delay, and an averaged count number over 22 other neighboring pulses. It is noted that the autocorrelation measurement for SE (Figure 2.8A) is performed on relatively large patterned GaAs squares (of $50 \mu\text{m} \times 50 \mu\text{m}$). This is to ensure that an adequate amount of signal can be collected and analyzed. It, however, does not change the nature of the SE from the

materials composing the nanolaser. Due to their larger size, the photoluminescence intensity of these squares is several times greater than that of the single GaAs nanocylinder used to measure $g_2(0)$ in the ASE and lasing regimes, which explains why the coincidence count number in [Figure 2.8A](#) is substantially higher than those of [Figures 2.8B,C](#). The $g_2(0)$ for the SE case is calculated to be 1.002, which is significantly smaller than the theoretical value for an ideal thermal light source (*i.e.*, $g_2(0) = 2$). This is because the coherence time of the SE is much lower than the detection limit of our experimental setup (~ 81 ps); it is in accordance with similar observations reported in earlier studies.^{19, 130} Nevertheless, we are able to measure a clear decrease in $g_2(0)$ upon transitioning between the ASE and lasing regimes, as shown in [Figures 2.8B,C](#), with the $g_2(0)$ reducing from 1.16 (for ASE regime, $F = 382 \mu\text{J}/\text{cm}^2$) to 1.008 (for the lasing regime, $F = 433 \mu\text{J}/\text{cm}^2$) and, thus, clearly indicating the coherent behavior of the nanolaser.

Figure 2.8. (A) Intensity autocorrelation histogram for $50 \mu\text{m} \times 50 \mu\text{m}$ GaAs square at the pumping laser fluence $F = 764 \mu\text{J}/\text{cm}^2$ and for the single GaAs nanocylinder of diameter 500 nm and height 330 nm for the pumping fluences (B) $F = 382 \mu\text{J}/\text{cm}^2$ and (C) $F = 433 \mu\text{J}/\text{cm}^2$.

We now prove that lasing is obtained for the third azimuthal order supercavity mode by studying how a slight detuning of the geometrical parameters from the optimal ones affects the system's performance. For that, lasing and dark-field scattering measurements are carried out for cylinders of the same height, namely $H = 330$ nm, but with diameters above and below the optimum one. In particular, [Figure 2.9A](#) shows the dark-field scattering spectra of GaAs cylinders with a diameter ranging from 420 to 653 nm with an increment of ~ 5

nm. The mode highlighted by the green dashed line in Figure 2.9A corresponds to the third azimuthal order mode TE_3 from the simulation. Higher and lower order modes can also be seen from the scattering data in Figure 2.9A. For better visualization of the supercavity mode formation, the scattering spectra for several selected radii around the optimum one are depicted in Figure 2.9B. It should be noted that due to the presence of high absorption at wavelengths below the excitonic resonance (see Figure 2.4), it is not possible to observe the full evolution of the scattering peak associated with the formation of the supercavity mode (from broad to narrow and back to broad) but only the narrowing of the resonance. Thus, we explore an alternative experimental method, relying on the analysis of lasing threshold, to prove that the third azimuthal order TE mode used for lasing indeed undergoes the formation of the supercavity regime, as explained below.

Figure 2.9. (A) Normalized experimental dark-field scattering spectra of GaAs nanocylinders with fixed height $H = 330$ nm and varying diameter $D = 420$ – 653 nm with an increment of ~ 5 nm. The green dashed line is a guide to eyes indicating the mode TE_3 . (B) The same normalized experimental dark-field scattering spectra plotted in 1D. The Dashed line indicates the mode TE_3 , which undergoes the supercavity regime in the vicinity of parameters shown by the red dashed ellipse.

Figure 2.10A shows the theoretically obtained (using Q factor simulations) peak positions of the quasi-normal eigenmodes in the span of parameters λ and D of our interest. In the case of the mode TE_3 , the corresponding Q factors are also plotted by red circles, so that the supercavity regime can be readily identified. The green triangles depict the experimental lasing peak positions obtained for various disk diameters. As can be seen, there is a good agreement between the simulated peak positions and the lasing peaks observed in the experiment. The discrepancy between theory and experiment is minimal and lies within the error of diameters determined by SEM and the possible slight deviations in the material parameters. Importantly, we can achieve lasing for the third azimuthal order mode TE_3 for a relatively wide range of cylinder diameters due to the low divergence of the quality factor near the optimal value. This gives us the flexibility to tune the emission wavelength of the nanolaser within the gain spectrum of the active materials involved (note that, in our case, these refer not only to GaAs but also to the Si nanocrystals obtained inside HSQ resist after the e-beam exposure). Besides the mode TE_3 , lasing is also obtained for bigger nanocylinders supporting higher azimuthal order modes (up to fifth-order TM_{510} mode, as shown in Figure 2.10A).

It is important to note that, for nanocylinders with diameters close to the optimum ($D \sim 500$ nm), the third-order mode of interest (TE_3) and the fourth-order Mie mode (TM_{410}) are spectrally close (~ 20 nm away from each other, according to the Q factor simulations, as can be seen in Figure 2.10A). Thus, it is crucial to observe both the third- and fourth-order modes in the lasing experiments to prove that the mode TE_3 is identified correctly. The detailed view of the emission spectra contributed by both these modes simultaneously is shown in Figure 2.10B. Moreover, as can be seen in Figure 2.10A, the lasing peak positions for all modes (third, fourth, and higher azimuthal orders) match well with the theory, further supporting the correct identification of the mode TE_3 .

Figure 2.10. (A) Theoretically obtained peak positions of the modes supported by a single dielectric nanocylinder $\epsilon = 13$ on a substrate, retrieved from the Q factor simulations, shown in the region of dimensionless parameters x_H , ρ of interest. The alternative x- and y-axis correspond to the wavelength and diameter obtained when the height of the cylinder is fixed at $H = 332$ nm. The modes denoted as TE_{320} and TE_{312} are strongly coupled, which is manifested by the anticrossing. At optimal parameters, these modes destructively interfere in the far field, which leads to the formation of the supercavity regime for the mode TE_3 , for which the corresponding Q factors are indicated by red circles (their area is directly proportional to the value of the Q factor). Strong coupling of the modes TE_{420} and TE_{412} manifests itself as an avoided resonance crossing. Modes TM_{410} and TM_{510} are regular Mie modes. The green triangles show the experimentally obtained lasing peak positions for cylinders with fixed height $H = 330$ nm and different diameters. (B) Evolution of the lasing spectrum for GaAs nanocylinders with fixed height $H = 330$ nm and different diameters. For each diameter in the plot, the pumping was set at a different fluence but always above the lasing threshold. Arrows highlight the modes TE_3 and TM_{410} identified by numerical simulations.

Figure 2.11 shows additional information on the modes TE_3 and TM_{410} obtained from the simulations. The overlap of the near-field maxima (Figures 2.11B–E) with the sidewalls of the cylinder is more pronounced for the mode

TM₄₁₀ compared to that of the mode TE₃. Thus, resonances of higher azimuthal orders are more sensitive to the sidewall roughness created during the dry-etch process, which tends to lower the Q factor in experiments. Consequently, compared to the mode TE₄₁₀, the mode TE₃ near the supercavity regime seems to be more robust against fabrication imperfections and, therefore, more suitable to obtain lasing in practical devices.

The higher the Q factor is, the lower the lasing threshold is. However, the cavity's quality factor is not the only thing that the lasing threshold depends on. Thus, the Q factor is not the only parameter that can be accounted for during clarification of which mode is preferred for lasing (see the [Methods](#) for more details).

Figure 2.11. Simulated characteristics of the single dielectric nanocylinder ($\epsilon = 13$) on a substrate ($n_{\text{substrate}} = 1.46$) with HSQ cap on top ($H_{\text{HSQ}} = 150$ nm, $n_{\text{HSQ}} = 1.41$). (A) Theoretically obtained peak positions of the modes supported by a single dielectric nanocylinder $\epsilon = 13$ on a substrate, retrieved from the Q factor simulations, shown in the region of dimensionless parameters x_H , ρ of interest. The alternative x- and y-axis correspond to the wavelength and diameter obtained when the height of the cylinder is fixed at $H = 332$ nm. Q factors of the modes are shown by red circles, where the area of the circles is directly proportional to the value of the Q factor. Modes TE₃ and 'TE₃ experience avoided crossing (mode TE₃ undergoes the supercavity regime at optimal geometrical parameters). Mode TE₄ anti-crosses with the mode 'TE₄. Modes TM₄₁₀ and TM₅₁₀ are regular Mie modes. The green triangles

show lasing peak positions obtained from experiments for the cylinders with fixed height $H = 330$ nm and varying diameters. $Q = 270$ is the Q factor of the mode TE_3 at the point β_3 ($D = 465$ nm, $\lambda = 783.5$ nm). $Q = 600$ is the Q factor of the mode TM_{410} at the point ζ_4 ($D = 465$ nm, $\lambda = 798.3$ nm). In spite of the fact that the Q factor of the mode TM_{410} is nearly two times higher than that of the mode TE_3 at $D = 465$ nm, when the diameter exceeds 460 nm, lasing is favorable to happen in the mode TE_3 instead of 4th order Mie mode even though both of them are laying within the gain spectrum. This stems from the fact that resonances of higher azimuthal orders are more sensitive to the sidewall roughness created during the dry-etch process, which tends to lower the Q factor in experiments. In this regard, the overlap of the near-field maxima with the sidewalls of the cylinder is more pronounced for the mode TM_{410} compared to that of the mode TE_3 . (B, C) Simulated near fields of the mode TE_3 in the vicinity of the supercavity regime for the parameters corresponding to the point β_3 in XY (B) and YZ (C) planes of the cylinder. (D, E) Simulated near fields of the Mie mode TM_{410} for the parameters corresponding to the point ζ_4 in XY (D) and YZ (E) planes of the cylinder. White lines in (B–E) show cylinder boundaries. Consequently, despite having a lower theoretical Q factor, the mode TE_3 seems to be more robust against fabrication imperfections and, therefore, more suitable to obtain lasing in practical devices.

As mentioned above, in order to demonstrate the formation of the supercavity regime of the mode, the lasing threshold is measured and analyzed for several nanocylinders with different diameters around the optimum, as shown in [Figure 2.12](#). For each diameter, two to four similar nanocylinders are measured to increase the statistic accuracy. As can be seen from the plot in [Figure 2.12](#), the lasing threshold has a minimum in the vicinity of $D = 500$ nm, indicating an increase in the Q factor of the mode for this diameter, which is in good agreement with the theoretically obtained behavior for the supercavity condition.

Figure 2.12. Measured lasing threshold values (depicted as red stars) shown as a function of the diameter of the nanocylinder for the mode TE_3 for GaAs cylinders with fixed height $H = 330$ nm. The red shadowed area is a guide to the eyes. The vicinity of the optimal diameter value is highlighted by the blue dashed ellipse (where the mode TE_3 undergoes the supercavity regime in theory, $\rho \sim 1.47$). This has a slight discrepancy with experimental results, where the minimal threshold is obtained at $\rho \sim 1.51$.

2.4 Coupling Between a Single Cylinder and a Waveguide

The coupling between a cylinder and a waveguide is studied as an extension of the original work about lasing in a single GaAs nanocylinder. It is important to have an efficient mechanism of coupling of light to waveguides (which are the most fundamental elements of on-chip photonic devices). This system is considered from the point of view of practical applications of such lasers to show the possibility to use single-particle quasi-BIC lasers as on-chip photonic sources.

The dimensions of the cylinder are chosen at the supercavity mode point discussed above. The waveguide and the cylinder are placed side by side, as shown in [Figure 2.13](#). A rectangular waveguide of width 500 nm is considered to have the same height and refractive index as the cylinder. For the vacuum case and the case of the system on a substrate with an HSQ capping layer on top, the material parameters are the same as the ones mentioned above.

A single electric dipole is used to excite the supercavity mode of the character TE_{312} (see the [Methods](#) for more details). Coupling efficiency is defined as follows: $\eta = (P_1 + P_2)/P_d$, where P_i is the power transmitted through the i th power monitor located in the cross-section of the waveguide (see [Figure 2.13](#)); P_d is the power injected into the simulation region (dipole power). The mode that is excited in the rectangular waveguide has the TM_{02} character ($H_x = 0$, $E_x \neq 0$ where x is the axis of the waveguide); the first and second indices are the indices along y and z axes, respectively (see [Figure 2.13](#)).

Figure 2.13. Illustration of radiation coupling between a dielectric cylinder and a waveguide in vacuum. An electric dipole placed inside the cylinder is used to excite the supercavity mode. Distance between the edge of the cylinder and the facet of the waveguide d_{ef} is altered in numerical simulations. Power monitors 1 and 2 are used to record the power flow inside the waveguide.

Coupling efficiency is studied as a function of distance d_{ef} between the edge of the cylinder and the facet of the waveguide (see [Figure 2.14A](#)). The Q factor of the cylinder is recorded as well (see the [Methods](#) for more details). It is important to analyze the coupling efficiency and the Q factor simultaneously because the optimum configuration of the system involves having a non-negligible fraction of light going into the waveguide, but not too much either, since that would spoil the cavity.

For the vacuum case, the cylinder of diameter $D = 505$ nm, height $H = 345$ nm and $\epsilon = 13$ is considered. The Q factor of 970 is reached when the distance d_{ef} exceeds 320 nm. Such quality factor was also obtained for a standalone cylinder at the supercavity mode point, as shown in [Figure 2.1C](#). Distance $d_{ef} = 340$ nm corresponds to the quality factor $Q = 980$, $\eta = 4\%$ for $\lambda = 831$ nm.

Figure 2.14. Simulated coupling efficiency maps η for the system consisting of a GaAs cylinder and a GaAs rectangular waveguide ($\epsilon = 13$) in vacuum (A) and on a substrate (B) shown as a function of wavelength λ and distance d_{ef} between the edge of the cylinder and the facet of the waveguide. The peak positions and the Q factors of the supercavity mode TE_{312} , obtained from the Q factor simulations, are shown by white circles; their area is directly proportional to the value of the Q factor.

It is important to study the case of the substrate and HSQ presence to determine the minimal distance d_{ef} at which the Q factor is high enough for gain-loss compensation. Here the cylinder of diameter $D = 500$ nm and height $H = 340$ nm is considered. The resultant coupling efficiency map is shown in Figure 2.14B. When increasing the distance, the Q factor reaches 260 (minimal Q factor at which lasing was obtained for a standalone cylinder, see Figure 2.10A at $D = 520$ nm) at $\lambda = 832$ nm for distance $d_{\text{ef}} = 200$ nm. These parameters correspond to the coupling efficiency $\eta = 3.5$ %.

Such small values of the coupling efficiency can be explained by the fact that the radiation of the supercavity mode occurs predominantly out-of-plane (see Figure 2.2D). Around one-third of the power is being radiated in-plane, which results in the maximum coupling efficiency of around 35 %, as shown in Figure 2.14A (vacuum case). When the substrate is introduced, the losses are higher, and the maximum coupling efficiency drops to ~ 10 %, as depicted in Figure 2.14B. At moderate distances $d_{\text{ef}} \sim 200$, the quality factor is ~ 260 ,

which is enough to obtain lasing and get a coupling efficiency of $\sim 3.5\%$. This is presumably enough for detection in our experimental setup. For that, of course, couplers of different kinds can be designed.¹³¹ This can be outlined as a point in future research directions.

2.5 Summary

In summary, using the concept of quasi-bound states in the continuum or supercavity modes, we have demonstrated lasing action at cryogenic temperatures in a single GaAs nanocylinder fabricated on a quartz substrate. To my knowledge, it is the first demonstration of lasing using a supercavity mode in a single nanoparticle (see Figure 2.15A). The nanolaser has a size-to-wavelength ratio as low as 0.6, which, to the best of our knowledge, is one of the lowest values demonstrated for purely semiconductor nanolasers so far ($D = 500$ nm, $H = 330$ nm, $\lambda \sim 830$ nm). Compared to the device shown in ref 23 ($\zeta = 0.72$) we have obtained a smaller size-to-wavelength ratio $\zeta = 0.6$ (see Figures 2.15A,B). That became possible by engineering two resonances supported by the system, namely, the Mie resonance and the Fabry–Perot resonance, to achieve a quasi-bound state in the continuum, the so-called supercavity mode, *via* mutual destructive interference of the resonances in the far field at the targeted lasing wavelength. To our knowledge, it is the first fully semiconductor laser using the third azimuthal order mode. Moreover, as compared to previous approaches, the present design does not need the particle to be suspended in air and does not rely on modes of high azimuthal orders, which increases the robustness of the device.

Figure 2.15. Summary of Chapter 2. (A) The first demonstration of lasing using a supercavity mode in a single nanoparticle. (B) SEM image of one of the smallest fully dielectric-based nanocavity lasers so far.

In addition to that, lower-order supercavity modes (zero-, first- and second-

order) have been studied theoretically for the cylinders of smaller sizes (diameters of ~ 400 nm and below). This part is not presented in the thesis because, even though we have fabricated the GaAs nanostructures for that purpose, we have not managed to obtain lasing using these modes in semiconductor cylinders. That was not possible due to lower Q factor modes ($Q \sim 100$ and below, for a particle on a substrate), which did not allow the gain-loss compensation in the experiment.

Coupling between a semiconductor cylinder (supporting the supercavity mode) and a waveguide has been studied theoretically. The optimal distance $d_{ff} \sim 200$ nm between the cylinder edge and the waveguide facet has been obtained for the case of the system on a substrate. It has been optimized for maintaining a sufficient Q factor for lasing while having the highest coupling efficiency.

Chapter 3

3 Lasing in Cuboids

3.1 Concept

We have reached the lower size limit for the lasing realization in a single nanoparticle. The next logical step is to obtain lasing in the coupled particles of smaller size using lower order azimuthal modes. In the case of a single nanoparticle, the radiation pattern is omnidirectional. Coupled particles give an opportunity for directivity control, which can be useful for out-of-plane coupling into a waveguide. We started with a single cylinder. After that, a dimer consisting of coaxial cylinders could be considered. However, it is more practical to study coaxial cuboid dimers from a fabrication perspective.

It is critical to theoretically obtain high Q factors for nanocavity in order to reduce the lasing threshold in the experiment. Recently, a system of two coaxial cylinders has been studied where the Q factor enhancement has been achieved by designing a system to have multipole conversion from the lower order multipole to the higher-order multipole.^{132, 133} That can be interpreted as suppression of radiation into the initially dominant radiation channel, as discussed in the introduction (see [Figure 1.11D](#)).

One of the recipes for increasing the Q factor of the coupled coaxial cylinders consists of two steps. First, the geometrical parameters corresponding to the

supercavity mode point are obtained for a single cylinder. After that, two identical cylinders having the optimal dimensions obtained are displaced (see [Figure 3.1A](#)).¹³³ At a large distance between the cylinders $L \gg h$ (where h is the height of the cylinder), the resonances are degenerate and coincide at a single point shown as a cross in [Figure 3.1B](#). Once the distance is decreased, the degeneracy is lifted because of the disks' weak coupling *via* the leaky resonances.¹³² This results in symmetric and antisymmetric hybridizations of the resonant modes, which near fields are displayed in the insets of [Figure 3.1B](#). The symmetric and anti-symmetric modes (with respect to the XY -plane) can also be referred to as bonding and anti-bonding modes, respectively. That is because the lower frequency bonding mode induces optical binding force, which pushes the disks closer to each other. The higher frequency anti-bonding mode pulls the cylinders apart.¹³³ Significant enhancement of the Q factor is due to the suppression of the lowest order term in a multipole expansion of the far-field radiation.¹³³ At different optimal distances, the Q factor reaches the maximum for both these modes (see [Figure 3.1C](#)). The symmetric mode's Q factor reaches ~ 500 for diameters ~ 450 nm at $\lambda = 830$ nm (see panel D in [Figure 3.1](#)). This diameter is smaller than $D \sim 500$ nm for the mode TE_3 in the supercavity mode point discussed in [Chapter 2.3](#). Hence, this symmetric mode of zero azimuthal order can be considered as a candidate for lasing in cuboid dimers in my study. It is possible to further enhance the Q factor by simultaneously optimizing the cylinder dimer's geometrical parameters.¹³³

Figure 3.1. (A) Coaxial cylinders ($\epsilon = 12$) in vacuum irradiated by Bessel beams. Evolution of the resonances (B) and the Q factor (C) as functions of the distance between the cylinders for $a/h = 0.71$. (D) Peak positions of the high Q factor zero azimuthal order modes in coaxial cylinders. Panels (A–C) are adapted with permission from ref 133, © 2020 Optical Society of America.

For the case of $\epsilon = 12$ cylinders in vacuum, maximum Q factors obtained in ref 133 were around 5500 for $kD \sim 4.37$ and on the order of 15000 for $kD \sim 6.26$, where k is the wavenumber and D is the diameter of each cylinder (see panel D in Figure 3.1). We work with GaAs-based nanolasers; thus, our system has to be optimized for the wavelength 830 nm (center of the gain bandwidth in GaAs at liquid nitrogen temperature, as discussed in Chapter 2.1). If we consider using the two above-mentioned high Q factor modes from that study for obtaining lasing in our experiment, we need to work with cylinders of diameters $D \sim 580$ nm ($kD \sim 4.37$) and $D \sim 830$ nm ($kD \sim 6.26$, see panel D in Figure 3.1). We have already demonstrated lasing for a single GaAs cylinder of diameter as small as $D \sim 500$ nm (which is the transverse size of the structure). All in all, we need to study different modes to minimize the size of our nanolaser. As mentioned above, we decide to use cuboids (see Figure 3.2) instead of cylinders because coaxial cuboids are more practical in nanofabrication.

Figure 3.2. Illustration of dielectric cuboids in vacuum. (A) Single dielectric cuboid in vacuum with highlighted dimensions: length L , width W , and height H . (B) Cuboid dimer in vacuum with highlighted dimensions: the distance between cuboids centers d (referred simply as “distance” further in the text); distance between cuboids facets d_{ff} . Further in the text, the cuboids in the dimer are referred to as the first and second cuboid or the left and right cuboid.

This part of the thesis is about finding the highest Q factor modes in cuboid dimers for obtaining lasing. These findings do not allow obtaining the smallest possible dielectric nanolaser, but they are interesting from the fundamental perspective. Specifically, compared to a single nanocylinder, presented in the previous chapter, the structures of smaller transverse sizes supporting lower azimuthal order modes will be discussed further in the text. The goal is to reduce the size of the particles further to obtain lasing at even lower order modes but compensate for the loss in the Q factor by adding another coupled particle. Also, having a few coupled particles gives additional possibilities to design nanoantenna-type lasers with controlled emission directivity.

There are two main approaches to optimization used in this chapter. The first one consists of optimizing a single cuboid to support a quasi-BIC of zero azimuthal order and displacing two equivalent cuboids to obtain the optimal distance (which refers to the highest Q factor for the dimer). The second approach consists of simultaneous optimization of all geometrical parameters

of the system to obtain the maximum Q factor at the target wavelength.

3.2 Lasing by the Use of Zero Azimuthal Order Mode in Square Cuboids

3.2.1 Numerical Optimization of a Single Cuboid

The advantage of the first approach is its simplicity. As a first step, I study the modes of a single square cuboid in vacuum (Figure 3.2A). This geometry is chosen to closely mimic a cylinder to obtain a maximum Q factor for zero-order mode. The scattering efficiency map is depicted in Figure 3.3A. It is very similar to what was obtained for a single cylinder. The Q factors of zero-order mode near the supercavity condition are shown by black circles (see the Methods for more detailed information on scattering and Q factor simulations). Maximum Q factor is obtained for dimensionless parameters $x_L \sim 1.18$, $\rho_w \sim 1.27$ (point A). For $\lambda = 830$ nm, these correspond to a square cuboid of length $L \sim 312$ nm, width $W \sim 395$ nm, and $Q = 180$, which is slightly lower than for the similar mode in a single cylinder ($Q \sim 200$).³⁸

Figure 3.3. Simulated characteristics of a single GaAs cuboid ($\epsilon = 13$) in vacuum. (A) Scattering efficiency map Q_{sca} as a function of the dimensionless parameters $x_L = \pi L/\lambda$ (the size parameter, λ is the wavelength in vacuum) and $\rho_w = W/L$ (the cuboid aspect ratio). The alternative x- and y-axis correspond to the wavelength and width

obtained when the cuboid's length is fixed at $L = 312$ nm. The peak positions and the Q factors of the mode, obtained from the Q factor simulations, are shown by black circles; their area is directly proportional to the value of the Q factor. The results of the Q factor simulations are shown for the quasi-TE mode of azimuthal order $i = 0$. The Q factor reaches its maximum value for the optimal parameters indicated by the point A, which is the supercavity mode point. (B, C) Simulated near fields of the supercavity mode in the YZ (B) and XY (C) planes of the cuboid, respectively. The white lines in (B, C) show the cuboid boundaries. (D) Simulated 3D far-field radiation pattern corresponding to the point A. (E) 2D projection of the simulated far-field radiation pattern corresponding to the point A. The white dashed circle shows the range of wavevectors collected by a 0.55 NA objective (similar to the one used to collect the emission in the lasing experiment).

Beyond the observed peak in the Q factor, to complete the characterization of the supercavity mode, near fields and far-field radiation pattern are shown in [Figures 3.3B–E](#). [Figures 3.3B,C](#) highlight the fact that the mode discussed is the zero azimuthal order mode of character TE_{012} . [Figure 3.3D](#) gives an additional confirmation that the mode at the point A is indeed a supercavity mode (a similar far field was obtained for zero-order supercavity mode in a single cylinder).³³ Compared to a single cylinder, a cuboid does not have a continuous axial symmetry (only under discrete, 90-degree rotation). Thus, the far field does not have it either, which is easier to notice in the far-field projection in [Figure 3.3E](#).

3.2.2 Numerical Optimization of Cuboid Dimers. Optical Experiments

Let us consider a cuboid dimer where sizes for each cuboid matches the sizes for the point A in [Figure 3.3A](#). By altering the distance between cuboids (see [Figure 3.2B](#)), it is possible to increase the Q factor of the modes. As a first step, the normal excitation is studied for TE polarization ($E_x = 0$, $H_x \neq 0$, see the left inset of [Figure 3.4A](#)); the corresponding scattering map is depicted in [Figure 3.4A](#). However, due to the symmetrical nature of the source used, not all TE modes are excited (the antisymmetric ones are missing). A good idea is introducing asymmetry in the excitation to get a comprehensive picture of TE modes present. This is carried out by tilting the k -vector by an angle θ with respect to the z -axis. In particular, the angle of 40° is used for oblique excitation; angle dispersion, which is less than 10° , has been taken into account when computing the scattering efficiency (see the [Methods](#) for more detailed information). For the case of oblique irradiation (see the inset of [Figure 3.4B](#)), an antisymmetric mode has emerged in the spectra (see [Figure 3.4B](#)).

The Q factors of zero azimuthal order modes are plotted by black circles overlaid with the scattering maps (see [Figures 3.4A,B](#)). More details on Q factor simulations are discussed in the [Methods](#). A maximum Q factor of 510 has been obtained for the mode $^s\text{TE}_{01}$ in the point β in [Figure 3.4A](#). In this mode notation, TE stands for the dominant polarization, s refers to a symmetric mode with respect to the YZ-plane, the first numeric index denotes the azimuthal order of the mode while the second index denotes its radial order. The axial index is not used for this particular case. It should be noted that the Q factor has a broad peak as a function of distance. This can be explained as follows. Cuboids designed in an above-described way act like low-effective-reflectivity mirrors, and at a moderate distance, the condition of the Fabry–Perot resonance is fulfilled, which results in a maximum Q factor ($Q = 510$) in the point β in [Figure 3.4A](#). The Q factor simulations in vacuum show that the symmetric mode $^s\text{TE}_{01}$ is advantageous compared to the

antisymmetric mode $^a\text{TE}_{01}$ (see Figure 3.4B). Thus, initially, it has been chosen as a candidate for lasing.

Figure 3.4. Simulated characteristics of a GaAs square cuboid dimer ($\epsilon = 13$) in vacuum. Width and length of each cuboid is fixed: $W = H = 393$ nm, $L = 312$ nm. (A, B) Scattering efficiency maps Q_{sca} as a function of wavelength and distance between the cuboid centers obtained using the normal excitation (A) and the tilted excitation (B). Schematics of the plane-wave irradiation are displayed in the insets. The right inset of panel (A) shows the near field of the mode in XZ-plane (the white lines highlight the cuboids' boundaries). The peak positions and the Q factors of the symmetric $^s\text{TE}_{01}$ and antisymmetric $^a\text{TE}_{01}$ modes obtained from the Q factor simulations are shown by black circles; their area is directly proportional to the value of the Q factor. The Q factor reaches its maximum value of 510 for the optimal parameters indicated by the point β where distance is $d = 612$ nm).

The near fields and the far-field radiation pattern are shown in Figure 3.5 to complete the characterization of the mode $^s\text{TE}_{01}$ at optimal parameters. Figure 3.5A confirms that the mode has zero azimuthal order. Figure 3.5B shows the mode structure in XY-plane, which matches well to that in XZ-plane (the right inset of Figure 3.4A), as expected for the case of a dimer in vacuum. The symmetric nature of the mode is demonstrated by Figures 3.5C,D. The far-field radiation pattern, shown in Figures 3.5E,F, exhibits strong out-of-plane radiation lobes, which is exactly what we want.

Figure 3.5. Simulated characteristics of a GaAs square cuboid dimer ($\epsilon = 13$) in vacuum at optimal parameters for the mode $^s\text{TE}_{01}$: $W = H = 393$ nm, $L = 312$ nm, $d = 612$ nm, $\lambda = 830.7$ nm (the point β). (A, B) Simulated near fields in the YZ (A) and XY (B) planes of the system, respectively. Simulated near-field components in the XZ (C) and XY (D) planes of the system, respectively. White lines in (A–D) show the cuboids' boundaries. (E) Simulated 3D far-field radiation pattern. (F) 2D projection of the simulated far-field radiation pattern. The white dashed circle shows the range of wavevectors collected by a 0.55 NA objective (similar to the one used to collect the emission in the lasing experiment).

Before discussing the experiment, it is important to consider the numerical simulations for the structures on a substrate with HSQ capping layer on top (see Figure 3.6A). For a single square cuboid, when going from the vacuum case to the substrate case, the Q factor drops from 180 to 110. At the same time, the optimal parameters are roughly the same, within a 5 nm difference compared to the vacuum case. So, the same width and length of the cuboids are used when analyzing the dimers ($W = H = 393$ nm, $L = 312$ nm) placed on a substrate $n_{\text{sub}} = 1.46$ with HSQ capping layer $n_{\text{HSQ}} = 1.41$. Quality factors of both modes mentioned above are plotted by a set of red circles in Figure 3.6B. The mode $^a\text{TE}_{01}$ is antisymmetric or anti-bonding (see inset 2 of Figure 3.6B). The mode $^s\text{TE}_{01}$ is symmetric or bonding (see inset 5 of Figure 3.6B).

It can be seen that for the mode $^s\text{TE}_{01}$, the Q factor drastically decreases from 510 to 160 when introducing the substrate. This occurs due to the nature of that mode. Specifically, the near-field leakage into the substrate significantly reduces the Q factor. The near field leakage into the substrate is smaller for the antisymmetric mode $^a\text{TE}_{01}$ (see inset 3 of Figure 3.6B) than that of the symmetric mode $^s\text{TE}_{01}$ (see inset 6 of Figure 3.6B). Thus, the mode $^s\text{TE}_{01}$ is less likely to lase than the antisymmetric mode $^a\text{TE}_{01}$, which has a Q factor on the order of 200 for distances in the vicinity of $d = 710$ nm. The far-field projections for both modes are shown in Figures 3.6C,D. The mode $^a\text{TE}_{01}$ possesses out-of-plane radiation lobes, which is exactly what we want. Thus, the mode $^a\text{TE}_{01}$ is a promising candidate for lasing.

Figure 3.6. (A) Illustration of the Q factor simulations for a cuboid dimer on a substrate. (B) Peak positions of the modes supported by a GaAs square cuboid dimer ($\epsilon = 13$) on a substrate, retrieved from the Q factor simulations, shown in the region of dimensional parameters of interest. Width and length of each cuboid is fixed: $W = H = 393$ nm, $L = 312$ nm. The peak positions and the Q factors of the symmetric $^s\text{TE}_{01}$ and antisymmetric $^a\text{TE}_{01}$ modes obtained from the Q factor simulations are shown by red circles; their area is directly proportional to the value of the Q factor. High Q factor ($Q = 200$) for the antisymmetric mode occurs near a point α_1 where $d = 712$ nm (no pronounced maximum). For the antisymmetric mode, the Q factor reaches its maximum value for the optimal parameters indicated by a point β_1 where $d = 512$ nm, $Q = 160$. The insets 1–3 and 4–6 show the near fields of the modes

$^a\text{TE}_{01}$ (at the point α_1) and $^s\text{TE}_{01}$ (at the point β_1), respectively. The white lines in the insets 1–6 show the cuboids' boundaries. 2D projection of the simulated far-field radiation pattern corresponding to the points α_1 (C) and β_1 (D). The white dashed circles in (C, D) show the range of wavevectors collected by a 0.55 NA objective (similar to the one used to collect the emission in the lasing experiment).

The structures are fabricated by the same top-down fabrication method, as explained in [Chapter 2.3](#) and the [Methods](#). An exemplary image of the fabricated dimer is shown in [Figure 3.7A](#). The experimental optical setup used here is the same as described in [Chapter 2.3](#) and the [Methods](#). For the case of the single GaAs cylinder, the simulated Q factor was around 280. Here, for a GaAs dimer, a typical Q factor is ~ 200 , which makes the gain-loss compensation more challenging. We have managed to record a lasing peak for several cuboid dimers (see [Figure 3.7B](#)). However, the lasing peak is sustained in the spectra only for a short time of around 10 seconds. Such a short operation time of the laser can be explained by the fact that the lasing threshold exceeds the damage threshold of the system. High pumping fluences are used to obtain lasing: $F = 460 - 760 \text{ } \mu\text{J}/\text{cm}^2$. At lower pumping fluences, lasing has not been observed. For comparison, the lasing threshold for a single GaAs cylinder from [Chapter 2.3](#) of height $H = 330 \text{ nm}$, diameter $D = 500 \text{ nm}$ is equal to $300 \text{ } \mu\text{J}/\text{cm}^2$, and the device can operate in the lasing regime at $F = 433 \text{ } \mu\text{J}/\text{cm}^2$ for an extended period of time (1 hour of which, alone, was used to measure the autocorrelation function shown in [Figure 2.8C](#)).

The lasing peak obtained in the experiment is slightly blue-shifted (see [Figure 3.7B](#)) from the simulated peak position of the mode $^a\text{TE}_{01}$ (see [Figure 3.7C](#)). This can be attributed to the band-filling effect. It should be noted that for the lasing experiment, longer cuboids are used ($L = 325 \text{ nm}$) because for the cuboids of $L = 315 \text{ nm}$, lasing has been observed only for a TM mode at wavelengths $\sim 780 - 790 \text{ nm}$. The lasing spectra peak is in good agreement with the theory (see [Figures 3.7B,C](#)). We obtained lasing in a narrow range of distances where the Q factor is relatively high (see [Figure 3.7C](#)).

Figure 3.7. (A) Exemplary SEM image of the fabricated nanostructure. (B) Experimental lasing spectra for cuboids displaced by a set of distances (for the fixed cuboid dimensions $H = 390$ nm, $W = 390$ nm, $L = 325$ nm). (C) Theoretically obtained peak positions and Q factors of the antisymmetric mode $^aTE_{01}$ retrieved from the Q factor simulations for a cuboid dimer $\epsilon = 13$ on a substrate, shown in the region of dimensional parameters of interest. The area of red circles is directly proportional to the value of the Q factor. Width and length of each cuboid is fixed: $W = H = 393$ nm, $L = 312$ nm. The blue dashed ellipse highlights the vicinity of distances at which lasing has been obtained in the experiment.

3.2.3 Summary

In conclusion, the obtained Q factor for the square cuboid dimers operating in the zero-order azimuthal mode reaches a value around 200, which is not sufficient to observe sustained lasing for more than 10 seconds for our system. In general, there are three ways for improvement:

1. Designing a higher Q factor system.
2. Using a lower pulse repetition frequency to improve the operation time of the device.
3. Using new materials with better gain, resulting in a lower lasing threshold or a higher damage threshold for bringing these conditions far apart from each other.

While the second and the third ways are challenging from the experimental perspective, the first one still allows some improvement by numerical simulations.

In addition, we have studied similar designs based on a cuboid trimer and cuboid tetramer. Theoretically, we have managed to obtain Q factors above 300 (square cuboid trimer) and Q factors approaching 400 (square cuboid tetramer) for the particles on a substrate of width $W = H \sim 390$ nm and distances between particle facets $d_{ff} \sim 450$ nm. The structures have been fabricated, but lasing could not be detected by our experimental setup with numerical aperture $NA = 0.55$ because the radiation is predominantly in-plane when a larger number of particles is considered. Thus, the part with trimer and tetramer has not been included in the thesis. We have implemented a different approach for the Q factor maximization for a dimer, as discussed further in the text.

3.3 Optimization of an Arbitrary Cuboid Dimer for Operation in the Second Azimuthal Order Mode. Fabrication of the Nanostructures

3.3.1 Results and Discussion

The second simulation approach consists of optimizing all parameters simultaneously. For cuboid dimers, there are four parameters to optimize: height, width, length, distance; the Q factor needs to be high enough to obtain lasing at the target wavelength of 830 nm (from our experience, this means at least 280 in the numerical simulations for the structure on a substrate, as to obtain lasing for an extended time). So, I have defined a fitness function as a Q factor. This terminology is taken from ref [134](#). The purpose is to obtain the highest Q factor possible for dimers on a substrate with their height and width in the vicinity of 400 nm or below. This is to get a significant decrease in the nanostructure's transverse size from what was obtained for a single cylinder ($D = 500$ nm, as noted in [Chapter 2.3](#)).

As a first step, we consider the best strategy to vary only a subset of the parameters and study the structures on a substrate right away. Thus, first, only square cuboids are considered ($W = H$). In numerical simulations, I have defined the solution space^{[134](#)} as follows: the cuboids' length is fixed at $L = 300$ nm, and we scan $W = H = 300 - 500$ nm and $d = 300 - 500$ nm independently. The spectra retrieved from the Q factor simulations are analyzed using the following dimensionless parameters: $x_L = \pi L / \lambda$ (the size parameter where the characteristic dimension of the system is the length of the cuboid, λ is the wavelength in vacuum), and two aspect ratios of the system, namely $\rho_d = d/L$ and $\rho_H = H/L$. After the set of optimal dimensionless parameters x_L^* , ρ_d^* , ρ_H^* corresponding to the maximum Q factor is obtained, dimensional parameters corresponding to the same mode at the target wavelength $\lambda = 830$ nm are retrieved using the following formulas: $L = x_L^* \cdot \lambda / \pi$, $d = \rho_d^* \cdot L$, $H = W = \rho_H^* \cdot L$.

For square cuboid dimers, optimal parameters need to satisfy two conditions mentioned above (highest possible Q factor above 280 and transverse size in the vicinity of 400 nm or below). Specifically, the following conditions on geometrical parameters are considered: $H < 410$ nm, $L < 350$ nm for any d . Using this method and taking into account the conditions mentioned above, the optimal dimensionless parameters obtained using the Q factor simulations are (see the [Methods](#) for more details): $x_L^* \sim 1.176$, $\rho_d^* \sim 1.067$, $\rho_H^* \sim 1.267$. For an operational wavelength $\lambda = 830$ nm, these correspond to a square cuboid dimer with length $L \sim 311$ nm, distance $d \sim 331$ nm, and height $H \sim 393$ nm. The corresponding Q factor reaches a maximum value of $Q = 320$, which is presumably enough to obtain lasing. Exemplary one-dimensional graphs of the Q factor in dependence on distance and height in the vicinity of optimal parameters highlight the maximum Q factor (see [Figure 3.8](#)).

Figure 3.8. Simulated Q factor in the solution space for GaAs square cuboid dimers ($\epsilon = 13$) on a substrate. Q factor in dependence on distance (blue line, $H = W = 380$ nm) and height (green line, $d = 320$ nm) for square cuboids of length $L = 300$ nm. Upper axis shows the dimensionless parameters $\rho_d = d/L$ and $\rho_H = H/L$.

In the first step for the optimization of the parameters, a relatively wide range of dimensional parameters has been considered ($d = 300 - 500$, $H = W = 300 - 500$) using a big step of 20 nm for altering both parameters. This has been carried out to approximately determine the optimal parameters of the cuboid first, leaving a more precise analysis of the system at the second step.

At the second step, I narrow the solution space (near the optimum obtained at the first step), decrease the step, and alter more parameters (considering arbitrary cuboids). Here, the solution space is defined as follows, the length is fixed at $L = 310$ nm, and for each distance d in the range from 310 to 350 nm, both height H and width W are altered independently in the range from 380 to 420 nm. A step of 10 nanometres is used for altering all the sizes just mentioned. All the size ranges have been chosen in the vicinity of optimal parameters obtained for the square cuboid dimers. The simulation method consisting of the two steps described above is used to obtain the highest Q factor modes using a relatively short time consumed by simulations.

For arbitrary cuboid dimers on a substrate, the optimum parameters need to satisfy the two conditions mentioned above (the highest possible Q factor above 280 and transverse size in the vicinity of 400 nm). Specifically, the following conditions on geometrical parameters are considered: $H < 420$ nm, $W < 420$ nm, $L < 350$ nm for any d . The optimal dimensionless parameters obtained at the second optimization step are: $x_L^* \sim 1.17$, $\rho_d^* \sim 1.06$, $\rho_H^* \sim 1.23$, $\rho_w^* \sim 1.32$. For an operational wavelength $\lambda = 830$ nm, these correspond to a cuboid dimer with length $L \sim 310$ nm, distance $d \sim 330$ nm, height $H \sim 380$ nm, and width $W \sim 410$ nm. The corresponding Q factor reaches its maximum value of $Q = 400$, which is greater than that of the square cuboid dimer on a substrate ($Q = 320$).

The near fields and far-field radiation pattern of the mode at optimal geometrical parameters are shown in [Figure 3.9](#). This mode has a mixed polarization and cannot be identified as a TE or TM mode. However, it is still possible to classify the mode using indices of cylindrical coordinates. This mode has HE_{223} character, where the first, second, and third indices are azimuthal, radial, and axial indices, respectively, and HE denotes the non-transversal character of the mode. In other words, this is the second azimuthal order mode (see [Figures 3.9A–C](#)), and its Q factor 400 is presumably enough to obtain lasing. The 2D projection of the simulated far-field radiation pattern

shown in Figure 3.9D is promising for the lasing experiment (the radiation of this mode can be collected by our 0.55 NA objective).

Figure 3.9. Simulated characteristics of a GaAs cuboid dimers ($\epsilon = 13$) on a substrate at optimal parameters of the arbitrary cuboid dimer – length $L = 310$ nm, distance $d = 330$ nm, height $H = 380$ nm, and width $W = 410$ nm ($Q = 400$, $\lambda = 830$ nm). Simulated near fields (A–C). The white lines show the cuboids’ boundaries. The dashed lines 1 and 2 in (C) highlight the antinodes where near fields (A) and (B) have been recorded, respectively. (D) 2D projection of the simulated far-field radiation pattern. The white dashed circle shows the range of wavevectors collected by a 0.55 NA objective (similar to the one used to collect the emission in the lasing experiment).

For a single cuboid with dimensions $L = 640$ nm, $H = 380$ nm on a substrate, which would be equivalent to the volume occupied by the whole dimer, the Q factor reaches its maximum $Q = 340$ for width $W = 420$ nm. This means that filling the small ~ 20 nm gap between the cuboids (*i.e.*, the distance between the facets of the cuboids d_{ff}) reduces the Q factor. In other words, a dimer is indeed advantageous compared to a single particle with the same overall dimensions. However, in the experiment, it is hard to obtain such a small gap, as discussed next.

We have managed to fabricate cuboid dimers of height 400 nm, which is in the close vicinity of the optimal parameters. The exemplary images of the

fabricated nanostructures are displayed in [Figure 3.10](#). It is essential to note that for some cases, we had GaAs pillars formed on top of a quartz substrate (see [Figures 3.10A,C](#)). Pillar formation could occur after the final etching step for the following reasons. Residues from the upstream fabrication steps could act like a micromask. We used Cl_2 and N_2 gases for the etching of GaAs. Etching products could also act as masks. Pillars formation can be eliminated by optimizing the etching recipe, *i.e.*, flow rates, pressure, power, *etc.* Pillars have extremely small sizes. Thus, they cannot support optical modes at high wavelengths ~ 830 nm. So, they have a negligible contribution to the collected radiation, which we have checked in the experiments.

The smallest distance between the centers of the cuboids at which the gap between them is still distinguishable is $d = 370$ nm (this dimer is shown in [Figure 3.10A](#)). The length of each cuboid is $L \sim 305$ nm, which corresponds to a distance between facets of the cuboids $d_{\text{ff}} \sim 65$ nm. At a distance $d = 360$ nm between the cuboids, shown in [Figure 3.10B](#) ($L \sim 305$, $d_{\text{ff}} \sim 55$ nm), the cuboids are still separated by a gap, but it already becomes slightly narrower at the substrate level. For the smaller distances between the cuboids ($d_{\text{ff}} < 55$ nm), the gap between them is not distinguishable anymore (see [Figure 3.10C](#), where the layout distance between cuboid centers is 340 nm).

The numerically optimal distance between the cuboids is $d = 330$ nm, as discussed above (for $L = 310$ nm, this corresponds to $d_{\text{ff}} = 20$ nm). The smallest distance at which the fabricated cuboid dimer has a distinguishable gap is $d_{\text{ff}} = 55 - 65$ nm. This is higher than the optimal value, but it is important to check whether the Q factor of the mode is high enough to obtain lasing (Q factor has to be at least 280 in simulations, based on our previous results).

Figure 3.10. Exemplary SEM images of the fabricated cuboid dimers of height $H = 400$ nm. (A) Cuboid dimer with length $L = 305$ nm and distance $d = 370$ nm ($d_{ff} \sim 65$ nm). (B) Cuboid dimer with length $L = 305$ nm and distance $d = 360$ nm ($d_{ff} \sim 55$ nm). (D) Cuboid dimer with layout distance $d = 340$ nm (it was supposed to have $d_{ff} \sim 35$ nm).

The results of the Q factor simulations for dimensions closely matching the experiment are shown in Figure 3.11A. Mode 1 has a maximum Q factor at width $W \sim 410$ nm for wavelength $\lambda = 834$ nm denoted by a point β where $Q = 300$ (optimal point). The corresponding near fields in the different planes of the system are displayed in Figures 3.11B–E. This mode has HE_{223} character (the same as discussed in Figures 3.9A–C). This is the second azimuthal order mode. Using a lower axial order supercavity mode TE_{212} (also of the second azimuthal order) in a single cylinder on a substrate allows shrinking the system twice in the longitudinal direction. However, the Q factor of the second-order supercavity mode for such a small cylinder on a substrate is ~ 100 . For two cuboids, we have a Q factor ~ 300 , which is presumably enough to obtain lasing.

The 2D far-field projection corresponding to the point β is depicted in Figure 3.11G. When the width is detuned from the optimum (point β in Figure 3.11A), the far-field projection changes drastically, as shown in Figures 3.11F,H. This occurs because of different dominant multipoles in the radiation (it will be explained further in the text).

Figure 3.11. Simulated characteristics of a GaAs ($\epsilon = 13$) cuboid dimer of height $H = 400$ nm, distance $d = 360$ nm and length $L = 305$ nm on a substrate. (A) Peak positions and Q factors of the modes retrieved from the Q factor simulations, shown in the region of dimensional parameters of interest. The area of circles is directly proportional to the value of the Q factor. Mode 1 is a candidate for lasing in the experiment (that mode is highlighted by red color). Near fields (B–E) corresponding to the point β . The white lines show the cuboids' boundaries. The dashed lines 1 and 2 in (E) highlight the antinodes where near fields (B) and (C) have been recorded, respectively. 2D projection of the simulated far-field radiation pattern corresponding to the point α , width $W = 370$ nm (F), point β , width $W = 410$ nm (G), and point γ , width $W = 480$ nm (H). The white dashed circles show the range of wavevectors collected by a 0.55 NA objective (similar to the one used to collect the emission in

the lasing experiment).

The quality factor $Q = 300$ has been achieved for $\lambda = 834$ nm at height $H = 400$ nm, $W = 410$ nm, length $L = 305$ nm, distance between cuboid facets $d_{ff} = 55$ nm. If this gap is filled, meaning that a long cuboid of length $L = 665$ nm and height $H = 400$ nm is considered on a substrate, the Q factor for the similar mode reaches the same value $Q = 300$ for $\lambda = 870$ nm at width $W = 410$ nm (the mode peak redshifts when the gap is filled). Furthermore, at greater width $W = 430$ nm, the Q factor reaches a maximum value of 350 for wavelength $\lambda = 883$ nm. The geometrical parameters can be easily rescaled to obtain the same Q factor at the desired wavelength $\lambda = 830$ nm. This means that when choosing between a dimer and a single long cuboid from the experimental point of view, the second one is better because it is easier to fabricate and it has a slightly higher Q factor ($Q = 350$ compared to a dimer with $d_{ff} = 55$ nm, $Q = 300$). However, in theory, a cuboid dimer is better than a long cuboid because introducing a small gap $d_{ff} = 20\text{--}30$ nm for a dimer gives an advantage ($Q = 400$ for a dimer compared to a long cuboid with $Q = 350$). Thus, such a small gap needs to be obtained in nanofabrication to get a higher Q factor for a cuboid dimer. That can be carried out using a thinner HSQ cap, which can be outlined as a recommendation for further research.

It is important to explain how the Q factor reaches its maximum at optimal parameters in the point β in [Figure 3.11A](#). For the case of this system, the discussion has been about the dimer on a substrate. This has been performed intentionally to obtain high Q factors for a more realistic situation. At this point, the cuboids can be analyzed in vacuum to explain the concept.

The results of the Q factor simulations for the cuboid dimer in vacuum having the dimensions discussed above are shown in [Figure 3.12A](#). Mode 1 has a maximum Q factor at width $W = 400$ nm for wavelength $\lambda = 819.6$ nm, denoted by the point β , where $Q = 1230$ (optimal point). It is important to note that the corresponding near field patterns ([Figures 3.12B–E](#)) match well with what has been discussed for the optimal point for this mode when the dimer is on a

substrate (Figures 3.11B–E). This confirms that the same mode is being analyzed for both cases (dimer in vacuum and on a substrate).

The 2D far-field projections are depicted in Figures 3.12F–H. They match well with what has been shown for the case of the substrate (Figures 3.11F–H). The three-dimensional far-field radiation pattern for the points α , β , and γ are depicted in Figures 3.12I–K. The mechanism for obtaining the maximum Q factor here is the same as for a quasi-BIC in a single cylinder. It consists of optimizing the structure's geometrical parameters, where the optimal parameters refer to the case of suppression of the radiation through the initially dominant radiation channel, as discussed in Chapter 1 (see Figures 1.11D,E).

In general, the far field can be expanded into a multipole series, and each multipole can be referred to as a radiation channel.⁴⁰ At optimal parameters, the radiation of the initially dominant channel is suppressed. This can be interpreted as a decoupling of a leaky mode from the radiation continuum, which creates a quasi-bound state in the continuum. The optimal parameters correspond to the supercavity mode point,³⁸ and at that point, radiation through the initially dominant channel becomes negligible (suppressed), and it predominantly occurs through a different channel (*i.e.*, a different multipole). A good example of this concept has been demonstrated in ref 40. There, for a single dielectric cylinder, at diameters below and above the optimum, the dominant radiation channel is an electric dipole. The radiation occurs predominantly through a magnetic quadrupole for the optimum cylinder diameter, while the radiation through the electric dipole (the initially dominant channel) is suppressed (see Figure 1.11D). Due to multipole conversion, the far-field radiation pattern drastically reshapes while passing the supercavity mode point, as discussed in Chapter 1 (see Figure 1.11E).⁴⁰

For our system, the situation is similar. When increasing the width from $W = 330$ nm (point α in Figure 3.12A) to $W = 400$ nm (point β in Figure 3.12A), the initially dominant radiation channel is suppressed, which results in a significant change of the radiation pattern (see Figures 3.12I,J). The same

thing occurs (see Figures 3.12J,K) when decreasing the width from $W = 440$ nm (point γ in Figure 3.12A) to $W = 400$ (point β in Figure 3.12A). All in all, for both cases of cuboids on a substrate (Figure 3.11) and in vacuum (Figure 3.12), the mechanism for obtaining the maximum Q factor consists of engineering the system to support a supercavity mode.

Figure 3.12. Simulated characteristics of a GaAs ($\epsilon = 13$) cuboid dimer of height $H = 400$ nm, distance $d = 360$ nm and length $L = 305$ nm in vacuum. (A) Peak positions and Q factors of the mode 1 retrieved from the Q factor simulations, shown in the region of dimensional parameters of interest. The area of circles is directly proportional to the value of the Q factor. Near fields (B–E) corresponding to the point β . The white lines show the cuboids' boundaries. The dashed lines 1 and 2 in (E) highlight the antinodes where near fields (B) and (C) have been recorded,

respectively. 2D projection of the simulated far-field radiation pattern corresponding to the point α , width $W = 330$ nm (F), point β (optimal point, $Q = 1230$), width $W = 400$ nm (G), and point γ , width $W = 440$ nm (H). The white dashed circles show the range of wavevectors collected by a 0.55 NA objective (similar to the one used to collect the emission in the lasing experiment). Simulated 3D far-field radiation patterns corresponding to the point α (I), point β (J), and point γ (K).

The dimensions considered for the cuboid dimers in vacuum and on a substrate (Figure 3.11 and Figure 3.12, respectively) have been chosen to match the dimensions of the structures we managed to fabricate ($H = 400$ nm, $L = 305$ nm, $d = 360$ nm, shown in Figure 3.10B). However, the optimal dimensions for the vacuum case are slightly different. We targeted the optimization for the substrate case from the beginning because it is more relevant from the experimental point of view. Besides, to have a full picture, I have optimized the dimers' parameters in vacuum and obtained the mode's characteristics at the quasi-BIC point.

To optimize the square cuboid dimers in vacuum, the following conditions have been considered for the geometrical parameters: $H < 450$ nm, $L < 350$ nm for any d . The optimal dimensionless parameters obtained are the following: $x_L^* \sim 1.19$, $\rho_d^* \sim 1.13$, $\rho_H^* \sim 1.27$. For an operational wavelength $\lambda = 830$ nm, these correspond to a cuboid dimer with length $L \sim 315$ nm, distance $d \sim 360$ nm, height $H = W \sim 400$ nm.

To ensure that the mode discussed here is the same as in Figure 3.12, the near fields are displayed in Figures 3.13A–C (they match well with Figures 3.12B–E). The corresponding Q factor reaches its maximum value of $Q = 1300$ (see Figure 3.13), which is greater than that of the cuboid dimer discussed in Figure 3.12, where the maximum Q factor is $Q = 1230$ (there the dimer dimensions match the parameters of the fabricated dimers which are slightly off the optimum). Even such a small change in size (from $L = 305$ for the cuboid in Figure 3.12 to $L = 315$ in Figure 3.13) results in a more symmetric radiation pattern (see Figures 3.13D,E compared to Figures 3.12G,J).

Figure 3.13. Simulated characteristics of a GaAs square cuboid dimers ($\epsilon = 13$) in vacuum at optimal parameters of the dimer – length $L = 315$ nm, distance $d = 360$ nm, height $H = 400$ nm ($Q = 1300$, $\lambda = 830$ nm). Near fields (A–C). The white lines show the cuboids' boundaries. The dashed lines 1 and 2 in (C) highlight the antinodes where near fields (A) and (B) have been recorded, respectively. (D) 2D projection of the simulated far-field radiation pattern. The white dashed circle shows the range of wavevectors collected by a 0.55 NA objective (similar to the one used to collect the emission in the lasing experiment). (E) Simulated 3D far-field radiation pattern.

3.3.2 Summary

In [Chapter 3.2](#), where the square cuboid dimers were studied, the optimal parameters for a dimer on a substrate were $W \sim 390$ nm, $Q = 200$, $d_{ff} \sim 400$ nm, $L \sim 310$ nm for operation in zero azimuthal order mode $^aTE_{01}$. Thus, we obtained a smaller transverse size than that of a single cylinder ($D = 500$ nm, see [Chapter 2.3](#)). The design approach used in numerical simulations in [Chapter 3.2](#) consisted of first, optimizing a single cuboid to support a quasi-BIC of zero azimuthal order at the target wavelength, utilizing a single dipole for mode excitation. And second, two equivalent cuboids were displaced to obtain the optimal distance (max Q factor). We managed to achieve lasing in such dimers for a short time because the lasing threshold was greater than the damage threshold (the Q factor was insufficient for stable operation).

For the case of the arbitrary cuboid dimers discussed in [Chapter 3.3](#), we have obtained a greater Q factor corresponding to the following optimal parameters: $W = 410$ nm, $Q = 400$, $d_{ff} = 20$ nm, $L = 310$ nm, $H = 380$ nm for operation in non-transversal mode HE_{223} of the second azimuthal order (supercavity mode). That design is promising to increase nanolaser lifetime because the Q factor is twice greater than the one obtained in [Chapter 3.2](#). In [Chapter 3.3](#), I have performed a simultaneous optimization of all geometrical parameters of the system to obtain the maximum Q factor at the target wavelength. That also includes using nine random dipoles for excitation to look for the highest Q factor mode among the modes of all possible polarizations. As a result, for the arbitrary cuboid dimers, the Q factor obtained for the mode HE_{223} at optimum is twice greater than that of $^aTE_{01}$ in the square cuboid dimers. Besides, HE_{223} is the highest Q factor mode discussed in the thesis for a nanostructure on a substrate. For the smallest fabricable distances, the optimal parameters for the mode HE_{223} are the following: $W \sim 410$ nm, $Q = 300$, $d_{ff} = 55$ nm, $L = 305$ nm, $H = 400$ nm. This Q factor is slightly greater than the maximum Q factor 280 for a single GaAs cylinder on a substrate, as discussed in [Chapter 2.3](#). Thus, the optical experiments were not performed for those dimers after they were fabricated. The smaller gaps

($d_{\text{ff}} = 20\text{--}30$ nm) need to be obtained in nanofabrication to get a significantly greater Q factor in a cuboid dimer. That can be carried out using a thinner HSQ cap, which can be outlined as a recommendation for further research.

Analyzing the GaAs cuboids on top of a quartz substrate, I conclude that the highest Q factors are achievable using the concept of supercavity modes for simultaneous optimization of all geometrical parameters of a nanostructure while considering modes of all possible polarizations.

Chapter 4

4 Conclusion and Recommendations

4.1 Conclusion

In summary, using the concept of quasi-bound states in the continuum or supercavity modes, we have demonstrated lasing action at cryogenic temperatures in a single GaAs nanocylinder fabricated on a quartz substrate. To my knowledge, it is the first demonstration of lasing using a supercavity mode in a single nanoparticle. The nanolaser has a size-to-wavelength ratio as low as 0.6, which, to the best of our knowledge, is one of the lowest values demonstrated for purely semiconductor nanolasers so far ($D = 500$ nm, $H = 330$ nm, $\lambda \sim 830$ nm, see [Figure 4.1A](#)). Compared to the device shown in [ref 23](#) ($\zeta = 0.72$) we have obtained a smaller size-to-wavelength ratio $\zeta = 0.6$. That became possible by engineering two resonances supported by the system, namely, the Mie resonance and the Fabry–Perot resonance, to achieve a quasi-bound state in the continuum, the so-called supercavity mode, *via* mutual destructive interference of the resonances in the far field at the targeted lasing wavelength. To our knowledge, it is the first fully semiconductor laser using the third azimuthal order mode. Moreover, as compared to previous approaches, the present design does not need the particle to be suspended in air and does not rely on modes of high azimuthal orders, which increases the robustness of the device.

Coupling between a dielectric cylinder (supporting the supercavity mode) and a waveguide has been studied theoretically (see Figure 4.1B). The optimal distance $d_{ff} \sim 200$ nm between the cylinder edge and the waveguide facet has been obtained for the case of the system on a substrate. It has been optimized for maintaining a sufficient Q factor for lasing while having the highest coupling efficiency.

Figure 4.1. Summary. (A) We have reached the lower size limit for the lasing realization in a single dielectric nanoparticle ($D = 500$ nm, $H = 330$ nm, $\lambda \sim 830$ nm). Lasing threshold $\sim 300 \mu\text{J}/\text{cm}^2$ and stable operation at $F = 430 \mu\text{J}/\text{cm}^2$ for an extended time have been achieved for such GaAs nanolaser. One hour of that time, alone, has been used to measure the autocorrelation function. Optimal dimensions in numerical simulations for a GaAs cylinder ($\epsilon = 13$) on a substrate at $\lambda = 830$ nm: $D = 500$ nm, $Q = 280$, $H = 340$ nm. (B) We have obtained the optimal distance for sufficient Q factor and coupling between a single cylinder and a waveguide. (C) We have achieved a smaller transverse size for a square cuboid dimer having out-of-plane radiation lobes. Optimal parameters in numerical simulations for a GaAs square cuboid dimer ($\epsilon = 13$) on a substrate at $\lambda = 830$ nm: $W = H \sim 390$ nm, $Q =$

200, $d_{ff} \sim 400$ nm, $L \sim 310$ nm. Lasing for such GaAs dimers has been obtained for a short time. The lasing threshold of around $400 \mu\text{J}/\text{cm}^2$ is greater than the damage threshold. (D) We have obtained a twice greater Q factor for an arbitrary cuboid dimer. Optimal dimensions in numerical simulations for an arbitrary cuboid dimer of $\epsilon = 13$ on a substrate at $\lambda = 830$ nm: $W = 410$ nm, $Q = 400$, $d_{ff} = 20$ nm, $L = 310$ nm, $H = 380$ nm. That design is promising to increase nanolaser lifetime.

We have achieved a smaller transverse size for a square cuboid dimer having out-of-plane radiation lobes for a zero azimuthal order mode. Optimal parameters in numerical simulations for a GaAs square cuboid dimer ($\epsilon = 13$) on a substrate at $\lambda = 830$ nm: $W = H \sim 390$ nm, $Q = 200$, $d_{ff} \sim 400$ nm, $L \sim 310$ nm (see [Figure 4.1C](#)). Lasing for a square cuboid GaAs dimer of dimensions $W = H = 390$ nm, $d_{ff} \sim 380$ nm, $L = 325$ nm has been obtained for a short time of ~ 10 seconds (unlike for the case of the single cylinder, $t \sim 1$ hour and beyond). The lasing threshold of around $400 \mu\text{J}/\text{cm}^2$ was greater than the damage threshold. The Q factor of such a square cuboid dimer was insufficient for gain-loss compensation. It motivated us to use a different approach for Q factor maximization.

Using the concept of supercavity modes, we have obtained twice greater Q factor for an arbitrary cuboid. The second azimuthal order mode has been used for that purpose. The smaller transverse size of the structures has been reached (width $W = 410$ nm compared to a cylinder of diameter $D = 500$ nm), resulting in Q factors on the order of 400 for the distance between cuboid facets $d_{ff} = 20$ nm. Optimal dimensions in numerical simulations for an arbitrary cuboid dimer ($\epsilon = 13$) on a substrate at $\lambda = 830$ nm: $W = 410$ nm, $Q = 400$, $d_{ff} = 20$ nm, $L = 310$ nm, $H = 380$ nm (see [Figure 4.1D](#)). That design is promising to increase nanolaser lifetime because the Q factor is twice greater than for the approach discussed above. Such small gaps $d_{ff} = 20$ nm need to be obtained in nanofabrication to get a significantly greater Q factor in a cuboid dimer. That can be made using a thinner HSQ cap, which can be outlined as a recommendation for further research.

Analyzing the GaAs cuboids on top of a quartz substrate, I conclude that the

highest Q factors are achievable using the concept of supercavity modes for simultaneous optimization of all geometrical parameters of a nanostructure while considering modes of all possible polarizations.

4.2 Recommendations for Further Research

The approach of minimization of the laser size using the concept of supercavity modes can further be brought to room-temperature operation by using higher gain materials or surface passivation techniques. GaAs is known to have strong nonradiative recombination due to surface defects, which significantly lowers its gain property. There are two ways to reduce the surface recombination: first, using surface passivation by chemical method (passivating using sulfide compounds such as P_2S_5 , $(NH_4)_2S$, Na_2S , etc.), and second, using a passivation layer like AlGaAs or multi-quantum-well (MQW) structure of AlGaAs/GaAs. Such a MQW structure, which is widely used in high-efficient LEDs and lasers, is a good candidate for room-temperature operation for this type of laser if we can preserve its emitting property during the dry-etching process.

Among the passivation methods mentioned, the best positive impact will be from using a passivation layer of $Al_xGa_{1-x}As$ /GaAs structure. The layer of wide bandgap semiconductor $Al_xGa_{1-x}As$ on top of the GaAs nanolaser surface acts as an electronic barrier to prevent carrier diffusion to the surface, suppressing surface recombination. The outer layer of GaAs prevents the $Al_xGa_{1-x}As$ layer from oxidation.¹³⁵ Each of the layers mentioned above can be less than 10 nm thick.^{135, 136} Passivation of GaAs using sulfide compounds such as ammonium polysulfide can also be implemented. However, this method results in shorter-term surface stability as compared to GaAs passivation using wide-bandgap semiconductors like $Al_xGa_{1-x}As$ just mentioned.¹³⁷

In the future, with the use of electrical pumping and radiation outcoupling strategies, the presented concepts can be useful for smaller, Ohmic-loss-free nanolasers, with potential applications in photonic circuits and beyond.

While several scientific groups have studied a directivity control in dielectric nanoparticle array-based lasers, research about directive lasing in a few nanoparticles-based designs still has many goals to accomplish. This direction is particularly challenging for out-of-plane configurations of small

dielectric lasers.

Chapter 5

5 Methods

5.1 Numerical Simulations

5.1.1 Single Cylinder

Numerical simulations were performed using FDTD commercial software (Lumerical FDTD Solutions). There are two types of numerical simulations carried out in this work: nanoparticle scattering simulations and Q factor calculations. The cylinder was irradiated by a plane wave using a total-field scattered-field (TFSF) source in nanoparticle scattering simulations. This source separates the scattered field from the incident field, which allows us to compute scattering efficiency Q_{sca} . The scattering efficiency was obtained using the following relation,¹³⁸ $Q_{sca}(\lambda) = W_{sca}(\lambda) \cdot (I_s(\lambda) \cdot D \cdot H)^{-1}$, where λ is the wavelength of radiation, W_{sca} is the power scattered through a box monitor (closed surface) surrounding the cylinder, I_s is the intensity of the source irradiating the nanoparticle, D is the diameter of the cylinder, and H is its height. Two types of plane waves were considered for excitation; namely, TE-polarized ($E_z = 0$, $H_z \neq 0$, [Figure 2.1A](#)) and TM-polarized ($H_z = 0$, $E_z \neq 0$), where z is the axis of the cylinder and E_z and H_z are the projections of electric field and magnetic field onto the z -axis, respectively. From the scattering simulations, the scattering efficiency map $Q_{sca}(\lambda, D)$, shown in [Figure 2.1C](#),

was obtained.

To compute the quasi-normal eigenmodes and the corresponding Q factors, one electric dipole was placed inside the dielectric cylinder to excite the modes selectively. To do so, the dipole was placed in the vicinity of the antinode of the electric field of the desired mode with its dipole moment parallel to the electric field at that point (as, for example, depicted in [Figure 2.2A](#), for excitation of quasi-TE modes). It is important to note that a single-dipole excitation was performed as a final step after the eigenmodes in the cylinder were obtained by the use of nine electric dipoles randomly placed inside the cylinder. The latter approach allowed me to find the antinodes of the modes and make sure that I did not miss high Q factor non-transversal modes in the spectra in the vicinity of parameters of interest when exciting the modes selectively using a single dipole at a subsequent step. Selective excitation of the modes by a single electric dipole improves the accuracy of the Q factor simulations compared to that while using nine random dipoles. That is because a smaller number of peaks need to be fitted in the spectra (separately for each polarization) at the next step discussed further in the text. Time-dependent electric and magnetic fields were recorded using nine pointlike time monitors, randomly placed inside the cylinder. After that, the spectrum was calculated for each monitor using a fast Fourier transform (FFT). Then the spectra were averaged over the number of monitors obtaining the spectrum for Q factor calculations. The resonant peaks in the spectrum were fit by a Lorentzian function to obtain Δf_i , fwhm for the i th resonance. The Q factor of the i th resonance was calculated using $Q = f_i/\Delta f_i$, where f_i is the frequency of the i th resonance. The far-field 3D pattern of the modes was computed by the use of a box monitor completely surrounding the cylinder. The modes' far-field 2D pattern was obtained using a plane monitor (perpendicular to the z-axis) located above the cylinder.

To account for the real experimental conditions, additional numerical simulations and optimization were performed considering that the dielectric nanocylinder is placed on a quartz substrate and has a capping HSQ layer on top, as can be seen in the SEM image in the inset of [Figure 2.3B](#). In those

simulations, we assumed that the cylinder had a constant relative permittivity $\epsilon = 13$, that the HSQ cylinder on top had a height of 150 nm and a constant refractive index $n_{\text{HSQ}} = 1.41$, and that the substrate had a constant refractive index $n_{\text{substrate}} = 1.46$.

In numerical simulations, there was zero absorption, emission, and gain. Those assumptions were made for the proof of concepts. The imaginary part of every material's dielectric constant was equal to zero in all numerical simulations presented in the thesis. That assumption allowed me to observe the full evolution of the resonant peak in the scattering spectra associated with the formation of the supercavity mode (from broad to narrow and back to broad, as in [Figure 2.1C](#)). During my PhD, I performed several Q factor simulations taking into account the absorption and dispersion of GaAs. The effect of loss (absorption) on the Q factor was $\sim 20\%$ reduction. For $\epsilon = 13$ and zero absorption, the maximum Q factor for a cylinder on a substrate was 280. For non-zero absorption and dispersion of GaAs cylinder at 77 K, the maximum Q factor dropped to ~ 230 in numerical simulations for a cylinder on a substrate. That Q factor was sufficient for lasing, which was proved by our experiments.

The Mie-FP mode formulation was used in the thesis. Thus, it is important to explain its feasibility for the systems I studied. Mie modes or whispering gallery modes are formed by the total internal reflection of the radiation traveling along the circumference of a spherical particle. The Mie theory was developed for spherical particles. However, 3D Mie theory can also be applied for a cylinder of height significantly smaller than the wavelength: $H \ll \lambda$.¹³⁹ For a high cylinder $H \gg \lambda$, 2D Mie theory can be implemented to solve the scattering problem. For a cylinder of moderate height $H \sim \lambda$, the modes supported are hybrid Mie–Fabry–Perot modes.¹³⁹ Mie modes or whispering gallery modes are formed by the total internal reflection of the radiation traveling along the circumference of the cylinder. Thus, their peak positions in the spectrum are highly sensitive to the cylinder diameter. Fabry–Perot (FP) modes are the modes formed by the multiple reflections of the guided mode

supported by the cylinder at its top and bottom interfaces. Thus, their peak positions in the spectrum are not highly sensitive to the cylinder diameter. This is one of the criteria by which Mie- and FP-type modes can be distinguished.

In general, the modes studied in the cylinder in [Chapter 2.2](#) are hybrid Mie–Fabry–Perot modes. However, in some cases, it is possible to highlight the mode's dominant character using the near-field pattern or the criteria mentioned above. In the supercavity mode point (SM point), the dominant character (Mie or Fabry–Perot) cannot be revealed. The supercavity mode (quasi-BIC) is a hybrid FP-Mie mode. Hence, the SM point can be interpreted as the point where Mie and Fabry–Perot resonances are degenerate.

The higher the Q factor is, the lower the lasing threshold is. However, the cavity's quality factor is not the only thing that the lasing threshold depends on. Thus, the Q factor is not the only parameter that can be accounted for during clarification of which mode is preferred for lasing. The gain-mode overlap analysis can give more insight into why one of the spectrally close modes is preferred for lasing. As discussed above (see [Figure 2.11A](#)), two spectrally close modes overlap with the gain spectrum – modes TE₃ and TM₄₁₀. Above in the text, these modes are compared in terms of the quality factor. The lasing threshold (the threshold photon density pump rate) can be expressed as follows:⁴⁹

$$P_{th} = \frac{h\nu (1 + \beta)}{\eta A} \left[\frac{\gamma}{\beta \Gamma} + F \frac{2n_0 V_a}{\tau_0} \right] = P_1 + P_2 \quad (5-1)$$

Here, $h\nu$ is the photon energy of the emission; η is the proportion of pump power absorbed by the gain medium; A is the area of the pump beam; β is the spontaneous emission factor; γ is the cavity loss rate (excluding intrinsic loss attributed to the gain material); gain-mode overlap Γ is the fractional overlap of the lasing mode volume V_m and the volume of the active medium V_a ;⁴¹ F is the Purcell factor; $2n_0$ is the carrier density at twice the transparency value; τ_0 is the natural spontaneous emission lifetime.⁴⁹

Equation 5-1 shows that the lasing threshold has two contributions: P_1 refers to the cavity mode loss; P_2 refers to the intrinsic loss of the gain medium.⁴⁹ In my research, P_1 is minimized by obtaining the maximum Q factor to reduce radiative losses. Taking into account the following expressions: $P_1 \propto \nu\gamma/\Gamma$; $\gamma \propto 1/Q$; $\Gamma \approx V_m/V_a$; $\nu = c/\lambda$, we can estimate $P_1 \propto (\lambda Q \Gamma)^{-1}$. We performed modal volume simulations and used the following formula to calculate the modal volume: $V_m = \int \epsilon |\mathbf{E}|^2 dV / \max(\epsilon |\mathbf{E}|^2)$, where the integral is taken over the box monitor volume surrounding the cylinder.

Using this approach, I can compare the contribution P_1 into the lasing threshold for the modes TE₃ at the point β_3 and TM₄₁₀ at the point ζ_4 (see Figure 2.11A). The calculated parameters for these two points in the solution space are as follows, β_3 ($H = 300$ nm, $D = 420$ nm, $\lambda = 708$ nm): TE₃, $Q = 270$, $V_m = 1.68 \cdot 10^7$ nm³ = $17.75(\lambda/2n)^3$, $\Gamma \approx V_m/V_a = 0.4$. ζ_4 ($H = 300$ nm, $D = 420$ nm, $\lambda = 721.4$ nm): TM₄₁₀, $Q = 600$, $V_m = 9.98 \cdot 10^6$ nm³ = $9.97(\lambda/2n)^3$, $\Gamma \approx V_m/V_a = 0.24$, where $n^2 = 13$. The solution space dimensions were used during numerical simulations. After I input the desired height $H = 332$ nm, the dimensions are recalculated using the size parameter x_H and the aspect ratio ρ : β_3 ($H = 332$ nm, $D = 465$ nm, $\lambda = 783.5$ nm): TE₃, $Q = 270$, $\Gamma \approx 0.4$. ζ_4 ($H = 332$ nm, $D = 465$ nm, $\lambda = 798.3$ nm): TM₄₁₀, $Q = 600$, $\Gamma \approx 0.24$. From these expressions, it is clear that the gain-mode overlap for the mode TE₃ is ~65 % greater than that of the mode TM₄₁₀.

Implementing the formula $P_1 \propto (\lambda Q \Gamma)^{-1}$ discussed above, we can estimate the following ratio in theory: $P_1(\beta_3) / P_1(\zeta_4) \approx 1.36$. In experiment $Q(\text{TE}_3) \approx 230$, $Q(\text{TM}_{410}) < 200$. That is because for the mode TE₃, larger fields are away from the lateral surface of the resonator compared to the mode TM₄₁₀ (see Figures 2.11B–E). Thus, surface roughness has a greater effect on the Q factor reduction for the mode TM₄₁₀. Consequently, a more realistic estimation: $P_1(\beta_3) / P_1(\zeta_4) \approx 0.5$. Thus, when the Q factor of the mode TE₃ is high enough (the dimensions are close to the supercavity mode point), its contribution P_1 to the threshold is nearly twice smaller than that of the mode TM₄₁₀. So, near the supercavity regime, lasing first occurs in the mode TE₃. It saturates the

gain to balance its losses. The saturated gain value is already not enough for loss compensation of the mode TM_{410} , which is why its power vanishes.¹⁴⁰ This explains why we did not obtain lasing for the mode TM_{410} for diameters above 465 nm (see Figure 2.11A). There the mode TE_3 lases alone.

5.1.2 Cuboid Dimers

Numerical simulations for cuboid dimers were performed in the same way as for the single cylinder with the following exceptions.

Scattering simulations in Figure 3.4B were performed using a TFSF source with oblique incidence with respect to the z-axis. In that case, the injection angle was wavelength-dependent. The central angle of incidence was $\theta = 40^\circ$, and the dispersion was $\sim 10^\circ$. The scattering efficiency was calculated using the following formula: $Q_{sca}(\lambda) = W_{sca}(\lambda) \cdot \cos(\theta) \cdot (I_s(\lambda) \cdot 2L \cdot W)^{-1}$, where λ is the wavelength of radiation, W_{sca} is the power scattered through a box monitor (closed surface) surrounding the dimer, I_s is the intensity of the source irradiating the nanoparticle, L is the length of the cuboid, and W is its width. The factor of 2 stands for a cuboid dimer.

Q factor simulations in Chapter 3.2 were performed in the following way for the cuboid dimers: one electric dipole was placed inside the cuboid 2 in the antinode of the mode excited (see Figure 3.6A). The signal was collected by 9 time monitors randomly placed inside the cuboid 1.

The same approach for collecting the signal was used in Chapter 3.3. There, the modes were excited using nine electric dipoles randomly placed inside the cuboid 2. That is a better approach for non-transversal modes excitation compared to a single dipole case.

The Q factor simulation results were additionally checked using the approach of placing the nine time monitors randomly inside the volume comprising both the cuboids and the gap in between. The results are in good correspondence with the primary simulation method mentioned above.

5.2 Sample Nanofabrication

GaAs wafer with an epitaxial structure of i-GaAs (500 nm)/AlAs (100 nm)/undoped $\langle 100 \rangle$ GaAs (625 nm) was purchased from Semiconductor Wafer, Inc., Taiwan. Epitaxial lift-off¹²⁰ technique was used to transfer the top, high-quality GaAs film (500 nm) from the wafer to a quartz substrate. In the process, black wax (*i.e.*, Apiezon W) was applied on the top i-GaAs layer and thermally annealed at 90 °C for about 30–60 min to realize a slightly domed surface. After that, hydrofluoric acid (HF) solution (~5 wt % in H₂O) was used to etch the sacrificial layer (AlAs), and GaAs epitaxial film was then lifted off. A direct bonding process was used to attach the film to the quartz substrate using a wafer bonder system (EVG 510). Inductively coupled plasma reactive ion etching (ICP RIE) tool (Oxford PlasmaLab System 100) was used to etch GaAs film to the desired thickness. Standard electron-beam lithography procedure (Elionix ELS-7000 system) was used to fabricate nanocylinders mask using hydrogen silsesquioxane resist (HSQ, Dow Corning XR-1541-006) along with ESpacer (E-spacer 300Z) to improve the conductivity during e-beam exposure. The unexposed resist was removed using a 25 % tetramethylammonium hydroxide (TMAH) developer. Finally, the patterned GaAs film was etched using ICP RIE. Nanocylinders were fabricated with a 20 μm spacing. That period was chosen in such a way that each cylinder would be individually irradiated during subsequent optical measurements, thus acting as an isolated nanoparticle.

5.3 Optical Characterization

All optical measurements were performed in a microspectrometer setup, consisting of an inverted microscope (Nikon Ti-U) and a spectrometer system (Andor SR303i spectrograph coupled with a 400×1600 -pixel Electron-Multiplying Charge-Coupled Device - Newton 971 EMCCD). For lasing and photoluminescence measurements, a Ti:sapphire femtosecond laser (Coherent, MIRA 900) was used. The pumping wavelength was tuned to ~ 530 nm using an optical parametric amplifier (Coherent OPA 9450, RegA). The pulse duration and repetition rate after OPA are ~ 200 fs and 20 kHz, respectively. The laser beam was focused on the nanocylinder using a 50X dark-field (DF) microscope objective (Nikon, CFI LU Plan DF, the numerical aperture NA = 0.55), resulting in a laser spot of ~ 5 μm in diameter. The dark-field scattering was measured in a backward scattering configuration using the same microscope objective with white light excitation from a halogen lamp. Transmission of GaAs thin film was measured using another halogen lamp excitation. The absorption was calculated by the formula $A = 2 - \log_{10}(\%T)$. All spectrally resolved measurements were carried out with 150 grooves/mm, 800 nm blading grating, and the spectrograph slit width of 100 μm , resulting in a spectral resolution of ~ 1 nm. All measurements were performed at cryogenic temperatures. For that, the sample was placed onto the cold finger of a cryostat (Janis, ST- 500). The chamber was vacuumed to 3×10^{-3} Torr by an oil pump before being cooled to 77 K using liquid nitrogen.

During the lasing experiments, we did not consider the modal distribution of the 530 nm pump and its overlap with the modes of the emission at 825 nm. If the overlap is good, most absorbed photons will translate into emission photons of the lasing mode of interest (the target mode). If the overlap is poor, some absorbed photons will couple to other modes that will not contribute to lasing of the target mode, increasing its threshold. In other words, lasing in the target mode can be facilitated by reducing its threshold, which can be carried out by increasing the spatial overlap between the target mode and the

pump profile.¹⁴¹

We used a uniform pumping with a beam diameter of $\sim 5 \mu\text{m}$. While the pumping is uniform, several modes can lase at the same time if the gain can compensate their losses simultaneously. For example, in [Figure 2.10A](#) at diameters $\sim 600 \text{ nm}$, we observed lasing in two modes simultaneously. For smaller diameters, ~ 500 and below, the gain could compensate the losses for one mode, not allowing multimode lasing. In the disks of greater size, the number of lasing modes can increase significantly. In this case, if tuning of the lasing wavelength is required, a specific mode can be selected for lasing by the use of nonuniform pumping.

In ref [141](#), which is about microdisk lasers, it was shown that by the use of nonuniform pumping, it was possible to select any lasing mode to be the dominant one by suppressing all other modes *via* increasing their lasing thresholds. The optimized pump profile has a smaller spatial overlap with non-target modes while keeping the spatial overlap with the target mode almost the same as during uniform pumping. The target mode does not always have to have the lowest lasing threshold (to be the first one to lase) at optimum pump profile. The target mode can grow faster than the other modes and become the dominant one at high pumping levels. The spatial intensity distribution of the pump profile was shaped by the use of a spatial light modulator.¹⁴¹ From this study, I conclude that a poor spatial overlap between the pump profile and the target mode increases the lasing threshold for this mode. A poor modal overlap Γ (fractional overlap of the lasing mode volume V_m and the volume of the active medium V_a) also increases the threshold, as can be seen from [Equation 5-1](#). A higher threshold means higher power consumption, which lowers the efficiency of the emitter.

5.4 Autocorrelation Measurements

Autocorrelation measurements were carried out using a Hanbury–Brown–Twiss setup. The emission signal in our microscopic lasing measurement setup was routed into a multimode fiber (Thorlabs GIF625, 62.5 μm core). The signal was then sent to a 50:50 optical fiber beamsplitter with each node coupled with a low-jitter avalanche photodiodes (Micro Photon Devices, PDM series with a timing jitter of 35 ps fwhm). Coincident clicks on the two photodiodes were recorded using a time-tagging device (qutools, QuTAU, timing resolution 81 ps).

Author's Publications

Research Article Published:

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Poster presentation at the Conference:

Mylnikov, V.; Ha, S. T.; Pan, Z.; Valuckas, V.; Paniagua-Domínguez, R.; Demir, H. V.; Kuznetsov, A. I. Lasing Action in Single Subwavelength Particles Supporting Supercavity Modes. *ICMAT 2019*, Singapore (BEST POSTER AWARD).

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