

Active and Nonreciprocal nanophotonics

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A thesis submitted for the degree of

Doctor of Philosophy

The Australian National University

Research School of Physics



April 2023

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*Dedicated to Shri Guru Nanak Dev ji, Shri Guru Teg Bahadur Shrine Babe Ke in
Goulburn, Baba Nahar Singh Ji, Baba Kapoor Singh, Mata Kamaljeet Kaur and the
Sikh Community.*

Disclaimer

I hereby declare that the work undertaken in this thesis has been conducted by me alone, except where indicated in the text. I conducted this work between July 2020 and February 2024, during which period I was a PhD student at the Australian National University. This thesis, in whole or any part of it, has not been submitted to this or any other university for a degree.

This thesis has been compiled as a Thesis by Compilation in accordance with relevant ANU policies. Each of the 3 main chapters in this thesis have been published in peer reviewed journals. I have made significant contribution to each of these journal articles and have written the text of the papers myself, except where indicated otherwise.

A handwritten signature in black ink, reading 'Aditya Tripathi', with a horizontal line extending from the end of the name.

Aditya Tripathi

7 June 2025

Acknowledgements

I express special thanks to the spiritual support and blessings from His Holiness Sant Baba Kapoor Singh, Mata Kamaljeet Kaur and Guru Teg Bahadur Babe ke Shrine in Goulburn, New South Wales.



Figure 0.0.1: Sant Baba Kapoor Singh

Sant Baba Kapoor Singh Ji (Fig 0.0.1) is the main spiritual teacher at the Shrine as well as a leading exponent in the Sikh faith. Being a very noble and affectionate soul, he inherited exemplary sense of service and visionary approach from his great father and the founder of this organization, His Holiness Dhan Dhan Sant Baba Nahar Singh Ji (Fig 0.0.3). Today he owes responsibility to manage all the religious and educational institutions under the umbrella of Babe Ke Group of Institutions.

Mata Kamaljeet Kaur (Fig 0.0.2), the better half of Sant Baba Kapoor Singh Ji as well as a key figure behind the operations of the organization.

In the fields of creativity like research and performing arts, we encounter a peculiar phenomenon called the writer's block. Wherein people experience difficulties ranging from blankness of mind or thought to extreme laziness in coming up with a new piece or conclusion. Last year (2023) was one such time frame for me. No matter how hard I tried or worked, I couldn't come up with new results or even publish the existing ones. Whenever, I attempted complex experiments my mind went completely blank. The experiment of optical nonreciprocity was one such case, even though I knew what had to be done, and what I should be expecting I was petrified even by the thought of going inside the lab and searching for the components. It



Figure 0.0.2: Mata Kamaljeet Kaur

was something like losing the battle even before fighting it just as the French armies experienced against the advancing German armies in World War II. Consequentially, I contemplated giving up the field of Physics and pursuing a career in service sector or as a chef.

I was experiencing a cocktail of venomous emotions which were extremely depressing in nature, I was driven to the point of having no hope or urge to even live. Even the easily doable things like writing a review or plotting a simple Z-scan curve seemed nearly impossible. The last nail in the coffin were the unsurmountable financial difficulties that I was facing at the time pertaining to my sisters' education. Basically, life seemed like a sinking ship without a lifeboat. This issue till date is unsolvable for most of the psychiatrics and counsellors. Everyone simply told me that time will heal it and so on.

In this situation of hopelessness, I went to Guru Teg Bahadur Babe Ke Shrine on 494 Carrick Road, NSW. I had the fortune of meeting the main spiritual master of the Shrine: Sant Baba Kapoor Singh Ji. His loving smile and welcoming hug were enough to detoxify my cocktail of hazardous emotions. I spent a total of good ten days in his divine association.

He asked me a lot of scientific questions in this tenure. One such incident was when he asked me about termites and powderpost beetles. The question he posed was that why both termites and powderpost beetles feed on wood, but the termites excrete out mud and the powderpost beetles excrete out a white flour. I was dumbfounded by this question and tried to ask other scientists as well. Then after my enquiry, I

realized that science had no answer to why different products for same inputs. The entire congregation of devotees laughed in pleasure and I accepted that the divine creation can never be completely understood by gross science.

Every day, my morale and emotions boosted up and made my outlook towards life extremely optimistic. On the end of his trip, I asked him whether I would be able to graduate. He gave me a hug and said, “no issues”. I got the blessing I needed and started working extensively on thesis writing and publishing papers and surprisingly, everything started improving drastically.



Figure 0.0.3: Sant Baba Nahar Singh

I even started going regularly on Sundays to the Shrine to cook food. The shrine hosts a monthly program on the last Sunday of every month. I started doing regular cooking and food serving as a part of my regular community service. The devotees of the shrine and the Sikh community kept on blessing me for the successful completion of this thesis.

This Shrine on Carrick Road, NSW (Fig 0.0.4) was established in 2000 by His Holiness Sant Baba Nahar Singh Ji (father of Sant Baba Kapoor Singh Ji). Sant Baba Nahar Singh Ji is the grandson of Sant Baba Nand Singh Ji and one of the pioneers of Nanaksar tradition of Sikh faith. Sant Baba Nahar Singh Ji established the Babe Ke trust in India and built many educational institutions and hospitals. His temperament of relentless service is an inspiration for me.

The Shrine boasts of a huge 136-acre campus on a beautiful hillside in the council of



Figure 0.0.4: Guru Teg Bahadur Shrine Babe Ke 494 Carrick Road, Goulburn, NSW

Goulburn, NSW. As of now, the infrastructure hosts the main shrine, kitchen and a few rooms for living all free of charge. In the future, it would be developed as a beautiful spiritual retreat that showcases journeys of the ten spiritual teachers of the Sikh faith. Having personally experienced the bliss, peace and blessings provided by the shrine, I can say for sure that the place has the power to bless everyone alike.

Science and physics explain a lot of things, but this Shrine explains everything in our hearts and lives that cannot be explained by physics. Just like how I was trying my best, putting in efforts, yet couldn't generate results. Or how someone ends up depressed even after having everything and how even after having absolutely nothing people can be happy.

Since then, my hard work was blessed by the divine grace of Sant Baba Kapoor Singh ji and the Sikh community of Guru Teg Bahadur Shrine Babe Ke to yield this thesis. I can say with full confidence that anyone who visits the Shrine on 494 Carrick Road, NSW 2580 will return with unprecedented blessings and peace of mind.

Abstract

Recently, the field of nanophotonics has seen rapid developments in the area of dielectric metasurfaces governed by Mie scattering. The dielectric metasurfaces have shown remarkable versatility of their functionalities. Ranging from Huygens metasurfaces controlling directionality and phase of light scattering to sharp Fano-resonators to quasi-BICs and nonlinear metasurfaces supporting higher order nonlinear effects. However, active and nonreciprocal nanophotonics with metasurfaces has been facing challenges like miniaturization to the nanoscale and extreme suppression of Mie scattering by active media. Active and nonreciprocal metasurfaces acting as light sources and switches were difficult to realise. The last seven years several different strategies for the harmonious combination of optically active media and dielectric metasurfaces have been developed and demonstrated. However, the scope of fundamental approaches, detailed methodology and experimental demonstration of functional active nanophotonic devices remained a challenge. This hindered the development of active and nonreciprocal metasurfaces from fundamental to applications as prospective nano-optical devices.

In my Thesis, I focus on the design, development and demonstration of working active and nonreciprocal nanophotonic devices and their practical applicability as prospective nano-optical devices. I developed a general framework for design and demonstrated both light generation, active light modulation and directional control capabilities in the near infrared wavelength range. Amongst light sources, I demonstrated the capability to generate polarized luminescence at the nanoscale as well as nanolasing. And in case of light modulation, I demonstrated amplitude modulation and spectral modulation. The directional control is shown in nonreciprocal transmitters. Ultimately, I addressed the design and development challenges in the realization of nano-optical devices and their role in emergence of nanophotonics as a major player in upcoming industry.

In, Chapter 1 I describe the overall motivation behind this work and the concept of active and nonreciprocal nanophotonics in detail.

Chapter 2 delves into the methods used (both theoretical and experimental) throughout this research.

Chapter 3 and 4 elaborate on the design of active metasurfaces as light sources. Specifically, Chapter 3 talks about active metasurfaces which can produce polarized photoluminescence. These metasurfaces rely on coupling of polarized scattering from dielectric nanoparticles to the nanoparticles of rare earth doped nanoparticles. Our work is one of the first approaches that demonstrate polarization control in fluores-

cence emission from rare earth nanoparticles. In Chapter 4, highly confined fields are generated in membrane based dielectric nanostructures which have thin layers of quantum wells sandwiched within them leading to the earliest demonstration of topological and anapole nanolasing at room temperatures.

In Chapter 5, I elaborate on the maiden demonstrations of nanophotonic nonreciprocal light control devices. The light control is achieved by the deployment of a layer of phase change or highly nonlinear materials in the structure of the metasurface. First, I demonstrate amplitude and spectral modulation using vanadium dioxide based metasurfaces. Next, I show nonreciprocal transmission using ITO based metasurfaces.

In the final chapter, I summarise my findings and outline the future scope.

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Introduction

0.1 Nanophotonics

Prof. Richard Feynman mentioned in his talk 'There is plenty of room at the bottom' [1]. He said that if individual atoms are placed side by side forming a layer, standard physics may no longer be applicable to the system. This idea expanded into different fields such as nanochemistry, nanoelectronics, and nanophotonics. Nanophotonics is an extension of nanotechnology that studies the peculiar physics of light-matter interaction at dimensions comparable to the wavelength of light. Majority of research in the field is basically development and study of different light trapping mechanisms like plasmonics and metaphotonics. All these mechanisms exploit either the wave nature of light or the particle nature or in many cases a combination of both [2].

The first advancement in nanophotonics came with the introduction of plasmonic systems in the 1957. In plasmonics we deal with oscillations of collective charge in a metal (plasma)[3]. In case the space of oscillations is confined to nanoscopic dimensions in all directions, we observe localized surface plasmon resonances (LSPR) like in the case of gold or silver nanoparticles[4]. The other case deals with propagating charge oscillations also known as surface plasmons, where the charge movement is confined in one direction only thereby enabling motion along the metal surface[5]. Study of these effects have led to developments in the fields of plasmonic sensing [6], Surface Enhanced Raman Spectroscopies [6] and Surface Plasmon Resonance microscopy[7].

The second major boost to nanophotonics came with the development of photonic crystals in 1987 by Eli Yablonovitch[8] and Sajeev John[9]. They exploited Bragg diffraction between grating layers to replicate a bandgap-like phenomenon in semiconductors and came up with 'photonic bandgaps'. Consequently, these led to research in fields like photonic bandgap engineering, super continuum generation[10], localization[11] and hyper-transport[12] of light. This research ultimately culmi-

nated in the development of products like photonic crystal fibres, reflective layers for VCSELs and LEDs and waveguides which are manufactured by companies like OPALUX and IPG Photonics Corp.

The third and by far the most promising giant stride in the field was provided by the advent of dielectric metasurfaces and metapotonics in 2014 (Capasso)[13]. The Bragg-diffraction based photonic crystals are often constrained by the lattice period being greater than or at least equal to the effective wavelength of incident light. This limits the technology to mostly micrometer scales for infrared photonics. The need of multiple diffractors or a proper homogeneous lattice also adds significant size constraints. The way around was offered by collective interference of Mie scattering modes of individual dielectric nanoparticles [14, 15, 16]. The scattering from the individual Mie modes (Electric and Magnetic Dipole) in dielectric nanoparticles and their potential use in nanophotonics was discussed in [15]. These modes don't impose any constraint on the lattice constant thereby enabling subwavelength nanostructures. Also, the interference doesn't necessarily have to be from modes of different nanoparticles. Different modes from the same nanoparticle can also interfere to provide a myriad of optical effects. Basically, metasurfaces are artificial sheet based structures that manipulate electromagnetic fields in space using Mie modes [16]. This field has led to the development of many promising applications like flat lenses, holography, beam steering, retroreflectors and mid infrared components offered by companies like Samsung and Metalenz.

Another distinct area of advancement is the development of quantum optics. Experimental Quantum optics was first brought into emphasis after Hong-Ou-Mandel's interferometric experiment in 1990, where the many experimental aspects of quantum entanglement were revealed[17]. Eventually various platforms were suggested to study the quantum nature of light like spontaneous parametric down conversion (SPDC)[18], semiconductor quantum dots[19] and NV centers[20] in diamond. Nanophotonic structures are being considered for implementation of quantum optical systems. The underlying principle of quantum nanophotonic systems is the strong coupling between a single photon and a single pseudo atom of the metasurface.

Materials science plays an essential part in nanophotonics research. The innate properties of individual materials play a key role in deciding the role of the material in a system. Just like how different ingredients impart a certain taste in a dish. In case of nanophotonics, the selection of material can alter the working wavelengths as well as fundamental nature from being a scatterer to an absorber [21]. The choice of materials can give us control over many other properties like refrac-

tive index, nonlinear effects, phase changes and many more. Hence employing the right material can give us the desired light matter interaction in the nanophotonic device. For example noble metals like gold, silver can be used in doing plasmonics as they have a high free carrier concentration. And III-V semiconductors can be used for optoelectronic applications like making a nano Light emitting diode or even a nanolaser[22]. Materials like Silicon or titanium dioxide are great for engineering photonic band gaps or tuning interference of Mie resonances for making perfect reflectors or absorbers. Finely fabricated materials like semiconductor quantum dots under cryogenic conditions can give generate entangled photons on demand for quantum optical applications. Similarly rare earth doped nanoparticles can be used to develop devices with upconverting properties.

Applications of nanophotonics include sensing, holography, specialized optical components, specialized photovoltaics and nano-optoelectronics[23]. Plasmonic sensors are great for the purpose of detection of even the slightest change in the chemistry and the refractive index of the surrounding media. Owing to the strong light matter interaction and electromagnetic coupling of surface plasmons to the surrounding media. Arrays of Quantum dots have been demonstrated to show the ability to generate entangled photon pairs[19] and cluster states[24]. Semiconductor quantum dots hold great potential in becoming potential quantum processors and routers.

III-V semiconducting nanostructures in solar cells have been shown to demonstrate exceptional functionalities like use of nanoantennas for enhanced absorption, increment of the effective refractive index of the substrate and increment of local optical density of states.

Another application is the field of nanolasers[22], where III-V semiconductors are structured in the form of nanowires, quantum wells or other nanostructures like bound states in continuum. These are usually integrated with other chip scale devices like photonic integrate circuits, waveguides or modulators to provide stable platforms for optical data processing and computing.

0.2 Metasurfaces

Engineered materials which held desired properties or metamaterials had been introduced in science in 1967 [25]. This concept was developed as an idea to mimic natural materials and then tweak them to manipulate properties. Solid state physics unravels wonderful features like band gaps, grain boundaries, bragg diffraction and permanent magnetism. Metamaterials were the first attempt at artificial creation of solid state effects and hence allowed a greater property control than natural ma-

terials.

In early 2000's the idea of metamaterials had found its application in photonics in the fields of photonic crystal fibres, cloaking materials and negative index materials [26]. Success with 3D metamaterials inspired a nanotechnological shift in the field owing to the enticing prospect of reduced size and unexplored physics. This nanotechnological perspective was provided by the introduction of metasurfaces which were two dimensional metamaterials. These 2D metasurfaces were nothing but patterned thin films grown using epitaxial or sputter methods. This development facilitated practical demonstration of optical properties like retroreflection, perfect transmission or reflection and many more at the nanoscale.

Structurally, metasurfaces are two-dimensional lattices of individual light scatterers or resonators of sub wavelength dimensions. Each individual light scatter or resonator produces a set of electromagnetic scattering modes or Mie modes which are then allowed to interfere constructively or destructively to provide different fields and hence distinct functionalities[27]. Physically, metasurfaces are a system of interfering sub wavelength resonators which enable a myriad of possible optical consequences by altering individual resonators. One can look at it as an analogy of Riemann integrals, where the integral of a function is simply the sum of areas of small infinitesimal rectangles. Similarly, here our 'function' is the desired optical waveform and the 'infinitesimal rectangles' are the modes emanating from individual resonators. These are arranged in different geometries and individual shapes to ensure the closest fit to the final waveform.[28]

One must note that metasurfaces are fundamentally different from photonic crystals. Photonic crystals are based on Bragg diffraction or interference from lattice scattering. However, in case of metasurfaces, there is interference of individual Mie modes in the far field. As a result, there are no angular diffraction spots in the K space.

The key elements in a metasurface are its substrate, its individual resonator and its periodicity. The substrate determines its fabrication, and overall near fields. Most commonly used substrates are fused silica, Silicon on sapphire and glass.

The individual resonator, or the 'meta-atom' is the key ingredient in deciding the main interfering mode. For example, if we want the interference of electric dipole modes only, we must ensure that the resonator has a dominating electric dipole mode at the desired wavelength. These nanoresonators can take on various shapes and sizes like nanodisks, nanoholes, nanosquares or even nano ellipses. Each of these structures has its own modes and symmetry properties hence individual resonance wavelengths and phase modulation capabilities.

Periodic arrangements of the resonators guides the collective response of the electric fields from the individual building blocks. This functionality can be compared mathematically to the limit of a Reimann summation [29], where the number of terms usually tends to infinity and yet the sum is a finite value. The periodic arrangement defines the sharpness of resonance, overall phase shifts in the output and amplitude control in the metasurface output. Different possible arrangements are square lattices, hexagonal lattice, quasi crystals, amorphous arrangements and disordered arrays. Square and hexagonal lattices are generally used for sharp resonance applications where the priority is to ensure strong constructive interference. Quasicrystal arrangements are extremely useful in enabling multiple functionalities and holographic effects and amorphous or disordered arrangements can be employed for studying random and rogue effects.

Material selection in a metasurface is important as the material index influences the resonance wavelengths and the absorption decreases the quality of resonances. Generally non-absorptive materials are used in the fabrication of the metasurfaces to ensure best quality of resonances [30]. For example, in the infrared regime, Silicon metasurfaces are popular and in the visible regime a popular choice is Titanium Dioxide. These materials have their own standardised fabrication protocols and hence are not that difficult to fabricate.

0.3 Fundamentals of Metasurfaces

The fundamental working principle of metasurfaces is interference of scattering modes of subwavelength resonators. Different scattering modes in themselves form a complete set of functions capable of mapping the entire 3D vector space. This implies that through the aforementioned means we can at least mathematically generate any integrable wavefunction. In practice however we are limited to a few primary functions which can still be used in generating a lot of interesting functions.

One of the basic concepts in metasurfaces is the understanding of scattering by an individual nanoparticle. There are two types of scatterings from individual nanoparticles: Rayleigh scattering and Mie scattering. Rayleigh scattering is from nanoparticles much smaller than the wavelength of incident light. This scattering is generally weak and inversely proportional to the fourth power of incident wavelength. The dominating scattering mechanism in sub-wavelength resonators is the Mie scattering where the particle is comparable to the wavelength of light. [31] The Mie scattering is characterised by strong angular dependence, polarization dependence and sharp

resonances. The angular distribution is dependent on the size, shape, and refractive index of the nanoparticles. The scattering is also dependant on the directionality, Optical angular momentum and polarization of light with respect to the shape of nanoparticle. The Mie modes are generally sharp resonances in the scattering cross-section plots.

The Mie modes are characterised by electromagnetic multipoles like Electric dipole, Magnetic dipole, Electric quadrupole, Magnetic quadrupole and so on. Most of the nanoparticles are generally limited to these four modes only, as the higher order modes generally require complex excitation beams and shapes, however certain special BIC structures are also capable of hosting octupolar modes like [32]. Each of these modes has its own quality factor and near field profile. The scattering from any individual nanoresonator can be analysed as a combination of these modes. Specialized nanoresonators are designed in a way that one mode dominates or overlaps with another. These modes are however strongly dependent on the surrounding media like the substrate and the presence of lattice.

The lattice arrangement is crucial as it determines both the near field and the far field properties of the metasurface. The spacing, and overall arrangement of individual nanostructures controls the interference of the modes in the far field as well as the overlaps of the angular field distributions in the near field. It can lead to confinement of most of the energy to near fields by destructive interference in the far field or a constructive resonance like in the case of perfect reflector.

The working wavelength range of the metasurface can be engineered by the design of individual nanoresonator. By changing the refractive index of the material, one can red shift or blue shift the resonant modes or the effective wavelength range of the metasurface. Similarly, the scaling of the resonator accordingly scales the wavelengths. However the aspect ratio or the shape of the resonator can change both the quality of modes and the relative positions of individual modes in the scattering spectrum. In a nutshell, scaling or changing the refractive index mostly brings quantitative changes and changing the shape or aspect ratio brings qualitative changes to the scattering spectrum.[33]

Overall optical property is achieved by carefully designing the lattice arrangement of the nanostructures. Regular rectangular or hexagonal periodicity in lattices can result in sharp resonances due to collective constructive interference in near fields. Theoretically this interference can reach infinitely high quality factors depending on the design.

By slightly changing the geometry or index of the periodic resonator after a period or number of particles, we can impart gradual or abrupt changes to the phase map

or polarization of the incident beam. Such metasurfaces also known as gradient metasurfaces possess the capacity to engineer or interact with the wavefront of incident light beam at high precision due to the spatio-structural variation of unit cells over the surface. These metasurfaces play the role of light modulator instead of field confiner in an optical system.

The main selling point of gradient metasurfaces is their ability to transform the wavefront and precise modulation of the input signal. This can help in both qualitative and quantitative operations such as focusing, beam steering, or holography, in a planar and ultrathin form. These metasurfaces are highly configurable, implying that they can be used as a universal system for implementation of lenses, retroreflectors and holographic displays. As an example, metasurfaces can circumvent bulky lenses in cameras, reducing the size of optical systems with better and more precise performance.

The ability of modulating phase, amplitude and polarization of light can also provide new insights into complex mathematical operations like spatial differentiation, imaging and fourier transforms. These three degrees of control over the incident wavefront can also produce peculiar effects like phase singularity. Altering the size of resonators periodically leads to an altering phase accumulation over different parts of the metasurface. It can result in creation of vortex beams, beam deflectors and berry phase modulators.

In terms of functionality, metasurfaces can be tuned to have near field hot spots having extremely high electric fields. These near field hot spots can be used to demonstrate nonlinear processes like non perturbational harmonic generation [34, 35], multiphoton absorption [36] and even higher harmonics [35, 37]. Such metasurfaces also commonly referred to as nonlinear metasurfaces are a relatively new and unexplored territory in the discipline.

Unlike their traditional linear counterparts, the nonlinear metasurfaces also use material properties along with phase, amplitude and polarization control to exhibit nonlinear effects. These metasurfaces have the capability to demonstrate effects beyond the scope of regular light matter interactions like high harmonics and multiphoton absorptions. This feature allows them to work with multiple wavelengths simultaneously by exploiting effects like harmonic generations, frequency mixing, nonlinear switching, phase transitions and even ultrafast modulation.

Nonlinear metasurfaces enable the miniaturization of bulky nonlinear optical devices thereby facilitating integration into compact multifunctional systems. They demonstrate applications in lasing, sensing, telecom, nonreciprocity, solid state UV

sources and biophotonics. All these applications make use of the ability to generate extremely strong nonlinear response in very low physical volumes.

In summary of the foregoing, metasurfaces are by far the most flexible and reconfigurable platform the field of nanophotonics has to offer till date. Metasurfaces are close to lossless structures as the resonators as well as the substrate are both non absorbing dielectrics unlike metals in plasmonics. Even in terms of versatility, metasurfaces can be employed for almost all the applications nanophotonics is used in. This cannot be said of plasmonics because they are not good as sharp resonators because of losses and photonic crystals as they are not very successful in manipulating phases. Because of versatility in optical response, metasurfaces are widely employed in adaptive optics, beam steering, holography and general nano-optics.

0.4 Introduction to Active Nanophotonic Devices

The field of active nanophotonics is a unique combination of the fields of materials science and nanophotonics to manipulate light matter interaction at the nanoscale. The word active here refers to active media or media which interacts with incident light in a particular way or interact with external stimuli in any way. Different active media combined with different nanophotonic structures give a myriad of possibilities for nanophotonic device development.

Most observed light matter interactions include reflection, refraction, scattering, transmission, absorption and emission. Amongst these, of special interest in nanophotonics are scattering, transmission and absorption.

At atomic level, it is well known that incident light is absorbed by the electrons of the media it falls on. We observe scattering if the electrons oscillate in a dipolar arrangement. Implying that the absorbed energy is re radiated in different directions. In case of transmission the electrons absorb and re emit the same energy with a temporal delay (phase shift). However in case of absorption, the atoms of the material take the incident photon energy and increase the energy level of an electron to cross the forbidden band. After this crossing there are a lot of different possibilities like emission of photoluminescence, absorption of multiple photons, generation of harmonics or even non radiative energy loss.[38, 39]

Most research in this field is based on absorptive active media like III-V semiconductors, rare earth doped nanoparticles, Indium Tin Oxide, Transition metal Dichalcogenides, Perovskites and even organic molecules. III-V semiconductors are

widely used in optoelectronic applications like light emitting diodes and lasers where we need to convert the absorbed energy into light. The rare earth doped nanoparticles or core shell nanoparticles are of keen interest as they can produce upconverted light after absorbing two photons which makes them a prospective candidate for solid state UV lasing. TMDCs demonstrate unique dirac cone like band diagrams and hence can be useful for electro-optical technologies.[40]

Most of the active media (except graphene) have been used in functional devices at millimetre or microscale. The field of active nanophotonics promises to deliver functional optical devices at the nanoscale, which would be pivotal in the development of optical communications and quantum information. The biggest challenge in this field is that there is an absence of a universal idea of how to integrate active material with extremely sensitive nanostructures so that they complement each other's functionality. For example, when we use III-V semiconductors with a high Q nano resonator, we need to be careful that the near fields aren't too high to burn the semiconductor right away. To ensure harmonious coexistence, the concepts of gain/loss of active media and near field enhancement of the nanostructure must be clearly understood. The interplay of gain/loss factors of active media and the quality factor of resonance of the nanostructure can tune device performance as well as functionality which was not possible with passive nanophotonic devices.

For example, semiconductor dots are active media, whose band gap and emission properties can be controlled by adjusting the dimensions. This helps to get the emission at a desired wavelength as well as adjusts the value of maximum permissible gain until burning. The corresponding nanoresonator should have a sharp resonance and near field hotspots where we can expect to place some of the quantum dots. The Q factor shouldn't surpass the maximum permissible gain of the quantum dot to ensure robust light emission.

In active nanophotonics, metasurfaces are some of the most promising structures to integrate active media and their functionalities. Owing to their immense flexibility in design, reconfigurability and standardised fabrication protocols, addition of an active material is usually just one extra step in the procedure. As far as design is considered, well defined theoretical tools like multipolar decompositions are available to scrutinize the design of nanoresonator for selection of scattering modes.[41] The optimisation can be carried out by using commercially available full wave simulation softwares like COMSOL or CST.

Most of the active metasurfaces involve the use of a thin film of active media like quantum wells, vanadium dioxide or Indium Tin Oxide. In this case, the fabrication challenges are overcome by the means of coating the active layer with a very thin film

of Silicon dioxide (upto around 5-10 nm). In the cases where the active material is incorporated or embedded in the nanostructure, the etching process becomes slightly complicated

Switchable or actively controllable media offer another degree of freedom in nanophotonic device capability. Adding dynamic control to nanophotonic devices opens possibilities for optical isolation, optical switching as well as even optical logic at the nanoscale, which can be a great boost to the integrated photonics industry. The dynamic control can be either discrete or analogue, depending on the controllable media used as well as the functionality of device. The analogue control is achieved by using media which have a gradient response to external stimuli like electric, current or light or sound like photorefractive materials. A metasurface made on a photorefractive substrate can be converted into an index grating using external optical excitations.[42] This can give us a distinctly functioning metasurface every time there is a particular optical excitation on the substrate. The discrete control, (more often sought) is the achieved by using phase change media [43] where the state change or phase change is triggered by an external stimulus. An example are the Vanadium dioxide based switchable metasurfaces, where Vanadium dioxide changes its state from insulating to conducting at 68°C. This allows for two differently functional metasurface states.

The process of phase change is essentially the transition of a system's energy from one local equilibria to another local equilibria in the internal energy curve [44]. This transition is facilitated by the consumption of energy which helps in overcoming the activation barrier [44, 45]. For example, there is a transition between ice and water, ice as well as water are both stable phases on their own, but they are separated from each other by a barrier called the latent heat of fusion. This energy can be supplied by different methods [45, 46].

In our case, these different methods of energy supply are essentially the different mechanisms of phase change. Mostly used methods of energy transfer are electrical, optical, thermal and mechanical. The electrical energy transfer uses the means of charge motion to transfer the energy to material molecules [47]. Optical method exploits material photon absorption to transfer energy [48]. The thermal method also the oldest known method of energy transfer to entire mankind directly increases the kinetic energy of material molecules by increasing temperature [49]. And finally, the mechanical method relies on the application of strain to increase the elastic potential energy to the point that the material structure is completely deformed into a new one [50].

Phase change materials in nanophotonics are capable of exhibiting transitions be-

	Thermal	Electrical	Optical	Mechanical
Chalcogenides (GST, Sb_2S_3 , Sb_2Se_3)	Transition at (150-300 °C)	Possible	Possible	Not Available
Vanadium Dioxide (VO_2)	Transition at ~68 °C	Possible	Possible	Possible
Transition Metal Dichalcogenides (MoTe_2 , WTe_2)	Transition at (250-300 °C) or (50-70 °C)	Possible	Possible	Not Available
Perovskites (CsPbI_3 , Halide Perovskites)	Transition at (50-150 °C)	Possible	Possible	Not Available
Organic Polymers	Transition at (30-100 °C)	Not Available	Possible	Not Available

Figure 0.4.1: Tuning methods for phase change media.

tween two or more structural or electronic states. Each of which have a characteristic difference in optical properties like refractive index, absorption and reflectivity. Based on the consisting materials, phase change materials used in nanophotonics are broadly classified into five categories namely: Chalcogenide-Based Phase-Change Materials (GST, Sb_2S_3 , Sb_2Se_3), Vanadium Dioxide (VO_2), Transition Metal Dichalcogenides (TMDs) (MoTe_2 , WTe_2), Perovskite Phase-Change Materials (CsPbI_3 , Halide Perovskites) and Organic Phase-Change Materials (Poly(N-isopropylacrylamide) (PNIPAM), Poly(3,4-ethylenedioxythiophene) (PEDOT)) [51, 52, 53, 54]. The table above (Table 0.4.1) gives an overview of the energy transfer methods possible for the aforementioned classes of phase change materials.

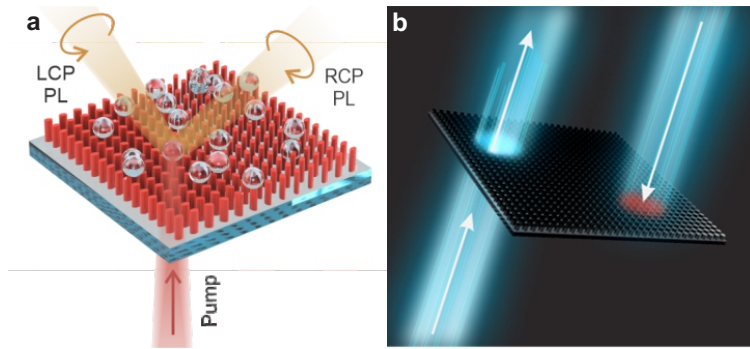


Figure 0.4.2: Types of Active Nanophotonic devices. Metasurfaces for (a) producing polarized upconversion photoluminescence (light generation) and (b) optical isolation (light control).

Overall, active nanophotonic devices can be classified into two types, control devices and light generating devices (Figure 0.4.2). From the viewpoint of optical data processing, the light generating devices such as nanolasers provide the optical analogue of current in an electronic logical operation. The laser light once generated at the

nanoscale can be guided using waveguiding channels (interconnects) all around the circuit board. The control devices (which can be controlled using external stimuli) provide the optical analogue of switches or logic gates in the same operation. Together the two capabilities if developed can be used to develop solutions for on chip all optical communication and data processing.[55]

Other applications of the field span across various fields like telecommunications, photovoltaics, sensing, imaging, catalysis and biomolecules. Switching applications of active nanophotonics find their use in the field of telecommunications for enabling ultra-fast switching, larger bandwidths and high speed data transmission. In medical science, plasmonic biosensors have proven to be extremely useful in detecting and identifying DNA molecules. Plasmonic nanotweezers are being used to perform drug delivery to locations where syringes cannot reach. Biomarkers have also proven to be an effective imaging tool from the perspective of cellular imaging. In the field of photovoltaics, active nanophotonic solutions have shown to add various new functionalities like hydrogen splitting and enhanced efficiencies of individual components by significant factors.

The major roadblocks in the field of active nanophotonics are mainly fabrication costs, fabrication challenges, and integration with other nanophotonic devices. The fabrication costs of a single active metasurface are not economical for consideration for production. Different active media have different chemistry, making it difficult to find a standardized procedure for fabrication. Eventually, achieving a common platform on which all the designed devices could work together harmoniously could be the last nail in the coffin toward bringing active nanophotonics to the market.

0.5 Nonreciprocal nanophotonics

The effect of nonreciprocity in optical systems is a consequence of breaking of time reversal symmetry. The effective permittivity tensor of such an optical system is skew symmetric, instead of a perfectly diagonal matrix. The skew symmetricity is introduced by the addition of magnetic fields [56] (magneto optical effects like Faraday rotation), acousto-optic effects (mechanical manipulation of optical properties) and nonlinear optical effects.

The Faraday effect (also used in nanophotonic devices), is basically the rotation of optical polarization by application of high intensity magnetic fields [57]. Which implies different properties for forward and backward propagation of light. As an example in case of optical isolators, due to Faraday rotation there are different accumulated polarizations for both directions of propagation. There are polarizers placed

on both the ends of the Faraday rotator, which ensure that only one polarization is allowed through implying only one passing direction.

The acousto optic effect of non-reciprocal Brillouin scattering is basically using a control acoustic signal in a waveguide with whispering gallery modes. The control signal ensures that the resonator scatters light coming from one direction and absorbs light from another [58]. This effect leads to optical isolation. In principle, non-reciprocal Brillouin scattering is a technique that can be used to integrate devices in optical isolators to integrated photonic framework. However the size of the devices is limited to a few micrometers, thereby making it suitable for microwave optics.

Optical nonlinearity can also add skew symmetric elements to the effective permittivity tensor. Effects like Second harmonic Generation, Kerr Effect and two photon absorption have been known to break the time reversal symmetry [59]. These effects are an extension of the perturbation theory of quantum mechanics where there is an additional momentum or intensity dependant potential. The most common examples of nonlinearity usage in nonreciprocity are kerr effect, nonlinear absorption and phase transition. Phase transitions are generally nonlinear processes which are marked by sudden change in structure upon reaching certain optimal conditions. For example at 100 degrees water is heated further to release steam. Hence making the curve like a step function (nonlinear).

Simulated gauge fields or abelian fields in optics are created by carefully engineering geometrical phase in the photonic crystal or metamaterials.[60] These fields can emulate the effects of magnetic fields or nonlinear fields.

An important aspect in the study of nonreciprocity is the clear understanding of the fact: what is nonreciprocal and what is not nonreciprocal. As a matter of fact, there is a huge confusion between asymmetric propagation of light and non-reciprocal propagation of light. Asymmetric propagation can be almost anything which produces different outputs after propagation through the structure (thereby resulting in asymmetry along the axis of propagation).[61] For example, there are structures where you send a signal from one direction, you get one spatial distribution of third harmonics and if you illuminate from the other side, you get another distribution. However, non-reciprocity is a different phenomenon. If we send an input signal (A) from one direction (let's call it direction 1) and get an output signal (B) (let's call it direction 2) on the other, we can call the system non-reciprocal only if we send signal (B) as input in direction 2 and not get signal A as output in direction 1 (see Figure 0.5.1).[62] This is fundamentally different from sending the same signal A in both directions and getting different outputs.[63] This condition of nonreciprocity can be mathematically verified using the Lorentz nonreciprocity theorem.

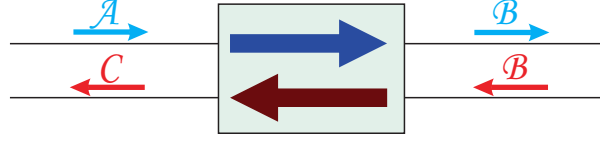


Figure 0.5.1: Optical Nonreciprocity. Sending wave A to get wave B and sending back wave B from opposite direction to get another wave C (different from A).

The design of a nonreciprocal system at the nanoscale, just like the design of active nanophotonic devices requires the assessment of three factors. The first is the working principle, where the mechanism for nonreciprocity and the function of the device are decided upon. The main mechanisms are magneto-optical effects, acousto-optical effects and nonlinear optical effects. Of these, the nonlinear optical effects are the most promising for use at the nanoscale. Nonlinear materials like lithium niobate, lithium tantalate, III-V semiconductor nanoparticles and poled ferroelectric crystals have high nonlinear coefficients and hence are known to show nonlinear effects at much lower incident intensities.

A new variety of nonlinear materials is the epsilon near zero materials like Indium Tin Oxide films. Epsilon near zero means the materials whose permittivity reaches zero at certain wavelengths. In the range of these wavelengths, these materials can exhibit exceptionally high nonlinear enhancements, both in absorption and Kerr effect. Phase change materials are also relatively new nonlinear materials, however the nonlinearity here isn't the conventional optical nonlinearity but the fact that their phase transition is a nonlinear process.

The device materials are decided to ensure that they demonstrate the properties desired for the device mechanism to function smoothly. This is followed by the device structure, which is basically an optimisation of different parameters to deliver the desired result. Another feature in design of devices like the optical isolator or optical diode is the fact that one must be very careful of the platform compatibility considering that it must be able to work properly with other devices as well. This feature is particularly useful for integrated photonics perspective.

In electronics, nonreciprocal devices like diodes/ half or full wave rectifiers, and logic gates have proven to be vital components for data processing operations. Especially the integrated circuits, field programmable gate arrays and hardware memory are completely dependant on them. In nano photonics, especially the field of optical data processing and communications, there are very few nonreciprocal devices available. Hence the field is mostly limited to linear and reciprocal applications. At nanophotonic level, nonreciprocal devices can provide rectification, improvement of signal to noise ratios, and introduce wavelength selectivity in optical signals.

In quantum computing and communications, nanophotonics plays a crucial role in transport and detection. Availability of nonreciprocal nanophotonic devices adds to the capabilities of quantum technologies by introducing logical operations, memory and preserving signal quality.

0.6 Applications

Active and nonreciprocal nanophotonics have diverse applications across most aspects of modern science and engineering technology. Ranging from core cellular biology to biotechnology in biological sciences, from inorganic chemistry to chemical engineering and from encryption to energy conversion.[64, 65, 66, 67]

There are five major sub disciplines of biological sciences where active nanophotonics finds its application. These include Biosensing, Imaging and microscopy, photothermal therapy, optogenetics and drug delivery. In Biosensing, plasmonic biosensors[68] are some of the most sensitive apparatuses available today, capable of sensing even the lowest concentrations of DNA and enzymes. The sensitivity is increased upon the use of active media in the device which enhances the light matter interaction and, in some cases, reduces the loss. Nonreciprocal optical isolators then take the available signal and improve the Signal to noise ratio by reducing back scattering. In Imaging or microscopy, superlenses or metalenses have given unprecedented focal lengths, depth of field and reduced aberrations. Active superlenses can dynamically adjust themselves to facilitate focusing, getting into deeper tissues and improve the overall image quality. Nonreciprocal photonic circuitry then improves the SNR by eliminating the back reflections from the tissues [69].

Phototherapy, also considered to be a safer alternative to chemotherapy or radiotherapy for cancer involves burning of cancerous cells using targeted light. Modern phototherapy techniques involve the use of optical nanotweezers with embedded silver or gold nanoparticles directed magnetically towards tumorous sites. Then they are excited externally using infrared radiation, and the plasmonic nanoparticles develop a lot of heat which burns the surrounding cells. Nonreciprocal light delivery is also essential in this operation as it improves the directionality of phototherapy [70, 71]. Optogenetics uses active waveguides to[72] modulate amplitude and polarization of incident light for control over gene expression.

In chemical sciences, physical chemistry and photochemistry are two types of application of active nanophotonic devices [73, 67]. Spectroscopy is an important tool in physical chemistry, as it helps in identification of different constituent atoms and energy levels of a material. In spectroscopy techniques work is done on both the sig-

nal detection aspect as well as the signal strengthening aspect. Active nanophotonic resonators are used to capture slight number of signals[74] and then resonate and amplify them to give a more measurable response. Nonlinear nanophotonic devices can amplify signals from nonlinear material transitions like Stokes or Anti Stokes Raman levels. The detection of signals is enhanced by nonreciprocal photonic circuitry, which over a few circulations in the circuit loop can increase the signal to noise ratio. Molecular imaging also utilises nanoantennas or plasmonic probes to enhance the received signals.[68]

In photochemistry, the catalysis is dependent on the optical conditions in the reaction zone. In the reaction zone, confinement of light using active nanophotonic cavities or waveguides manipulates light matter interactions of the involved materials. This can control reaction kinetics, reaction pathways and alter the activation energy.[75] In some cases, it can also influence product selectivity. Use of light sensitive active materials like photochromic molecules, azobenzene polymers, or up-converting nanoparticles in nanophotonic devices can initiate and modulate certain chemical reactions by acting as external light stimulus.

The field of energy conversion encompasses areas like energy harvesting, renewable energy generation and manipulation of light using renewable energy. Energy studies revolve around two major directions, increasing conversion efficiency[76] and discovery of new technologies.

Active and nonreciprocal nanophotonics has major contributions in improving the energy conversion efficiency across almost all the photonic platforms. In the area of Solar energy harvesting, active metasurfaces enhance the photon capturing efficiency by improving light trapping in their near field hot spots.[77, 78] Nonreciprocal optical concentrators can effectively recycle light by directing it on the semiconductor panel multiple times. This can reduce optical losses and improve overall conversion efficiency [79].

Photothermal energy conversion can also be enhanced using active plasmonic devices.[80] The ohmic loss of plasmonic metals can rapidly convert light to heat for usable purposes. The light absorption can be further enhanced by using nonreciprocal nonlinear light absorbers. Which as the name suggests recycles light by directing it to the absorbing material again and again, thereby increasing overall conversion to usable heat.

In integrated photonics, ensuring the flow of light through the interconnects[81] and maintaining its coherence is of utmost importance. Nanolasers offer a coherent source of coherent radiation at the nanoscale in the telecom wavelengths.[82, 83] Topological waveguides also come into play as potential interconnects.

An important aspect in integrated photonics is computation. Performing computation involves the use of logic as well as memory. Without these two, photonics is merely a communication channel, not a fully-fledged intelligent unit. The problem of optical logic is solved by logic gates based on active nano photonics.[84]

By the combination of modulators, nonreciprocal circuitry and metasurfaces, it is possible to mimic the function of neurons and eventually build a network of these photonic neurons. Such a network can realise learning and intelligence. However, active photonic neural networks still face integration challenges with other devices.[85, 86]

At last, but not the least, Quantum technologies are also a major application area for this field as photonics is by far the most stable and reliable platform for quantum communication.[87] Active nanophotonic devices function as repeaters and amplifiers for weak quantum signals. Nonreciprocal optical waveguides also function as quantum memories to ensure reliable data processing. Nanophotonic logic gates and nonreciprocal quantum circuitry can also be useful in combination with a reliable source of entangled photons.

0.7 Challenges

The progress in active and nonreciprocal nanophotonics is stalled due to two major types of challenges: development stage and product stage challenges. In the development stage, device fabrication and device integration pose major questions as most of the active materials don't have a well-developed recipe. Also, there is lack of a common platform where all the active devices can be built on, in other words there is lack of a breadboard where all the components could work together. In development of the product, there are performance limitations and manufacturing costs which need to be thoroughly researched.

In case of tunable metasurfaces, achieving dynamic control remains a challenge. At the nanoscale, controlling the action of external signals remains difficult, as it requires the addition of extra components to the device. Also, increasing the switching speeds is often met by material limitations like decay times and interconnect losses. The switching speed is also limited if the impulses provided are electrical. Because the speed limit to electrical signals is usually 1 nanosecond, If such a signal is being used as the control, it is impossible to get faster switching.

As an example, devices based on the pockels effect are limited in their switching speeds because of the slow nature of the effect and limited tuning ranges. This

limitation is also faced by phase change devices. In their case, repeatability and reversibility of their phase transitions is also a major challenge.

Most active materials don't exhibit broadband optical responses like Indium Tin Oxide films and semiconductor quantum dots. Their active band is small and strongly dependent on their geometry and material parameters. Increasing tuning range requires development of new hybrid materials or combination of more than one material.

The challenge of introducing external stimulus has an inverse relationship with the size of the device or simply to say the smaller our device, harder it is to ensure a controllable stimulus. Incorporating an electrical interconnect only complicates the overall design of an active nanophotonic device. It can even damage or destroy a device with the slightest of fabrication imperfections.

Optical switching also needs to ensure that the control wavelength range is beyond the device working wavelength range, otherwise we can observe significant interference with the device performance.

Materials like metal nanoparticles, Indium Tin Oxide and Vanadium Dioxide are active materials, however they also lose a significant amount of optical power on having light sent through them. For example, ITO and metal nanoparticles have ohmic losses and they lose upto half of their energy due to it.

Media like III-V semiconductors and upconversion nanoparticles are particularly challenged by saturation and burning issues. Which means that if the design is not cautious, excessive enhancement of near fields can lead to oversaturation of the gain medium and hence destruction of the device. Also, in certain cases, if the gain medium is too far away from the near field hot spots, we can observe underperformance issues.

High near fields do present themselves with an opening in the field of non-perturbational nonlinearity or simply higher order nonlinear effects which don't scale as powers of intensity [88]. This also poses the problem of generation of unnecessary stray signals like higher harmonics. Also, there are higher order nonlinear absorptions which are triggered at high field levels. They sometimes lead to sudden and abrupt losses.

In general, overcoming these challenges requires careful design consideration, new fabrication techniques and newer materials. In this research we try to solve these problems and develop functional active nanodevices.

0.8 Summary of research

In this research, we develop from scratch some active and nonreciprocal nanophotonic devices. We discuss the process from design to fabrication and measurement. We add complexity to functionality throughout this research to develop new devices. In other words, we start from the simplest of functionality and go on to complicated functionalities like optical isolation as described in Figure 0.8.1.

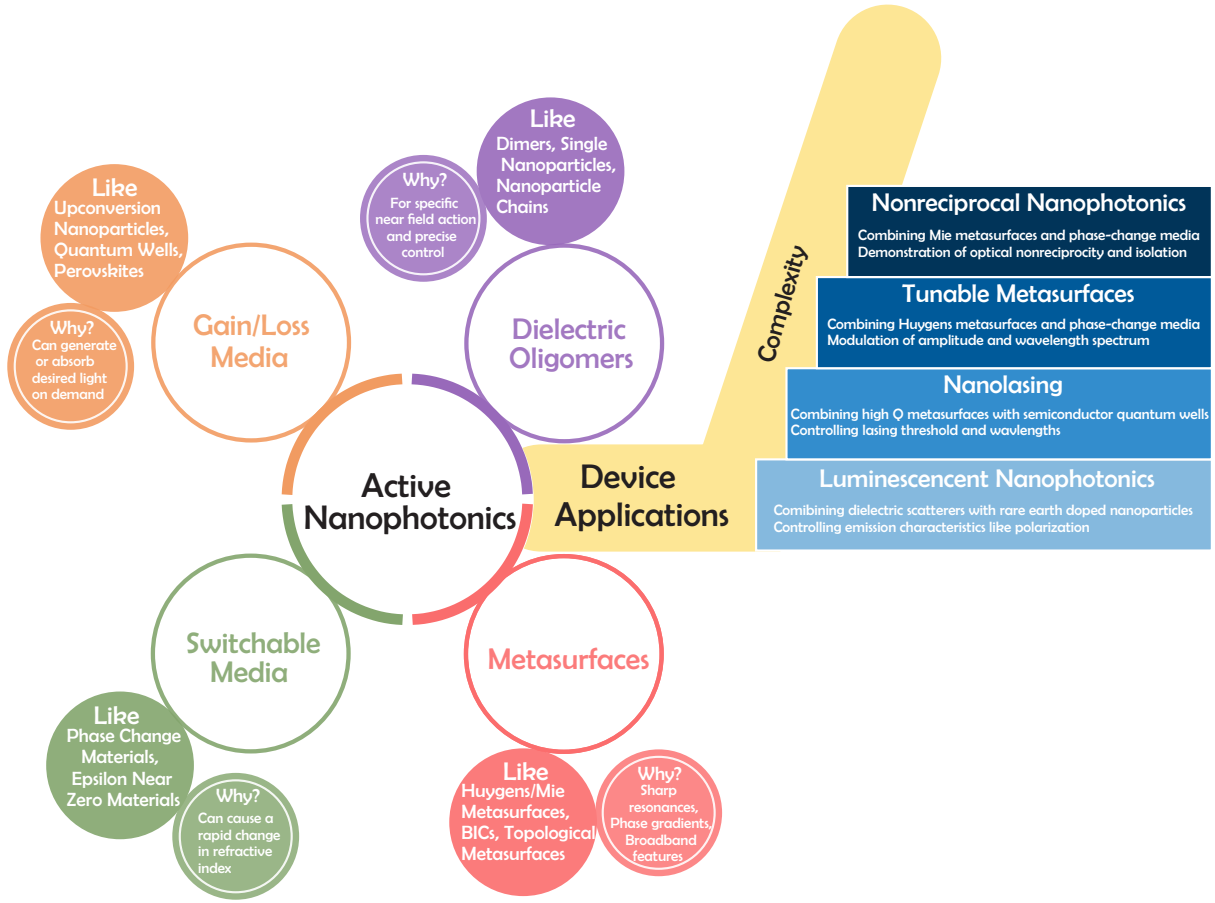


Figure 0.8.1: Conceptual map of research. Active nanophotonics is a blend of nanophotonic structures like oligomers and metasurfaces with optically active or switchable media whose consequence is development of devices. The complexity of application increases from simple light sources to nonreciprocal devices.

We browse across various active materials like III-V semiconductors, Upconversion Nanoparticles, PEDOT PSS, Quantum Dots, Phase change materials, Epsilon Near Zero materials and Perovskites. III-V semiconductors are ideal optical gain materials also used in a lot of commercially available optoelectronic devices like laser diodes and Light emitting diodes. We demonstrate the use of III-V semiconductors in quantum wells as optical gain medium in nanophotonic designs. The up conversion nanoparticles are core shell-based hybrid nanoparticles made from rare

earth metals. The upconversion wavelength depends on the doping and the type of the rare earth metal used. They are particularly easy to fabricate as the process involves mostly chemical synthesis only, after fabrication they can be drop casted or spin coated for optimal performance. We exploit their favourability in fabrication to demonstrate enhancement in upconversion photoluminescence. Phase change materials like vanadium dioxide are particularly special because of their ability to switch between two states, thereby making their use as switches a natural consequence. VO_2 is particularly special because of its close to room temperature phase transition and its retention of crystal structure in both states as well as during the phase transition. This makes the switching process repeatable over millions of times without causing lattice defects. This phase transition of VO_2 owing to its crystalline to crystalline nature and sharp nonlinear nature is the underpinning concept behind the working of our switchable and nonreciprocal metasurfaces.

Epsilon near zero materials like Indium Tin Oxide are highly nonlinear materials which can demonstrate a virtual phase change if the high nonlinearity in the wavelength range is triggered. This high nonlinearity makes the material ideal for demonstration of ultrafast nonreciprocal nanophotonics.

The nanophotonic structures in this research were chosen based on their compatibility with the chosen active media and their ability to bring out the desired device performance. To obtain sharp resonances, we use three structures, the Bound states in the continuum, topological corner states and the Anapoles. We use the topological corner states as optical confinement geometry to combine with semiconductor quantum wells to create topological nano lasers. Topological edge states are robust and resistant to disorders or fabrication imperfections. We also use 1D topological edge states to demonstrate enhancement in luminescence due to effective light trapping in rare earth doped nanoparticles.

The bound states in continuum are trapped states in space that don't decay into the continuum despite being embedded in the same. The decay is cancelled out due to strong destructive interference in the far field. The anapoles are similar conceptually as they involve the cancellation of Electric Dipole and Toroidal modes in the far field. We also combine Anapole geometry with quantum wells to create another nano laser. We use these concepts and implement them using a membrane-based structure so that at least some portions of the quantum wells coincide with the near field.

Vanadium Dioxide and Indium Tin Oxide are typically lossy materials. The high absorption leads to accumulation of greater heat in the material. So such materials cannot be used in tandem with structures that generate high near fields, as

there will be a surge in absorption and heat generated which can lead to burning and the eventual destruction of the device. We integrate them with Mie resonant metasurfaces with particularly broad resonances. Vanadium dioxide when combined with Huygens metasurfaces gives us switchable Huygens metasurfaces. Mie resonant metasurfaces with asymmetric absorption when integrated with vanadium dioxide produce nonreciprocal metasurfaces. The asymmetric absorption leads to different trigger points for vanadium dioxide phase transition. This allows for a range of optical powers between the different trigger points where we observe bistability, as well as nonreciprocity. Similarly, when Indium Tin Oxide is integrated with a metasurface with asymmetric near fields, we observe nonreciprocity because, the nonlinearity of ITO doesn't manifest itself at the same intensity for both illuminations.

The main purpose of this research is to overcome the challenges in the design and fabrication process of active and nonreciprocal nanodevices and demonstrate their potential as candidates for integrated photonics. We successfully demonstrate six working active nanophotonic devices as the proofs of our concepts. We develop designs which have a lot of further scope for improvement and optimisation, hence opening new pathways for development of commercial products.

0.9 Thesis Outline

In this work, we develop and demonstrate the working of seven active and nonreciprocal nanophotonic devices. The complexity of their function increases gradually throughout the progress of this research. We structure the entirety of this text based on functionality of nanodevices.

We begin by describing the methods used in this research. In this section we describe how we used simulation softwares to design nanophotonic structures and then performed multipolar decomposition to get accurate analysis of modal fields. We describe experimental techniques like microscopy, spectroscopy and stokes polarimetry and their use in measurement and optical characterization of the devices.

One of the most basic yet crucial functions: light emission. In general, light emitters are either basic illuminators like light emitting diodes or coherent and directional sources like laser diodes. In the first chapter, we demonstrate enhanced light emission from rare earth doped nanoparticles with a degree of polarization control. We show that by combining metasurfaces with light emitting particles we can make better light sources at the nanoscale, with control over even the most dynamic parameters like polarization.

In the second chapter, we describe our efforts in the development of nanolasers.

Nanolasers are a more complex version of nanoscale light emitters. The nanoscale light emitters offer enhanced and polarized photoluminescence, whereas nanolasers offer directional, coherent, and polarized stimulated emission. The nanolasers are an epitome of harmonious combination of semiconductor quantum wells with nanophotonic structures like anapoles and topological corner states via a membranous geometry. We successfully demonstrate nano scale lasing action at room temperatures and overcome the challenge of material losses.

One of the most challenging applications of active nanophotonics is the development of switchable and nonreciprocal nanoscale devices. A device that can switch its behaviour back and forth on the application of a stimulus is like a control. The third chapter talks about the use of phase change in vanadium dioxide to implement the switching action in a Huygens metasurface to show dual functionality. We overcome the challenges of adding external stimulus at the nanoscale by providing thermal energy to the entire substrate. This increased the temperature and facilitated the phase transition to enable dual functionality.

Then we describe the physics of nonreciprocity and the working principles of our nonreciprocal nano optical devices. We use the nonlinear phase transition of VO_2 to enforce nonreciprocal action. The nonlinear phase transition is triggered at different incident intensities for both the illuminations because of difference in absorption. A similar phenomenon occurs for ITO based metasurfaces, where the giant nonlinearity of ITO generates a change in the refractive index. This pseudo phase change also demonstrates the effect of nonreciprocity. We argue that at the nanoscale, the best pathway to achieve nonreciprocity is self-action due to nonlinearity as other pathways have fabrication difficulties on that scale.

The following is chapter wise list of research outputs.

(The [blue color](#) represents titles hyperlinked to the full texts.)

Luminescence

- [Topological nanophotonics for photoluminescence control](#) (A Tripathi, S Kruk, Y Shang, J Zhou, I Kravchenko, D Jin, Y Kivshar, Nanophotonics, 2020)
- [Metasurface-Controlled Photonic Rashba Effect for Upconversion Photoluminescence](#) (A Tripathi, A Zalogina, J Liao, M Wurdack, E Estrecho, J Zhou, D Jin, Nano Letters, 2023)
- Topology-controlled polarized photoluminescence from rare-earth doped nanocrystals (A Tripathi, S Kruk, Y Shang, J Zhou, I Kravchenko, D Jin, Y Kivshar, CLEO, 2021)
- Photonic Rashba Effect in Upconversion Photoluminescence (A Tripathi, A Zalogina, J Liao, M Wurdack, E Estrecho, J Zhou, D Jin, ICEE IEEE, 2022)

Lasing

- [Room-temperature lasing from nanophotonic topological cavities](#) (D Smirnova, **A Tripathi**, S Kruk, MS Hwang, HR Kim, HG Park, Y Kivshar, Light: Science and Applications 2020)
- [Lasing Action from Anapole Metasurfaces](#) (**A Tripathi**, HR Kim, P Tonkaev, SJ Lee, SV Makarov, SS Kruk, MV Rybin, Y Kivshar, Nano Letters, 2021)
- Room-temperature lasing from topological cavities (**A Tripathi**, D Smirnova, S Kruk, MS Hwang, HR Kim, HG Park, Y Kivshar, CLEO 2020)
- Low-Threshold Lasing from Anapole Metasurfaces (**A Tripathi**, HR Kim, P Tonkaev, SJ Lee, SV Makarov, SS Kruk, MV Rybin, Y Kivshar, CLEO 2021)
- Nano Lasing from Anapole Metasurfaces (**A Tripathi**, HR Kim, P Tonkaev, SJ Lee, SV Makarov, SS Kruk, MV Rybin, Y Kivshar, Photonics North 2021)
- Active and lasing dielectric metasurfaces (**A Tripathi**, SS Kruk, Y Kivshar, ICEE IEEE 2022)

Tunable and nonreciprocal metasurfaces

- [Tunable Mie-Resonant Dielectric Metasurfaces Based on VO₂ Phase-Transition Materials](#) (**A Tripathi**, J John, S Kruk, Z Zhang, HS Nguyen, L Berguiga, PR Romeo, ACS photonics, 2021)
- [Nonreciprocal optical nonlinear metasurfaces](#) (**A Tripathi**, C F Ugwu, V S Asadchy, I Kravchenko, S Fan, Y Kivshar, J Valentine, and S S Kruk, submitted: Nature communications)
- [Nonreciprocal metasurfaces based on epsilon-near-zero material](#) (**A Tripathi**, R H Aschwanden, B Sain, P Jangid, T Zentgraf and S S Kruk)
- Switchable Dielectric Metasurfaces (**A Tripathi**, J John, S Kruk, Z Zhang, HS Nguyen, L Berguiga, PR Romeo, Photonics North, 2021)
- Switchable Dielectric Metasurfaces based on vanadium dioxide (**A Tripathi**, J John, S Kruk, Z Zhang, HS Nguyen, L Berguiga, PR Romeo, OSA Fabrication Congress, 2021)
- Active and lasing dielectric metasurfaces (**A Tripathi**, SS Kruk, Y Kivshar, ICEE IEEE 2022)
- Nonreciprocal optical nonlinear metasurfaces (**A Tripathi**, C F Ugwu, V S Asadchy, I Kravchenko, S Fan, Y Kivshar, J Valentine, and S S Kruk, Frontiers in Optics, 2022)
- Nonreciprocal transmission through metasurfaces with phase transition materials (**A Tripathi**, C F Ugwu, V S Asadchy, I Kravchenko, S Fan, Y Kivshar, J Valentine, and S S Kruk, CLEO 2023)
- Optical nonreciprocity in metasurfaces with epsilon-near zero materials (**A Tripathi**, R H Aschwanden, B Sain, P Jangid, T Zentgraf and S S Kruk, SPIE Optics and Photonics, 2023)

The aforementioned works received the following awards and recognitions.

Awards and Achievements

- Invited talk on Active Nanophotonics at TIFR Mumbai 2022.
- Best presentation award at IEEE 5NANO 2022.
- Achieved the 1st place at OSA Innovation School 2021.
- Selected amongst 100 research students around the world for Seigman International School on Lasers 2020.
- Invited talk at ICEE IEEE Bangalore 2022.
- Emerging researcher award (Optical Society of India, 2022).
- VCs Travel Grant for HDR students

Media releases

- Featured author in Media Release July 2020; “Researchers develop tiny but tough lasers”.
- Seminar: “Active nanophotonics for robust chip scale optical devices”, Department of Physics and Engineering, ITMO University.
- Seminar: “Nanoscale optical devices”, Department of Physics University of Madras 2023.

...

Research Methods

In the previous chapter we discussed about the prerequisite conceptual framework which could be useful for the comprehension of this series of research . Building upon the contextual foundation this chapter serves as a transition from conceptual understanding to practical methodology by offering a detailed overview of the strategies used in research and achievement of objectives . The main idea behind this meticulous detailing is to achieve transparency integrity in research.

This work is mainly experimental, hence most of the methods described are optical diagnostic techniques. However, we also design our samples and perform theoretical analysis in certain cases, which we elaborate in computational techniques.

The following section therefore will provide a detailed examination of the methods employed in research.

1.1 Computational Techniques

1.1.1 Multipolar decomposition

In mathematics, there is a concept called decomposition of functions into smaller basis functions. For example, using hermite-gaussian polynomials one can rewrite any function as their linear combination. Such basis sets which can help us decompose any function are known as complete sets. It is a very common and useful practice in mathematics, physics and engineering to decompose complicated functions and fields into basic polynomials or functions.

In electrodynamics, decomposition into basic functions is a very important tool to ensure correct analysis of complex fields. Complex electromagnetic fields can be decomposed into understandable basic functions like the spherical harmonics. This approach of decomposition is also known as multipolar decomposition. The name multipolar comes from the fact that the basis functions are fields from pairs of equal and opposite charges (poles), here dipole means one pair similarly quadrupole means

two pairs and so on (see Figure 1.1.1). This decomposition makes the analysis of fields more intuitive due to analogy with separation or accumulation of pairs of charges.

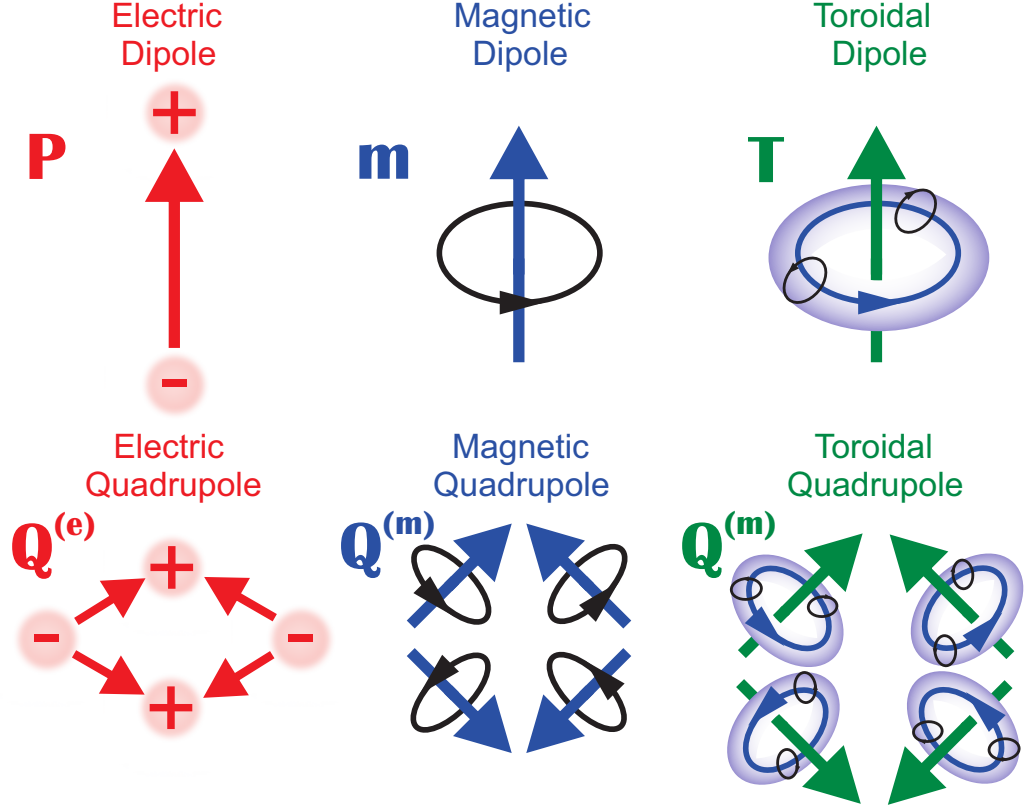


Figure 1.1.1: Electrodynamical Multipoles. The three columns contain charge-current distributions for respective multipoles (Electric, Magnetic and Toroidal).

In nanophotonics, multipolar decompositions provide a very strong insight of what is going on inside the nanoparticle upon exposure to light.[89] In electrodynamic systems, if we can estimate or even guess the currents and charges, we can get an idea of fields as well as magnetism using the four maxwells equations.

The multipolar decompositions give a clear understanding of both charges as well as currents. For example, if the decomposed fields of a nanoparticle have maximum energy in the electric dipole and magnetic dipole functions. We can safely assume that the nanoparticle must have a pair of opposite charges as well as one loop of current flowing in somewhere along the surface. Once this is understood, it becomes simple to understand what these nanoparticles might do in an array or a system.

In this research, we employ multipolar decompositions of fields to understand the physics of scattering from individual nanoparticles.[90] For example, to develop Huygens metasurfaces where we get perfect reflection or transmission, we need to ensure

destructive interference of scattered fields in both directions. First, we make sure that only two modes of radiation (only two spherical harmonics) should exist in the nanoparticle, otherwise ensuring destructive interference will be impossible. We mostly go for the basic electric dipole and magnetic dipole modes as they are the most basic and easily achievable in practice. Next, we ensure that the resonance wavelengths for both the modes coincide with each other to make sure that a clean interference effect is possible. Finally, we organize the lattice in such a way that in the far field the interference is perfectly destructive.

This approach helps in designing individual nanoparticles of the metasurface. Or rather gives a strong qualitative analysis of the near and far fields of the scatterer.[91] We also use the decomposition of fields to pinpoint the hotspots. For instance, if we want the field to be on top of the nanoparticle we might go for an electric dipole resonance and if we want the field to sit close to the substrate, we would likely go for a magnetic dipole resonance and design the nanoparticle accordingly. This concept extends to the cases of bound states in the continuum. In the BICs, we adjust the dimensions so that there is almost zero farfield decay or completely destructive interference. This implies that the entire energy is stored as the near field, hence giving us high Q factor resonances. This study is particularly useful in the design of nanolasers. A slightly advanced application is to ensure that the field hotspots interact sufficiently[92] with the used active material.

In switching or nonreciprocal devices, it is of great importance to initiate the switch or transition in the active material. The multipolar decompositions help to select nanoparticle scattering modes so that maximum electric field sits inside the active material. For example, we use strong magnetic dipole resonances to ensure that the vanadium dioxide substrate gets enough energy to facilitate its phase transition. This phenomenon becomes the underpinning concept for switchable metasurfaces. The design of nanoparticle is followed by the design of metasurface lattice which is done using full wave optical simulations.

1.1.2 Optical Simulations

As discussed earlier, the solution of maxwells equations can give accurate description of fields, charges, and currents within and outside the nanoparticle. In the farfield, the solution of maxwells equations can also be used to see the interference effects for a lattice of nanoscatterers[93] (metasurface).

In computational physics, a lot of problems are solved by discretising the system into small points or simply by making a mesh of points for calculation. Broadly speaking,

there are two popular techniques used in full wave simulations, the frequency domain and the time domain.

In the frequency domain method, plane waves of different frequencies in the computational band are sent into the sample. Their response is measured thereafter to give different spectra like transmission, reflection, and absorption.

The frequency domain works by division of calculation domain into finite spaces or cells. The size of cells is dictated by the need of spatial resolution to properly capture the optics of the system. Then the maxwells equations are discretised by the use of either the finite element analysis, finite element method or the finite difference method.[94] Then we set the boundary conditions for the interfaces which in our case are between air and nanoparticle, nanoparticle and substrate and air and substrate. After this, the equations are solved to obtain fields in every cell and for every frequency.

The time domain method involves sending a pulse on the sample which contains all the frequencies we need to measure. Then we calculate the response from the pulse and perform the fourier transform to get the result. This technique is slower than frequency domain method and is used for finer features like ultra-thin resonances which are difficult to observe in the frequency domain.

The working of time domain is particularly like the frequency domain in the sense of discretisation of computational domain and boundary conditions. However, instead of discrete frequencies the calculations performed are temporal. This means that in this method we try to calculate fields at discrete time intervals. This is performed using time stepping algorithms.

These full wave simulations are typically useful in adjusting the lattice parameters to ensure desired farfield properties are observed.[95] We use them in a typical parameter sweep for adjusting the transmission values, widths of resonances, reflection, and absorption spectra in the metasurfaces. For example, in the design of nonreciprocal metasurfaces made of vanadium dioxide we adjusted the contrast between absorption for excitation from both top and bottom directions by performing a parameter sweep to get optimal lattice period and thickness of film.

The data for optical constants for ITO and VO₂ used in my optical simulations is found in references [96] and [97].

1.1.3 Experimental device automation

We use programming to automate our optical experiments. This helps us in performing more efficiently repetitive experiments and reduce the need of human inter-

vention (hence facilitating multitasking) and reduce the possibility of human errors. Just like optical simulations, adding automation to experiments gives us the freedom to perform experimental parameter sweeps.

A good experimental automation requires developing raw codes for the functioning of all the important experimental components. This includes rotational and translational stages for the movement of samples or optical components, shutters, Laser pulses and wavelengths, cameras, power meters, photodiodes, photodetectors, and spectrometers. Before developing the code, the biggest challenge was to find a common language or platform. This was essential for the purpose of eventually combining all the components to develop a code for an experiment.

The languages most suited for input output applications is usually C/C++. However, certain devices like power meters supported Java. Hence, we chose MATLAB. MATLAB is a typically high-level scientific programming language. MATLAB has one of the best algorithms for calling libraries from most of the well-known programming languages. MATLAB also possesses a large library of mathematical and scientific functions which is extremely useful in the collection of experimental data.

We usually combined our automation with data processing codes to provide instant and in some cases even live results to check whether the experiments are going on perfectly. It was made possible due to MATLABs ability to handle large data sets, and its inbuilt visualization and data plotting tools.

Another requirement in the development of codes for device automation are the communication protocols. Once the programming platform is decided upon, it is important to have communication protocols in place for every device so that there is compatibility between computer and the device for data exchange, command transmission and reliability in execution of operations. We relied on ActiveX servers, COM objects or JavaScript wrappers as the communication protocols for our devices.

In MATLAB, ActiveX servers are a method to facilitate interaction between MATLAB and other applications or softwares with Graphical user interfaces that support this protocol. In a nutshell, ActiveX allows the user to use a GUI programmatically using MATLAB or simply it uses MATLAB to press the buttons in a GUI. We use ActiveX servers for writing codes and functions for our shutters, rotational and translational stages.

The Javascript wrappers are slightly advanced versions of ActiveX servers and Application Programming Interfaces (APIs).[98] They can encapsulate the APIs or GUIs, and hence allow for addition of extra functionalities. Basically, the wrappers

provide all the functions in the GUI as a function and hence they don't actually need to open the GUI to communicate with the device. We use wrappers for our spectrometers and power meters.

The Component Object Model Objects or simply COM objects are by far the most advanced and universal technology available for interaction with external devices and softwares. The COM objects can communicate with external softwares or device programs just like ActiveX servers and wrappers. The advantage lies in their ability to communicate directly with the hardware like LabView does.[99] Using COM objects we can call or modify Input/Output functions at the hardware level which is not possible using any of the libraries MATLAB offers. Due to their versatility, in this research we used COM objects to develop codes for cameras and lasers.

Our automated optical experiments helped us in scaling up our projects and experimental configurations. We increased our capabilities and scope of solving research challenges.

We share some of the codes used to automate experimental machinery.

```

1 RT.fName='C:\Users\sergey\Desktop\AT\automation\NonlinearSetup\
  MIOPA_labview\Users\alexander\Desktop\MIOPA\laserApp.vi';
2 RT.devicePanelHandle = figure('Visible','off');
3 RT.device_obj=actxserver('Labview.Application');
4 RT.viPath =RT.fName;
5 RT.lam=1550;
6 if ~exist(RT.viPath,'file')
7     f=0;warning('MIOPA vi not found!');return;
8 end
9
10 RT.vi = RT.device_obj.GetVIREference(RT.viPath) ; % 'Load the vi
    into memory
11 RT.vi.OpenFrontPanel(true,4);
12
13 %             RT.device_obj.StartCtrl;
14 %             invoke(RT.vi , 'SetControlValue', 'T2',100);
15
16 invoke(RT.vi, 'SetControlValue', 'N2Old',0);
17 if  invoke(RT.vi, 'GetControlValue', 'runFlag')==false
18     invoke(RT.vi, 'SetControlValue', 'Signal wavelength (nm)',1550);
19     invoke(RT.vi, 'SetControlValue', 'N2New',1550);
20     RT.vi.Run(1);
21 end
22 getLam(RT);
23 movingFlag=1;
24 pause(1);
25 while movingFlag

```



```

26     status=invoke(RT.vi,'GetControlValue','Status');
27     %           disp(status);
28     movingFlag=~strcmp(status,'Ready');
29     %           disp(movingFlag);
30     pause(0.1);
31 end
32
33 RT.connected=true;
34 disp('laser connected');
35
36 %chang wavelength to 1600
37 goLam(RT,1600)
38
39 function f=goLam(RT,lam)
40     oldLam=RT.lam;
41     if lam<oldLam
42         RT.lam=lam-10;
43         RT.goLam(lam-10);
44         RT.goLam(lam);
45     end
46     invoke(RT.vi,'SetControlValue','N2New',lam);
47     movingFlag=1;
48     while movingFlag
49         status=invoke(RT.vi,'GetControlValue','Status');
50         movingFlag=~strcmp(status,'Ready');
51         pause(0.1);
52     end
53     RT.lam=lam;
54     f=1;
55     pause(1);
56 end
57
58 function f=getLam(RT)
59     f=invoke(RT.vi,'GetControlValue','Signal wavelength (nm)');
60     RT.lam=f;
61 end

```

Listing 1.1: Working demo code for wavelength change for MIROPA optical parametric amplifier.(We thank Lei Wang for this piece)

```

1 function [power] = thorlabs_measure_power(lam)
2
3 NET.addAssembly('C:\Program Files (x86)\Microsoft.NET\Primary
    Interop Assemblies\Thorlabs.TLPM_64.Interop.dll');
4
5 import Thorlabs.TLPM_64.Interop.*;

```

```

6
7 %Create a dummy TLPM object to check for compatible devices.
8 handle = System.IntPtr(0);
9 device = TLPM(handle);
10
11 %Look for connected devices
12 try
13     [~,deviceCount] = device.findRsrc();
14 catch
15     print 'Unable to find compatible connected devices. Is the
16         device connected, on, and using the TLPM driver? This example
17         will not work with the legacy IVI drivers.';
18     return
19 end
20
21 if deviceCount == 0
22     print 'Unable to find compatible connected devices. Is the
23         device connected, on, and using the TLPM driver? This example
24         will not work with the legacy IVI drivers.';
25     return
26 end
27
28 %if only one device is connected, connect to this one.
29 deviceName=System.Text.StringBuilder(256);
30 %%if deviceCount == 1
31     device.getRsrcName(0, deviceName);
32 %if multiple are connected, ask which to use
33 % else
34 %     for i = 0:deviceCount-1
35 %         device.getRsrcName(i, deviceName);
36 %         disp(' ');
37 %         disp(['Device #', num2str(i)]);
38 %         disp(deviceName.ToString);
39 %     end
40 %     val=input('Select a device by the number from the above
41 %         detected devices: ');
42 %     if (floor(val) < deviceCount) && (floor(val) > -1)
43 %         device.getRsrcName(floor(val), deviceName);
44 %     else
45 %         print 'Invalid selection';
46 %         return;
47 %     end
48 % end
49
50 deviceName.ToString()
51
52

```

```

47 %reassign device to the selected power meter console
48 device = TLPM(deviceName.ToString(), false, false);
49
50 %set wavelength
51 wavelength= lam;
52 try
53     device.setWavelength(wavelength);
54     disp('Wavelength Setting (nm):')
55     disp(wavelength);
56 catch
57     disp('Error Setting Wavelength');
58     device.Dispose()
59 end
60
61 %measure power
62 try
63     [~, power]=device.measPower();
64     disp('Measured Power:');
65     disp(power);
66 catch
67     disp('Error Measuring Power');
68     device.Dispose()
69 end
70
71 device.Dispose()
72
73 end

```

Listing 1.2: Working code for obtaining power reading from thorlabs power meters.

```

1 function [spectrum,wavelengths]=measurespectra(t,n)
2 %% Checking if file path is added or not
3 try
4 wrapper = com.oceanoptics.omnidriver.api.wrapper.Wrapper();
5 wrapper.openAllSpectrometers();
6 catch
7     javaaddpath('C:\Program Files\Ocean Optics\OmniDriver\OOI_HOME\
OmniDriver.jar');
8     wrapper = com.oceanoptics.omnidriver.api.wrapper.Wrapper();
9     wrapper.openAllSpectrometers();
10 end
11 %% correcting electrical dark
12 wrapper.setCorrectForElectricalDark(0,1);
13 %t= Exposure time in microseconds
14 %n=1 No. of scans to average
15 %% Storing wavelengths
16 wavelengths = wrapper.getWavelengths(0);

```

```

17 %% Setting exposure time
18 wrapper.setIntegrationTime(0, t);
19 %% setting scans to average
20 wrapper.setScansToAverage(0,n);
21 %% getting spectrum
22 spectrum = wrapper.getSpectrum(0);
23 %% closing spectrometer
24 wrapper.closeAllSpectrometers();
25 end

```

Listing 1.3: Working code for obtaining spectra from Ocean Optics spectrometers.

```

1 function [picture] = clickpicture(t,n)
2 %UNTITLED2 Summary of this function goes here
3 % Detailed explanation goes here
4 vid = videoinput('gige', 1, 'Mono16');
5 src = getselectedsource(vid);
6 vid.FramesPerTrigger = n;
7 src.PacketSize = 9014;
8 src.ExposureTimeAbs = t; %in microseconds
9 preview(vid)
10 pause(2)
11 start(vid)
12 pause(2)
13 data = getdata(vid, vid.FramesAvailable);
14 data=squeeze(data);
15 stoppreview(vid)
16 delete(vid)
17 clear vid
18 clear src
19 picture=mean(data,3);
20 end

```

Listing 1.4: Working code for clicking picture in Bobcat 320 infrared camera.

```

1 %First capture camera calibration images which will be used for
  background
2 %subtraction
3 %Operate only once you know that you are on zero degrees (
  horizontal) on
4 %both QWP and Polarizer
5 clear; close all; clc;
6 global h; % make h a global variable so it can be used outside the
  main
7 % function. Useful when you do event handling and
  sequential move
8 global h1;

```

```

9         %% Create Matlab Figure Container
10 fpos      = get(0,'DefaultFigurePosition'); % figure default position
11 fpos(3) = 650; % figure window size;Width
12 fpos(4) = 450; % Height
13
14 f = figure('Position', fpos,...
15           'Menu','None',...
16           'Name','APT GUI');
17 f1 = figure('Position', fpos,...
18           'Menu','None',...
19           'Name','APT GUI');
20 %% Create ActiveX Controller
21 h = actxcontrol('MGMOTOR.MGMotorCtrl.1',[20 20 600 400 ], f);
22 h1 = actxcontrol('MGMOTOR.MGMotorCtrl.1',[20 20 600 400 ], f1);
23 %% Initialize Stages
24 % Start Control
25 h.StartCtrl;
26 h1.StartCtrl;
27 % Set the Serial Number
28 SN = 27254580; % put in the serial number of the Polarizer
29 set(h,'HWSerialNum', SN);
30 SN1 = 27254580; % put in the serial number of the QWP
31 set(h1,'HWSerialNum', SN1);
32 % Indentify the device
33 h.Identify;
34 h1.Identify;
35 pause(10);% waiting for the GUI to load up;
36 t=3; %pausing time
37 h.MoveHome(0,0); %checking home
38 h1.MoveHome(0,0);%checking home
39 %% Initializing Camera
40 vid = videoinput('gige', 1, 'Mono16'); %create video object
41 src = getselectedsource(vid); %create source object
42 vid.FramesPerTrigger = 5;    %No. of Images to be captured per
    iteration
43 src.PacketSize = 9014;        %Indicate packet size
44 src.ExposureTimeAbs = 500;    %Indicate exposure time (microseconds)
45 %t=src.MinimumFrameTime;
46 preview(vid)                 %opens live preview window
47 pause(2)
48 %% Horizontal Polarization
49 h.SetAbsMovePos(0,0), h1.SetAbsMovePos(0,0); %Set Polzarizer and
    QWP
50 h.MoveAbsolute(0,1==1), h1.MoveAbsolute(0,1==1); %Move Polzarizer
    and QWP
51 pause(t);

```

```

52 start(vid)      %Start Capture
53 pause(2)
54 H = getdata(vid, vid.FramesAvailable); %Collect data
55 H=squeeze(H);
56 %% Vertical Polarization
57 h.SetAbsMovePos(0,90), h1.SetAbsMovePos(0,0); %Set Polzarizer and
    QWP
58 h.MoveAbsolute(0,1==1), h1.MoveAbsolute(0,1==1); %Move Polzarizer
    and QWP
59 pause(t);
60 start(vid) %Start Capture
61 pause(2)
62 V = getdata(vid, vid.FramesAvailable); %Collect data
63 V=squeeze(V);
64 %% LCP Polarization
65 h.SetAbsMovePos(0,0), h1.SetAbsMovePos(0,45); %Set Polzarizer and
    QWP
66 h.MoveAbsolute(0,1==1), h1.MoveAbsolute(0,1==1); %Move Polzarizer
    and QWP
67 pause(t);
68 start(vid) %Start Capture
69 pause(2)
70 Lcp = getdata(vid, vid.FramesAvailable); %Collect data
71 Lcp=squeeze(Lcp);
72 %% RCP Polarization
73 h.SetAbsMovePos(0,0), h1.SetAbsMovePos(0,-45); %Set Polzarizer and
    QWP
74 h.MoveAbsolute(0,1==1), h1.MoveAbsolute(0,1==1); %Move Polzarizer
    and QWP
75 pause(t);
76 start(vid) %Start Capture
77 pause(2)
78 Rcp = getdata(vid, vid.FramesAvailable); %Collect data
79 Rcp=squeeze(Rcp);
80 %% D1 Polarization
81 h.SetAbsMovePos(0,-45), h1.SetAbsMovePos(0,-45); %Set Polzarizer
    and QWP
82 h.MoveAbsolute(0,1==1), h1.MoveAbsolute(0,1==1); %Move Polzarizer
    and QWP
83 pause(t);
84 start(vid) %Start Capture
85 pause(2)
86 D1 = getdata(vid, vid.FramesAvailable); %Collect data
87 D1=squeeze(D1);
88 %% D2 Polarization
89 h.SetAbsMovePos(0,45), h1.SetAbsMovePos(0,45); %Set Polzarizer and

```

```

    QWP
90 h.MoveAbsolute(0,1==1), h1.MoveAbsolute(0,1==1); %Move Polzarizer
    and QWP
91 pause(t);
92 start(vid) %Start Capture
93 pause(2)
94 D2 = getdata(vid, vid.FramesAvailable); %Collect data
95 D2=squeeze(D2);
96 %% Going back home
97 h.MoveHome(0,0);
98 h1.MoveHome(0,0);
99 %% Closing camera
100 stoppreview(vid)
101 delete(vid)
102 clear vid
103 clear src

```

Listing 1.5: Code for automated Stokes polarimetry.

1.2 Experimental Techniques

1.2.1 Near-infrared optical microscopy

Near infrared optical microscopy is the most regularly employed technique in this work. As the name suggests, this technique uses near infrared light (wavelength range 700-2500 nm) for illumination of the samples. This is particularly useful for our metasurfaces as most of the active materials we use operate in this regime. For example, the semiconductor quantum wells in our nanolasers have optical gain in this regime, Vanadium dioxide shows maximum difference in transmission for near infrared light, Indium Tin Oxide has its epsilon near zero region at the wavelength of 1243 nm and the upconversion nanoparticles absorb the wavelength of 980 nm to give upconverted signal at 800 nm.

In case of our active and nonreciprocal metasurfaces, the near infrared optical microscopy serves both the detection as well as the excitation purpose. To incorporate both these capabilities and allow for flexibility, we build our own customised setup (see Figure 1.2.1) which allows for a choice of detectors as well as excitation sources. The detection aspect is facilitated by the CCD imaging cameras for both visible as well as near infrared ranges.

The excitation is carried out by different lasers, ranging from femtosecond laser coupled to an optical parametric amplifier to get a range of wavelengths and pulses to simple laser diodes connected to electrical pulsers capable of generating nanosecond

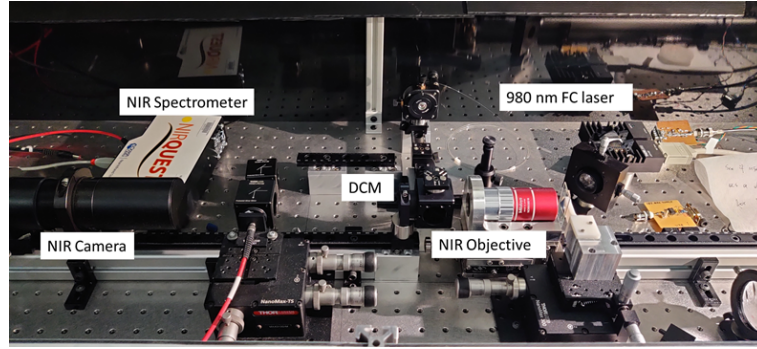


Figure 1.2.1: Experimental Setup. The picture shows the custom setup built for optical diagnostics experiments. The setup is capable of performing near infrared microscopy-/spectroscopy, nonlinear measurements and polarimetry. (FC Laser: Fiber Coupled Laser, DCM: Dichroic Mirror, NIR: Near Infrared)

pulses. Different wavelengths and pulses are chosen on the basis of the active media used in the nano device.

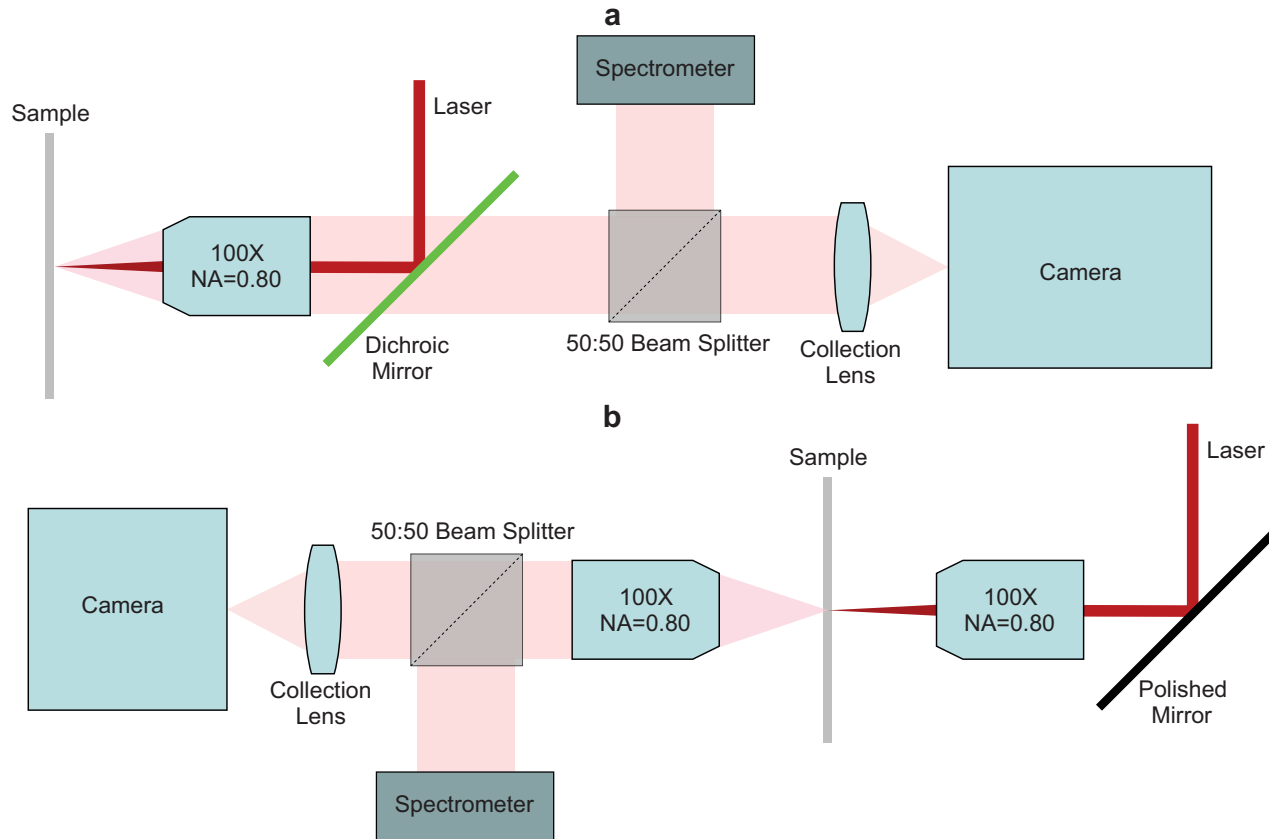


Figure 1.2.2: Near Infrared Microscopy. The (a) Reflection and (b) Transmission modes of near infrared optical microscopy used in this work. Simultaneous spectral and imaging measurements are possible due to beam splitter.

We employ two distinct types of detection techniques in our microscopy setup. The

first is reflection microscopy[100] where measurement is done in the direction opposite to that of propagation of incident light using the light reflected from the sample. The other method is the transmission microscopy [101] where the sample is imaged and measured in the direction of incident light (basically we measure transmitted light). Transmission microscopy needs two microscope objectives: one for focusing the incident beam on the sample and the other for collimating the collected light behind the sample. However, in reflection microscopy both the purposes are served by the same objective lens (see Figure 1.2.2). We use transmission mode for our work on nonreciprocity and rashba effect and reflection mode for our lasing projects.

Our microscopy setup can be extended to include other optical measurements like polarimetry, spectroscopy and interferometry.

1.2.2 Nonlinear optical microscopy

Nonlinear processes like two photon absorption and second harmonic generation are double frequency processes. Double frequency implies half the wavelength, and hence we get double the resolving power. The two-photon photoluminescence as well as second harmonic generation provide fluorescence signals which can be isolated using low pass filters, hence we can see sub-diffraction resolution imaging.[102, 103, 104] optical microscopy is an extension of the previous technique.

Nonlinear optical microscopy is an extension of the linear microscopy technique previously described. The only major difference lies in the part of excitation as it uses pulsed laser (Figure 1.2.3) excitation to trigger the nonlinear processes without damaging the material.

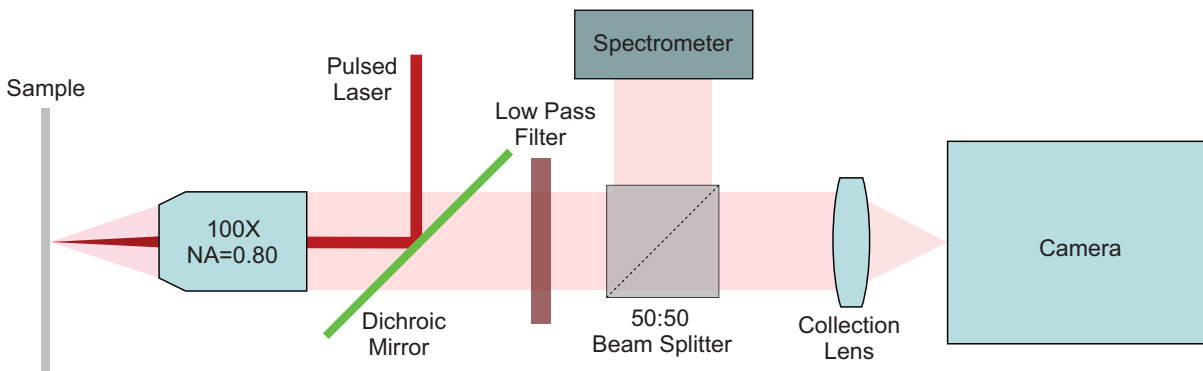


Figure 1.2.3: Nonlinear Optical Microscopy. Like NIR microscopy, a high power pulsed laser illuminates the sample and nonlinear signal is filtered out using a low pass filter.

In active nanophotonics, this technique is of great importance as most of the active

materials due to their strong light matter interaction also possess high nonlinear coefficient. For example AlGaAs nanoparticles are capable of demonstrating even the emission of higher harmonics upto 7th and 9th orders.[105]

In our research, we don't fully rely on this technique for imaging purposes, but for demonstrating new physics. Effects like non perturbative generation of high harmonics, three wave mixing and multiphoton photoluminescence[105] have been demonstrated by our optical setup for resonant dielectric nanostructures.

1.2.3 Laser diode pulser

As discussed in previous section, we use different diodes as illumination sources in our setup depending on the type of active material used.

However, to ensure flexibility in experiments, we must allow for the use of both Continuous wave as well as pulsed mode of operations for our sources. Most of the laser diodes used are CW light sources, without any mechanism to control their emission. The standard three pin or the fourteen pin butterfly packages of laser diodes are the most commonly available systems.

To fulfil the requirement of a customised pulsed source, we had to pulse the laser diodes externally. Q switching and mode locking as pulsing mechanisms were certainly out of question as they required a lot of optical equipment. Also, for a diode of a few millimetres dimension, one cannot use centimetre scale optics and design switches.

This leaves the option of electrical pulsing to get pulsed mode operation. Electrical pulsing happens to also be the most convenient option due to the presence of laser diode cathode and anode. Sending electrical pulses to any system has three requirements namely, transmission line, voltage to current converter and a function generator.

The function generator serves the purpose of generating pulses. We use an 80 GHz clock speed function generator to achieve voltage pulses as short as 8 nanoseconds with a repetition rate of mHz or more.

An integrated circuit that can convert voltage signals into current is needed to convert voltage pulses into current pulses for our laser diodes. We use IC TC4421AV for this purpose. We provide an external voltage bias using a voltage source. This bias defines the amplitude of the current pulses generated from voltage pulses (Figure 1.2.4). We solder it onto a breadboard for mechanical stability.

The transmission lines are particularly important in waveform or pulsed applications because of their characteristic of impedance matching. Impedance matching means

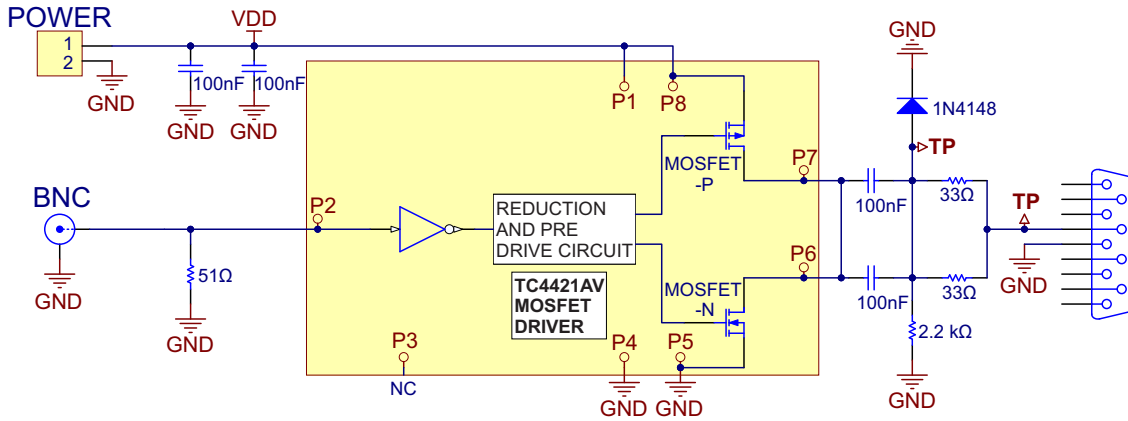


Figure 1.2.4: Laser Diode Pulser. The circuit diagram shows the electronic component arrangement for our laser diode pulser. The yellow box represents the internal circuitry of TC4421AV. The pulsed voltage input from function generator is provided at the BNC port and the pulsed current output at DB9 cable.

matching the ratio of voltage to current in the source as well as the load and the line. It is the only method to avoid pulse back reflections and eventual loss of repetition rate or even dispersion. We use external resistors and capacitors at the beginning of the transmission line to ensure impedance matching. Now we send the current pulses using our transmission line into the laser diode.

Making our own pulser gave us a choice of pulses, repetition rate and width. The pulse width could vary from as small as 8 nanoseconds to milliseconds. This was typically essential for pumping our nanolasers and high harmonic nanostructures.

Also, the control over current amplitude meant control over laser diode pulse amplitude. This reduced our dependence on external power controls like a wire grid polarizer for the purpose of reducing the intensity.

Pulsing also added to the safety of laser diodes by preventing them from burning due to heat generation from continuous supply of current.

1.2.4 NIR/UV Vis Spectroscopy

Just like how multipoles give an idea of light matter interaction in the nanoparticle theoretically, spectroscopy is the gold standard for all experimental measurements in the field of light matter interactions. Spectroscopy involves excitation of a sample using electromagnetic radiation of one or more frequencies and then observing the signal emitted. This emitted signal can be scattering, absorption, fluorescence or even transmission or reflection.

Research in nanophotonics generally comprises of fluorescence, absorption and

Transmission/reflection spectroscopy in the ultraviolet-visible range or the near infrared range. The UV/Vis methods are generally used to measure harmonics and upconversion signals as most of active nanophotonic devices work in the near infrared range.[106] However, the UV/Vis finds its use in the study of perovskite based metasurfaces as well as for quantum dots.

NIR spectroscopy on the other hand is more commonly used because of two reasons: most of the excitation lasers and pulsed lasers are designed to work in the near infrared region and most of the active materials are sensitive to wavelengths in the range 900-2000 nm. We also prefer designing near infrared devices because of ease in fabrication and flexibility in dimensions of the nanoparticles.

The spectroscopy methods we employ depend on the type of signal we expect from the nanostructure. For example, in case of lasing structures we study the luminescence spectrum. The luminescence spectrum is the spectrum obtained in the spectrometer after exciting the sample with a pump beam. As the pumping intensity is increased, we observe a gradual sharpening in the photoluminescence and formation of a sharp lasing type of spectrum. This spectroscopy method can help in identification of emission wavelengths, lasing as well as the threshold intensities. It can also help in identifying the source of any emission signal in photoluminescence experiments.

We also use reflection and transmission spectroscopy to study reflection and transmission as a function of wavelength. This can also provide us the absorption spectrum ($1 - R - T = A$). These measurements involve the use of a white light excitation or the use of a wavelength sweep (scan over a range of wavelengths, one wavelength at a time). We use these techniques in our experiments on switchable and nonreciprocal metasurfaces based on vanadium dioxide and indium tin oxide to demonstrate nonreciprocal transmission and absorption. Another important aspect of these experiments is that they can reveal the presence of any sharp resonances within the structure.

In our experiments, we perform spectroscopy by coupling the signal obtained through the near infrared microscope into an optical fibre. The optical fibre then sends the signal to the benchtop spectrometers[107] which gives us the obtained spectrum.

1.2.5 Fourier Imaging (Back focal plane Imaging)

In the physics of sound, we often perform a fourier transform on the sound signal to obtain its temporal frequency components. The temporal frequency components

describe that the sound we hear is composed of how many different frequencies. Each frequency component has its own importance, however there also are a lot of other stray components that contribute to noises. The existence of these individual frequency components implies that any sound we hear is a superposition of basic sounds which are sinusoidal waves.

Similarly, the intensity for a single frequency electromagnetic field has a sinusoidal form. This implies that any spatial distribution of intensities is also a superposition of different spatial frequencies. One spatial frequency refers to one particular fringe pattern in young's double slit experiment. Because of its completeness, any intensity pattern can be decomposed into constituent fringe patterns or spatial frequencies.

Fourier optics is the study of these spatial frequency components of light. They help in the clear understanding of interference as well as quality of any image. The low spatial frequency components are usually the bulk features of a pattern, and the high spatial frequencies are intricate details like the edges.

Experimentally, the spatial frequencies can be isolated using a 2f system.[108] In our setup, a lens is placed so that the back focal plane coincides with the microscope objective (see Figure 1.2.5). This performs the fourier transform of our pattern on the camera.

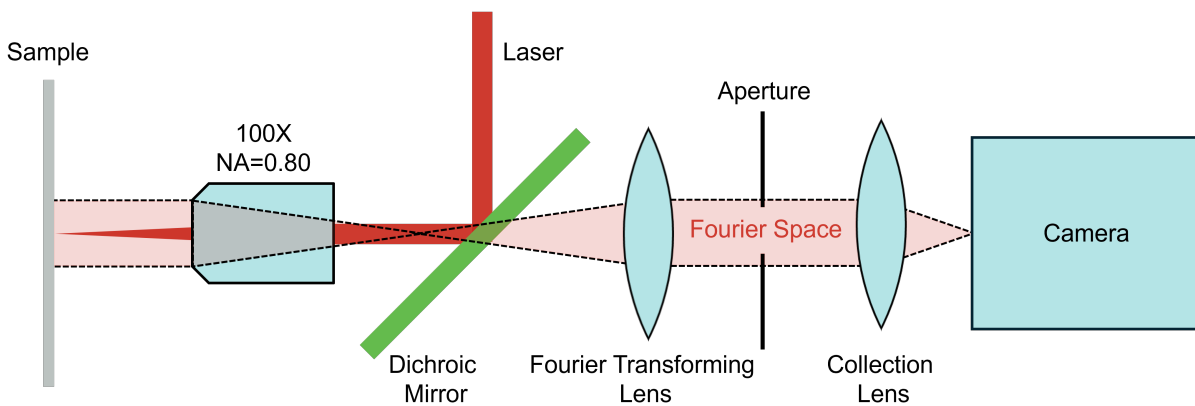


Figure 1.2.5: Back Focal plane imaging. The setup shows an arrangement to facilitate fourier imaging in reflection mode. The fourier transforming lens is placed so that the objective lies in its back focal plane. The aperture then cuts out the desirable features to project onto the camera.

We can add another lens to make our system a 4f system where we can perform both real space filtering and fourier filtering to see the angular dependence of light emitted

from our active nanodevices. This technique is particularly helpful in obtaining interference results to verify the coherence of our nano lasers as a typical light source.

Back focal plane imaging also provides basis for three very well-known techniques: dark field microscopy, bright field microscopy (regular microscopy) and phase contrast microscopy.[109] In dark field microscopy, only grazing incidence or scattered light is collected to reach the detector. Here the main illumination source is blocked using a spatial filter. Overall, it provides great contrast and sensitive detection of nanostructures.

The phase contrast microscopy involves the use of a phase mask along with the back focal plane imaging system described previously. Here, the phase mask ensures that the phase difference between different spatial components of light is adjusted in a way that we observe contrast in intensity due to their better interference.

In a nutshell, we use back focal plane imaging in our experiments on light emitting active nanophotonic devices for the verification of coherence and directionality[110] of radiation.

1.2.6 Stokes Polarimetry

The polarization of light is the optical analogue of the spin state in quantum mechanics. Hence, it is also represented on a Poincare' sphere (see Figure 1.2.6). The poles of the sphere represent left and right circular polarizations and the equatorial plane represents the horizontal, vertical and the diagonal polarizations. The polarization state of light is always a point on this sphere. If it lies on the equatorial plane, it is essentially linearly polarized otherwise it is elliptically or circularly polarized. The angle of projection of this point on the equatorial plane defines the orientation and the angle subtended with the polar axis defines ellipticity.

Stokes polarimetry is a method that helps us in finding our exact position on the Poincare sphere or simply the concise state of polarization. The four Stokes parameters define the total intensity, the degree of polarization, the orientation and the ellipticity respectively.

Experimentally we mostly employ the rotating polarizer polarimetry. In this technique, we use a rotating polarizer and a rotating quarter waveplate (connected on a controlled rotating stage) to select one basis polarization state at a time. This technique takes three known forms: transmission polarimetry, reflection polarimetry and spectroscopic polarimetry.[111] In transmission polarimetry the light transmitted through the nanodevice is sent into the rotating polarizer assembly and then

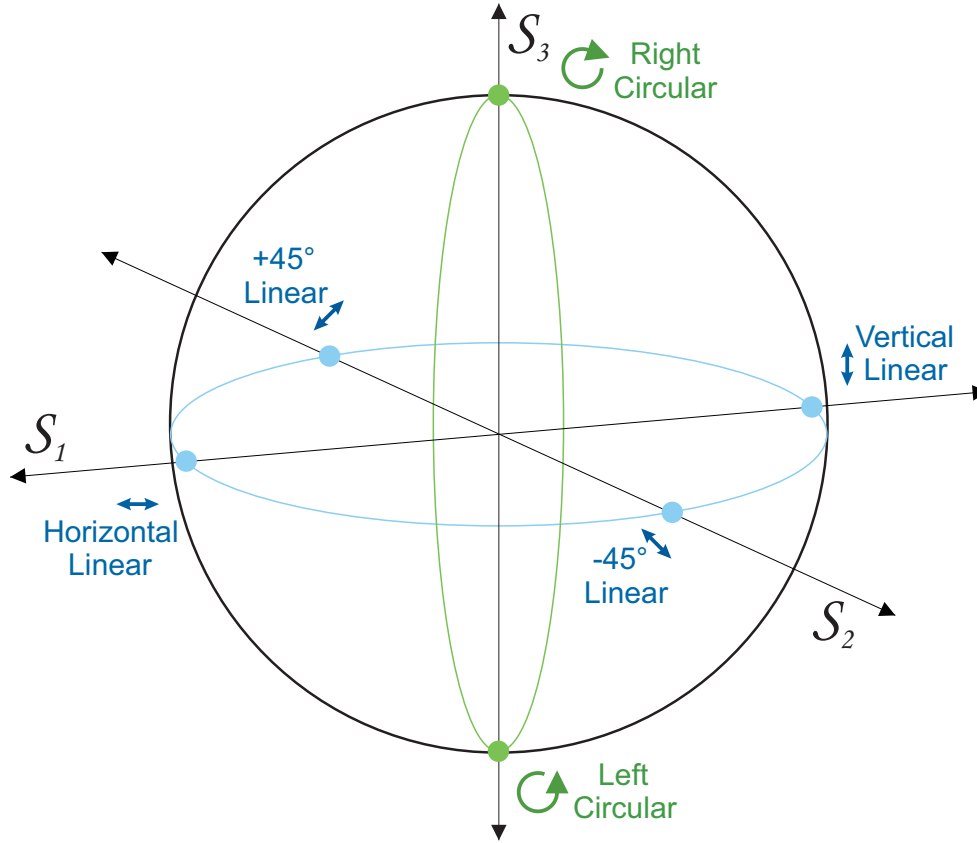


Figure 1.2.6: Poincaré Sphere. The poles of Poincaré sphere represent Left and Right circular polarizations and the horizontal plane represents the linear polarizations.

onto the camera. It is particularly used for experiments where the expected effect can only be observed in transmission like our Rashba effect experiment where we demonstrated polarized beam deflection of upconversion photoluminescence in transmission. Reflection polarimetry is like transmission polarimetry except for the fact that it uses the light reflected from our sample. It is often used in the cases where the substrate is reflective or opaque like GaAs, Sapphire or gold film. This is particularly useful for devices like the nanolaser, where transmission is usually blocked by the substrate and all the signal measured is in reflection only.

The spectroscopic polarimetry involves the use of spectrometer instead of a camera after the rotating polarizer setup. This provides spectral analysis of polarization and gives a clear understanding of which wavelengths are undergoing any kind of change in polarization. This is extremely useful in the cases where we observe two different emission wavelengths from the same sample like the nano lasers with two or more modes.

This technique is one of the most important secondary measurement techniques in nanophotonics. It will be a go-to technique with the advent of chiral metasur-

faces.

1.2.7 Temperature gradient optical diagnostics

Switchable and nonreciprocal nanophotonic devices often require an external stimulus to trigger switching action if they are not based on self-action. One of the most commonly used stimuli is heat or temperature variation.[112] Temperature change is the easiest applicable stimulus in the market as it doesn't need any complicated fabrication procedures to make electrodes or waveguides. It can be provided by simply providing a heater on the substrate.

This temperature gradient is provided across the sample to induce a change in the properties of materials used.[113, 114] For example, when we use vanadium dioxide in a metasurface we observe a change of phase at 68 degrees. We heat up the sample to temperatures greater than 68 degrees and wait for the device to reach a steady state. Once the steady state is reached we measure reflection, transmission and absorption. In practice, we use a copper mount with a hole, a ceramic heater and a Thorlabs temperature controller. The system is completely air cooled (no external cooling) because our temperature range is merely 20-80 degrees.

In nonreciprocal and switchable nanophotonic devices, study of repeatability and hysteresis is of particularly great importance. Temperature gradient optical diagnostics is used to heat and cool such devices repeatedly to provide hysteresis data in any optical parameter. This helps in understanding the device loss and the repeatability of transitions.

1.2.8 Nonreciprocal optical diagnostics

Nonreciprocal optical diagnostics is the experimental study of nonreciprocal optical devices. Using this method, one can identify nonreciprocity and even measure the degree of contrast. Usually, nonreciprocal optical action is achieved by magneto optics, nonlinearity, or time modulated crystals.

It is of great importance in the field of nonreciprocity to ensure that the optical components on both sides of the sample are identical. Which is to say that if we are measuring nonreciprocal transmission, we will have to ensure that the lens that is focusing the light on the sample and the lens that is collecting the light from the sample are identical.

In case of magneto optical nonreciprocity, the optical diagnostics are typically difficult. This is because we need to ensure the presence of uniform magnetic field. The testing of magneto optical devices involves the use of a reciprocal detection setup

with the addition of a variable magnetic field. The variable magnetic field tunes the faraday rotation effect and gives optimal nonreciprocity.

Time modulated nonreciprocity, requires time modulation of optical properties. Usually this is done in acousto-optic materials using acoustic signals. Setting up a source for acoustic signals and tuning optical properties for the purpose of breaking time reciprocal symmetry is the only difference in the setup.

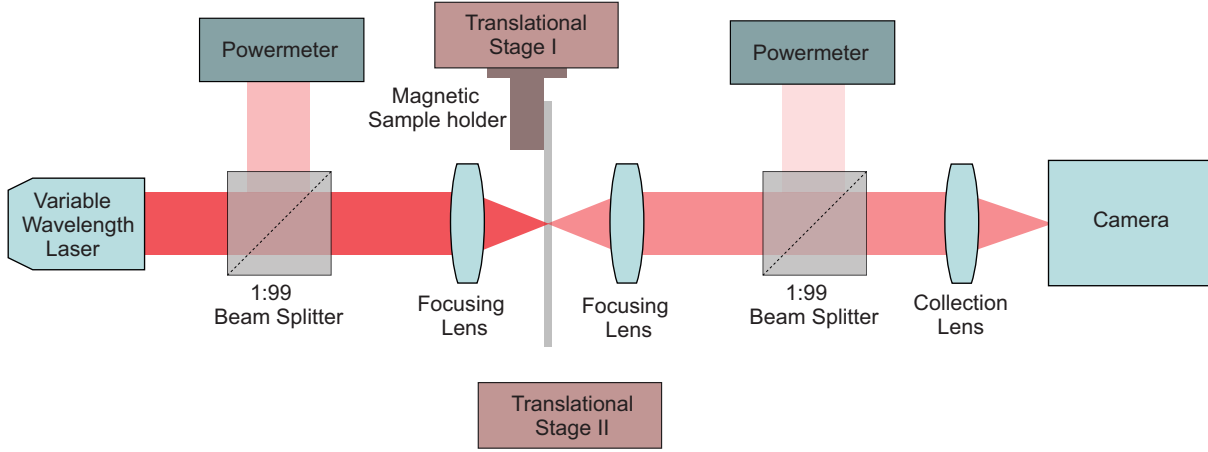


Figure 1.2.7: Nonreciprocal Optical Diagnostics. The schemataic represents the constructed setup for nonreciprocal experiments. The magnetic sample holder can be flipped and placed on an identical XYZ translational stage for reversing the direction of incidence. Identical focusing lenses are used to illuminate the sample and the powermeters measure power which is used to calculate the transmission at selected wavelength.

Nonlinearity driven nonreciprocity is typically the easiest to work with. This is mostly based on self-action, which means that the incident light is itself the stimulus that triggers the active material. The experiment mostly involves the use of a variable power and wavelength pulsed source in a simple transmission setup.

In our experiments on ITO and VO₂ based metasurfaces, we use an optical parametric amplifier with a wire grid polarizer to work as a variable power and wavelength source. For vanadium dioxide, we use the continuous wave mode of the amplifier (without sending the seed femtosecond laser). As shown in Figure 1.2.7, we use two XYZ translational stages with magnetic mounts to ensure flipping of the sample for backward excitation. Overall, we developed our own setup to verify non reciprocity in any nanophotonic device.

1.2.9 Zscan

The Zscan is one of the most fundamental and traditional techniques of nonlinear optics. It is used to characterize the nonlinear coefficients of a material. It can find

out two photon absorption coefficient and kerr coefficients by simple transmission measurements.[115]

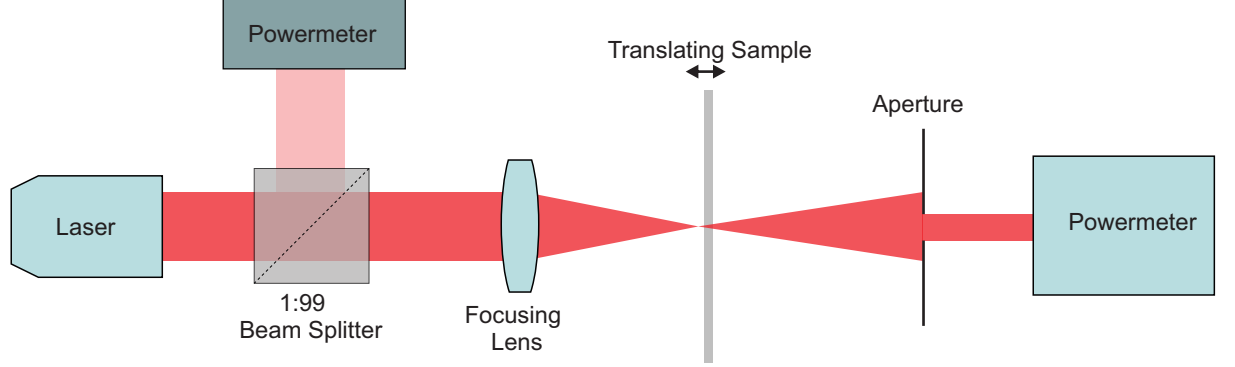


Figure 1.2.8: Z-scan setup. The closed aperture z scan employs an aperture in front of the detector. The sample is scanned in the propagation direction within the Rayleigh range using a translational stage.

In zscan, the sample is in the focal plane of a focusing lens. It is placed on a translational stage which is motile along the propagation direction of the illuminating beam. The sample is moved within the Rayleigh range of the gaussian beam (keeping the laser intensity constant) and the transmitted light is received in a bucket detector (photodetector). The intensity of transmitted light is noted for various positions of the sample within the Rayleigh range. The plot of this transmitted intensity against the position of the sample in the Z axis is popularly known as the Z scan.

The Zscan has two operational modes designed to measure different nonlinear coefficients.[116] These are the open aperture Z scan and the closed aperture Z scan. The closed aperture Zscan gives the estimate of the Kerr coefficient by calculating the axial phase shift ($\Delta\Phi_0$) from the difference in peak-valley transmission (ΔT_{p-v}) and transmittance of the aperture (S).

$$\Delta T_{p-v} \approx 0.406 (1 - S)^{0.25} |\Delta\Phi_0| \quad (1.1)$$

Where, $\Delta\Phi_0 = kL_{\text{eff}}n_2I_0$, $k = \frac{2\pi}{\lambda}$, $L_{\text{eff}} = \frac{1-e^{-\alpha L}}{\alpha}$, n_2 is the Kerr coefficient, α is the two photon absorption coefficient, L is sample length and I_0 is the peak intensity.

$$n_2 = \frac{\Delta\Phi_0}{kL_{\text{eff}}I_0} \quad (1.2)$$

The open aperture Z scan is used to measure nonlinear absorption. Here the photodiode is not shielded by any aperture and allows for all the light to enter. Typically

in this case we observe an overall dip in the curve, characterised by the nonlinear absorption. Quantification of this dip (T_{min}) is done with respect to known nonlinear materials to evaluate the nonlinear absorption.

$$T(z) = 1 - \frac{\beta I_0 L_{\text{eff}}}{2\sqrt{2}} \quad (1.3)$$

$$\beta = \frac{2\sqrt{2}(1 - T_{\min})}{I_0 L_{\text{eff}}} \quad (1.4)$$

Luminescence

Luminescence is of the most fundamental aspects in light matter interaction where a material produces light following absorption of energy. On the basis of method of absorption of energy, we observe four types of luminescence: fluorescence, phosphorescence, electroluminescence and photoluminescence. Each of these processes has been thoroughly studied and applied in different optical devices.

Fluorescence is the term coined for emission of light instantly upon the absorption of photons. Generally, a photon of smaller wavelength is absorbed to release photons of a longer wavelength. The characteristic feature of this process is the time frame which usually lasts from nanoseconds to microseconds. One of the most common applications of this effect is the use of biological marking dyes like janus green.

Unlike fluorescence, phosphorescence is not an instantaneous, but a much longer process. This is because phosphorescent materials have the capability to store optical energy chemically as triplet states. During the down transition from triplet states, we see gradual emission of photons.

Electroluminescence is one of the most widely used phenomenon in optoelectronics. The term refers to absorption of electrical energy in the form of current to give photons of desired wavelengths. It occurs mainly in direct bandgap semiconductors like AlGaAs where the electric current generates electron hole pairs and they undergo radiative recombination.

Photoluminescence however is an umbrella term that includes both fluorescence and phosphorescence. Biosensing uses quantum dots as labels and markers for the detection of biomolecules. The photoluminescence signal from quantum dots has implications in the field of medical diagnostics and bioimaging. Solar photovoltaics also utilises photoluminescence spectroscopy for material characterisation and recognition of recombination mechanisms. In certain cases it can also help in evaluating minority carrier concentration.

Upconversion photoluminescence is one of the most promising upcoming fields of re-

search in luminescence. They offer a mechanism to convert high wavelength photons to low wavelength photons by cascade effect. So far they happen to be the only solid state source of Ultraviolet radiation. Hence making their application more diverse as a universal light source.

However, the development of nanoscale light emitters is hindered by the challenges of efficiency, directionality and polarization control.

2.1 The problem statement

Manipulating luminescence features at nanoscale requires control over emission curves of the active materials used and the nanophotonic structure. Till date control over parameters like wavelength, intensity, quantum efficiency and lifetime has been demonstrated. The choice of active materials gives us the range of optical gain wavelengths and lifetimes, coupling with appropriate nanostructure allows us to choose specific wavelength, enhance intensity and the overall efficiency.

The ability to control optical properties of photoluminescence is of crucial importance in case of newer active materials like rare earth doped nanoparticles. The rare earth doped nanoparticles are used because of their strong photoluminescence in the near infrared region and for their ability to demonstrate upconversion photoluminescence in the UV-Vis region.[117]

Morphologically, these nanoparticles contain two units, a core doped with a rare earth metal and a shell made of a separate material. The core material is usually a semiconductor like Cadmium Selenide or a metal oxide like Titanium dioxide or even a dielectric like silica. The core nanoparticle is doped with a rare earth material like europium, erbium, ytterbium, or neodymium.[118, 119] This dopant serves the purpose of control parameter for the luminescent or magnetic properties of the nanoparticle.

The shell serves the purpose of providing biomechanical and chemical compatibility with its surrounding. Usually, the shell is inert in the bandgap region of the core material. Most used materials for this purpose are dielectrics or polymers.

These particles owing to their strong photoluminescence in the near infrared region serve the purpose of enhancing fluorescence in cells and tissues. A special class of these particles is also able to demonstrate frequency upconversion or simply converting lower frequency photons to higher frequency photons.[120, 121] This effect is used to enhance the efficiency of solar cells and to create solid state UV sources.

The synthesis involves purely chemical methods hence eliminating the need of bulky

vacuum chambers or cleanrooms. The core nanoparticles are prepared using colloidal synthesis, chemical precipitation, sol-gel synthesis or hydrothermal methods. The temperature, pH and precursor concentrations are the control parameters for the shape and size of the nanoparticles. The rare earth ions are introduced during the synthesis using precursors or via the post-synthesis doping methods. The most common methods to incorporate the rare earth metals is via the use of soluble salts or organometallic compounds. The surface of core nanoparticles is covered with a shell using methods like sol-gel coating and seed mediated growth. Again, here the reaction conditions can control the shell thickness.

Post synthesis, the nanoparticles are dissolved in an alcohol solution for deposition on samples. The concentration of nanoparticles in the solution determines the density of nanoparticles deposited. Once the solution is prepared, it can be deposited by simply drop casting on the sample or spin coating for nanometer scale precision. Due to the ease in fabrication and incorporation with photonic structures, rare earth nanoparticles are a very popular area of research.

Despite control over shape, size, materials, and intrinsic properties of these nanoparticles. The aspect of polarization of emitted light remains pretty much out of control.[122, 123] Unfortunately, locking of polarization is not possible via chemical methods. One of the most successful approaches in achieving control over polarization is the introduction of anisotropy.

Anisotropy implies the use of nanoparticles with shapes like an ellipse or a rod, all arranged in an order such that all the axes are parallel. However, this approach is extremely difficult to realise as it requires precise control over fabrication parameters.[124, 125]

An alternative and unused approach for achieving polarization control is coupling to resonators.[126, 127, 128] In this chapter, we try to couple the rare earth-doped nanoparticles to resonant dielectric nanoparticles in metasurfaces to solve the problem of polarization control. We start off with a more basic problem: controlling polarization of photoluminescence of rare earth doped nanoparticles using topological nanostructures. The photoluminescence lies in the near infrared region (telecom wavelength), hence making the fabrication of dielectric nanostructures easier (because of lesser size constraints).

In the second part, we solve a more difficult problem of isolating left and right circular polarizations in upconversion photoluminescence. Upconversion signal lies in the UV-Vis range which is half the wavelength of normal photoluminescence. This makes fabrication and design more difficult due to fabrication constraints. Also, the absorption of the dielectric should be negligible as the upconversion effect

requires extreme pump intensity. We demonstrate a beam deflector that separates the circular polarizations in upconverted light.

2.2 Polarized Luminescence using Topology

The photoluminescence from rare earth doped nanoparticles is a spontaneous emission effect. This implies that the emission is random in nature and hence there is random spin allocated to the emitted photons. However, if the emitters are coupled to a nano resonator or some sort of strong electric field, we can observe some degree of polarization in emitted photons.

The polarization of emission is controlled by altering the coupling to resonant nanostructures. Coupling of emitters to nanostructures is well studied for the control over emission properties. The polarization of light emitted is hence strongly dependent on the position of emitters with respect to the nano-resonators.[129, 130, 125, 131] This precise positioning of emitters is extremely difficult to ensure in nanophotonic devices as it involves the use of complex techniques like atomic force microscopy.

The coupling of a nanoresonator with a volume of rare-earth doped nanoparticles functions like a damped coupled oscillator.[132, 133] Here both the nano-resonator and the rare-earth doped nanoparticles are oscillators and the rate of conversion into luminescence is the damping coefficient.

To solve the problem of polarization control in photoluminescence, the only question on the table is which nano-resonator to use for optimal coupling.[134] The answer to this question lies in our application: polarization control. For selecting a particular polarization, we must couple our rare earth doped nanoparticles to a resonator that gives us a uniformly polarized near and far field.[135, 136] And to ensure optimal coupling, the resonance of the resonator must be broad and matching the luminescence wavelengths.

Most Mie nanoresonators can provide broad resonances, like the electric dipole and magnetic dipole modes, however, their near and far fields don't always have a uniform polarization. In this case, making a Mie resonant metasurface would only sharpen the resonance and not do much to the polarization.[137, 138] The implication of this fact is that isolated nanoparticles or oligomers would be a better choice than the metasurface or any kind of lattice.[139, 140] The second important factor to consider is the positioning of emitters with respect to the nano resonators. Ideally, we would desire a disorder robust resonator system to allow for imperfections in positioning of the emitters.

Our search is hence reduced to disorder robust nanophotonic structures that can polarize the mie modes of individual nanoresonators or oligomers.[141, 142] This effect can be engineered by using the topological phases of light.

Topological photonics is the physics of creation of robust and disorder resistant photonic devices.[143, 144, 145] Mie resonances like electric dipole and magnetic dipole hold special promise for implementations of topological orders of light.

One-dimensional topological structures have attracted a lot of attention in the fields of chemistry, photonics, and applied mechanics. The phenomenon of topological conduction of electricity in polyacetylene was awarded the Nobel Prize in chemistry in 2000. An analogous mechanical structure of chain of flip joints was demonstrated in nature physics in 2014.[146] Both these structures demonstrated the natural application of the concept of topological edge states. In polyacetylene, the conduction is facilitated by the pi electrons at the edge of the polymer chain, and in case of flip joints the edge joints have the maximum amplitude of oscillation.

The nanophotonic analogue of topological edge states was first studied in 2016 with the advent of one-dimensional zigzag arrays of dielectric nano resonators.[147, 148] The edge nano resonators of these chains are linearly polarized near field hotspots [149?]. Figure 2.2a is a schematic representation of our zigzag array, showing the near field hotspots (white discs) at the array edges. We can see the energy distribution in the full wave simulations showing the existence of linearly polarized edge states (see Figure 2.2c,d).

We exploit, these linearly polarized topological edge states in zigzag arrays to solve the problem of obtaining polarized photoluminescence from rare earth doped nanoparticles. In this project, we demonstrate coupling between zig zag nano resonator arrays (nanophotonic structure) and rare earth doped nanoparticles (active media) to control the polarization of luminescence.

2.2.1 Active media and nanophotonic structure

In our work, we employ the use of β -NaErF₄@NaYF₄ core shell nanoparticles having an average diameter of 25 nm in transmission electron microscopy image and schematic in Figure 2.1. The nanoparticles are prepared using layer epitaxial growth of dielectrics. These nanoparticles are chosen so that 976 nm wavelength laser is absorbed and photoluminescence at wavelength around 1532 nm is produced. This fluorescence is due to the orbital transition in the dopant: Er⁺³ ion. The round shape and small size of the nanoparticles is chosen to allow for easy agglomeration on the surface of the nano resonators.

Each individual nano resonator actually takes the form of a flat silicon disc. The silicon disk being a dielectric can host the desired Mie resonant modes (Electric and magnetic dipole). Figure 2.2b indicates that the electric dipole mode of the individual nanodisk is adjusted to match the photoluminescence wavelength of the core shell nanoparticles to facilitate the coupling between the active media and the nanophotonic structure.

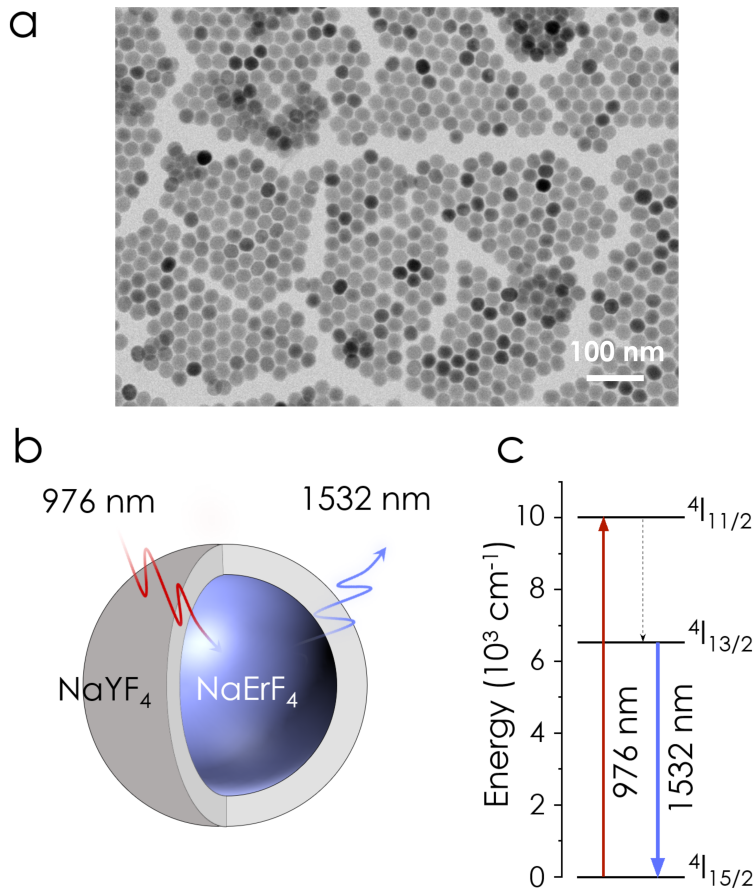


Figure 2.1: Structure and properties of Erbium doped core-shell nanoparticles.

(a) Transmission electron microscope image of nanoparticles. (b) Schematic of the core-shell structure of the nanoparticle. (c) Relevant energy levels of Erbium

In a zig zag array, the silicon nanodisks are coupled to their nearest neighbours. However, due to the zigzag nature the strength of mutual interaction between the nearest neighbour is high or low depending on the direction of polarization of the incident light. Such a system is described by a generalized Su-Schrieffer-Heeger (SSH)-type model with a gauge transforms independent topological invariant.

2.2.2 Topology of Zigzag array

Light localization via nontrivial topological states can be analysed using variants of the coupled dipole approximation also used in plasmonics.

The following is the expression for Hamiltonian of such a system. Here ω_0 is the resonance frequency, and the indices j and j' are the nanoparticles. a_{jv} is the annihilation operator for the multipolar eigenmodes having polarization v at the j^{th} nanoparticle. The electric dipole modes in the plane (x and y directions) are considered.

$$\mathcal{H} = \sum_{j,v} \hbar\omega_0 a_{jv}^\dagger a_{jv} + \sum_{\langle j,j' \rangle, v, v'} a_{jv}^\dagger V_{v,v'}^{\langle j,j' \rangle} a_{j'v'}$$

The coupling matrices are the planar unit vectors that are parallel and perpendicular to the linking vector between adjacent nano resonators. The coupling constants $t_{\parallel}$ and $t_{\perp}$ represent the co-polarized and cross-polarized coupling cases and $\otimes$ stands for the direct product. For our system the ratio of the coupling constants ($t_{\parallel}/t_{\perp}$) can be estimated to be -2.

The ratio of coupling constants is in fact the ratio of energy of interactions of magnetic dipoles in two different situations: the vector ($\mathbf{r}$) joining the parallel dipoles ($\mathbf{m}_1, \mathbf{m}_2$) is parallel or perpendicular to the dipole moments. In case the first case, the energy is given by $U = -\frac{2\mu_0}{4\pi r^3} |\mathbf{m}_1| |\mathbf{m}_2|$ and in the latter case the energy is $U = \frac{\mu_0}{4\pi r^3} |\mathbf{m}_1| |\mathbf{m}_2|$, hence giving us the ratio of -2 [150].

$$V^{\langle j,j' \rangle} = t_{\parallel} e_{\parallel}^{\langle j,j' \rangle} \otimes e_{\parallel}^{\langle j,j' \rangle} + t_{\perp} e_{\perp}^{\langle j,j' \rangle} \otimes e_{\perp}^{\langle j,j' \rangle}$$

Figure 2.2e shows the energy spectrum of the zigzag array prepossessing a band gap with two degenerate edge states associated with the near field distribution. Nonzero longitudinal components of k vector were accounted for in the coupling constants.

The topological invariant is 1 within the band gap, which implies the emergence of one polarization dependent state at every edge of the zigzag chain. We can visualize the resonant coupling between electric dipole modes of adjacent disks. The neighbouring dipoles can couple strongly or weakly depending on the mutual orientation. The weakly coupled disks at edges host the polarized edge state.

The edge states that result due to zigzag structure are topologically protected against system disorder and fabrication imperfections. In case of arrays with odd number of disks, we observe orthogonally polarized edge states and for even numbered arrays,

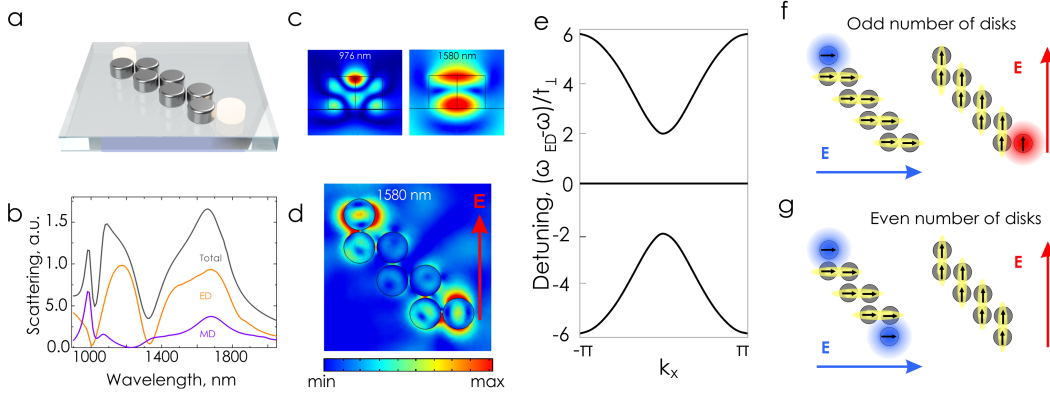


Figure 2.2: Topological zigzag arrays of dielectric nanoresonators. (a) Concept image of the zigzag array hosting topological edge states. (b) Multipolar decomposition of a constituent disk nanoresonator. (c) Near-field distributions of a single disk for plane wave excitation at around the pump and the photoluminescence wavelengths. (d) Full-wave simulation of edge localizations in the zig-zag array illuminated with a diagonal polarisation. (e) Dispersion curve for Bloch modes in periodic zigzag chain (f, g) Schematics of the edge states formation in arrays with (f) even and (g) odd number of nanoresonators. Blue and red colours are used to distinguish between two principal polarisation of excitation: horizontal and vertical. Yellow joints visualise stronger dipole-dipole coupling between the modes of the near-neighbour disk for the cases of horizontal polarisation excitation (left) and vertical polarisation excitation (right). Topological states (left side, blue color – horizontally polarised; right side, red color – vertically polarised) form at the edge disks that are weakly coupled to their near-neighbours.

we observe co polarized edge states (Figure 2.2f,g). The linear polarization of the edge modes makes them an ideal choice for coupling with the rare earth doped nanoparticles.

2.2.3 Experiment and Results

The fabrication is a four-step process is carried out on a 500 micron thick fused silica substrate using amorphous Silicon. In the first step we deposit a 300 nm layer of amorphous silicon using low pressure chemical vapour deposition. This is followed by the deposition of electron resist PMMA A4 950 on the sample, the pattern is then drawn on the resist using electron beam lithography. We evaporate a thin chromium film followed by a lift of process to harden the patterned mask. Finally, we use reactive ion etching to transfer the mask pattern on the Silicon film. The residual Chromium is removed using wet etching. Using the aforementioned process we fabricated zigzag arrays of Silicon disks having 300 nm thickness, 510 nm diameter and a mutual spacing of 30 nm between the nearest neighbours. Then we spin coat the erbium doped nanoparticles on the top of the dielectric nano resonator arrays.

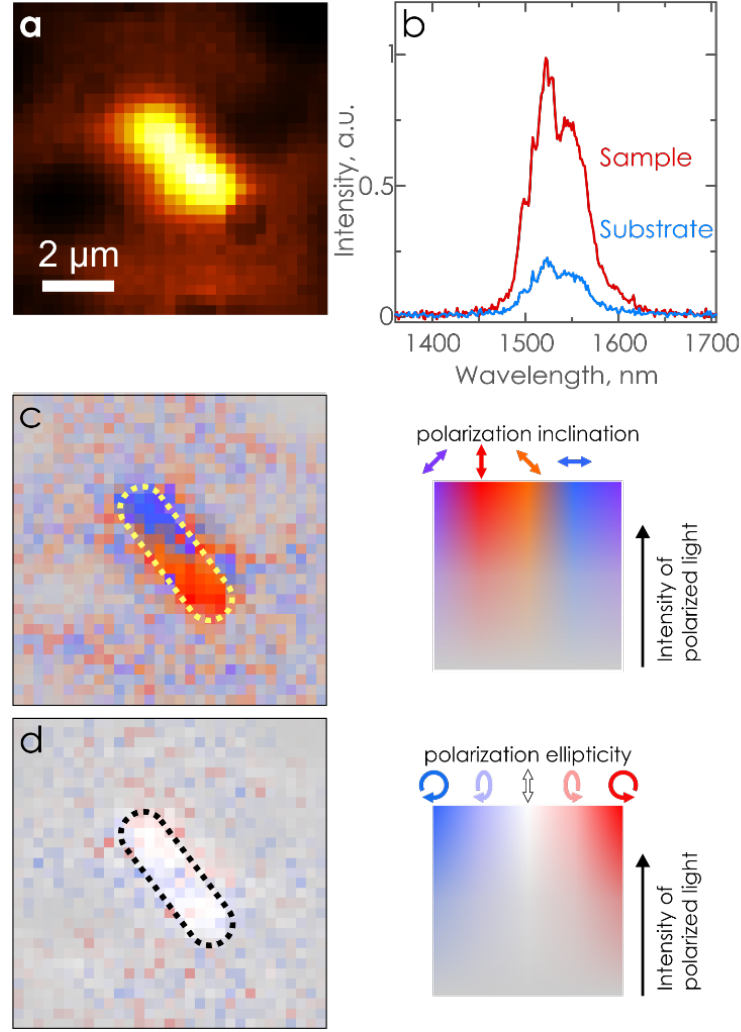


Figure 2.3: Topology-controlled photoluminescence of Er^{3+} core-shell nanoparticles. (a) Camera image of the emission enhancement from the Er^{3+} nanoparticles in the vicinity of the zigzag array. (b) Photoluminescence spectra of the Er^{3+} nanoparticles on top of a zigzag array vs on top of a bare substrate. (c, d) Spatially-resolved polarization states of photoluminescence showing (c) polarization inclination angles and (d) ellipticity of photoluminescence.

The optical diagnostics were carried out using our custom built near infrared microscopy setup (section 2.2.1). The pumping was provided by 976 nm CW diode laser. However, to narrow down the laser output to match the size of the zigzag arrays we made a telescope with a $f=125\ \text{mm}$ lens and objective lens Mitutoyo Plan Apo NIR HR (X100, 0.7NA). We measured the reflection of the zigzags using near infrared camera Xenics bobcat 320. The photoluminescence of erbium doped nanoparticles was verified using the NIR quest spectrometer from ocean optics. After verifying the presence of nanoparticles on the zigzag arrays, we compared the photoluminescence from nanoparticles coated on zigzags to those on the substrate to see five-fold enhancement due to coupling (Figure 2.3a,b). To quantify this en-

hancement further, we estimate the Purcell factor to be 190 using the Q factor, effective mode radius and the wavelength of the zigzag nanoresonator.

We proceed to the main experiment to measure the spatially resolved polarization states. We used polarizer and quarter wave plate to perform the rotating polarizer stokes polarimetry. We visualize the polarization inclination and observe that the PL from the edges of the zigzags is either co polarized or orthogonally polarized (Figure 2.4a,b). The color gray implies the presence of unpolarized light, red for vertically polarized and blue for horizontally polarized. The ellipticity of emission shows that the photoluminescence is strictly linearly polarized (Figure 2.3d), however the degree of polarization is not very high owing to nonuniform deposition of nanoparticles. We also perform polarimetry on linear arrays of nano disks to observe unpolarized emission (Figure 2.4d). Through our polarimetry experiments we conclude that the luminescence from erbium doped nanoparticles located on the edges of the zigzag arrays becomes polarized due to coupling with the topological edge states of the zigzag array.

We also compare the polarization of photoluminescence from odd and even numbered zigzag arrays. We find out that qualitatively the 9 disk, 11 disk and 15 disk arrays are similar as they possess orthogonally polarized edge states. However, quantitatively, the degree of polarization and overall enhancement of PL is greater for the longer array. This is because the longer chain becomes easily and noticeably depolarized. The 14 nanodisk array however shows copolarized edges. The bulk of the 14 disk array like the 15 disk array produces depolarized PL. However, the enhancement in case of even numbered disk array is slightly lesser than that of the odd numbered disk array.

2.2.4 Conclusion

We demonstrate a harmonious combination of erbium doped nanocrystals and nanoresonator array. We show a method to successfully integrate active media with dielectric nanophotonic structure by using the physical method of spin coating to couple the nanoresonator array with erbium doped nanocrystals. The nanoresonator array brings the functionality of polarized modes and the erbium doped nanoparticles serve the purpose of light generation. Hence we observe polarized light generation from a nanophotonic source.

The rare earth doped nanoparticles are a very popular source of upconverted photons in the UV-Vis range. Isolation of polarized upconverted photons is a highly inefficient process because of losses upon the use of external polarizer. As the next step, we add complexity to the function of our active nanophotonic device. We try

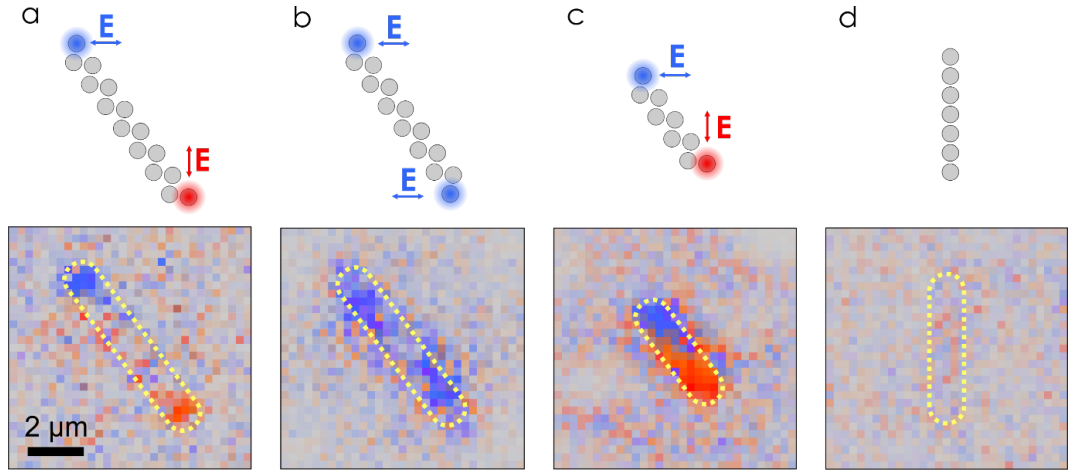


Figure 2.4: Polarization control of photoluminescence with topological edge states. Spatially resolved polarization states of photoluminescence for (a) 15-nanodisk zigzag array, (b) 14-nanodisk zigzag array, (c) 9-nanodisk zigzag array, and (d) topologically trivial straight array of resonant nanodisks. The color-map is analogous to that used in Fig. 2.3c.

to develop a device capable of generating beams of polarized upconverted photons right at the source.

Overall, we believe that this result can lead to the development of a crude understanding of active nanophotonics. The principles used in this study are holistic and applicable in the design of many more active nanophotonic devices. Our research primarily focuses on providing proofs of our concepts, however further research in these directions can provide useful industrial devices. Additionally, it can become a route for development of novel designs for disorder immune nanophotonics, high speed routing and even quantum light sources at room temperature.

2.3 Photonic Rashba effect in Upconversion

Rare earth doped nanoparticles as discussed in the previous sections are modern active materials usable in photonic devices. The most attractive property in their palette is the ability to upconvert lower energy near infrared photons to higher energy UV-Vis photons. The quality of upconversion is desired in various applications like solar energy harvesting, color displays, biomarking and photocatalysis. However the scope and prospects of their applications as upconversion nanoparticles is marred by their low conversion efficiency. The degree of control over emission properties is the gold standard of applicability as an active material in a photonic device.

So far, as discussed previously, control over polarization remains a stalwart challenge in their utilisation in industrial devices. Although it is possible to achieve anisotropic

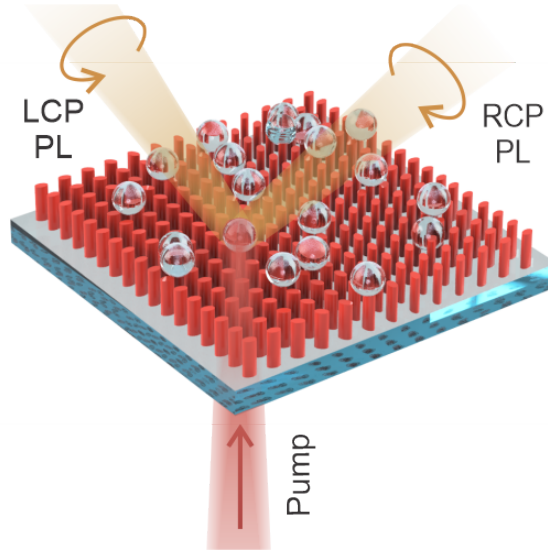


Figure 2.1: Concept image of a phase-gradient dielectric metasurface with dropcasted rare-earth doped nanocrystals demonstrating the photonic Rashba effect in emission.

polarization control using single or aligned nanoparticles. It is very difficult to realise this strategy with precise control. However as shown in the previous section, the rare earth nanoparticles can be coupled to nanophotonic structures which host polarized modes or can alter polarization in any way.

Here we aim to produce circularly polarized upconverted photons with maximum conversion efficiency. Which implies that we desire to produce circularly polarized photons from the get-go. The challenge in this functionality is the design of nanophotonic structure that can facilitate the generation of polarized light.

In this project, we couple the rare earth doped nanoparticles to a phase gradient dielectric metasurface to isolate circularly polarized upconversion photoluminescence as shown in Figure 2.1. This way, we eliminate the need of external polarizers and quarter wave plates and improve the overall efficiency of upconversion. We ensure that most of the upconverted photons are circularly polarized. This effect is also known as the Rashba effect in photonics.

2.3.1 Rashba effect

The Rashba effect is a relativistic effect for electrons having planar motion in the presence of an applied electric field. It is caused by the coupling between the spatial and spin degrees of freedom of the electrons. The effect is attributed to a term in the electron's Hamiltonian which accounts for the relativistic motion of the electrons. This term lifts the spin degeneracy in the electronic energy spectrum. As a

result we observe splitting of the electronic bands for both values of spin (Similar to zeeman splitting).[151] The Rashba effect is widely used as a tool in spintronics and magnetronics.

In photonics, Rashba effect is mostly a spatio-geometric effect. The polarization of light serves the purpose of photonic spin and plays a crucial role in spin orbit coupling.[152] As a result, the photonic rashba effect actually splits the opposite spins in opposite directions in the momentum space.[153, 154] Which in laymans language means splitting of opposite circular polarizations in opposite directions.[151, 155]

The photonic Rashba effect has also found its application in plasmonics.[156] One profound application is where circularly polarized light is coupled to surface plasmons generated in metasurfaces having broken inversion symmetry. In topological photonics, ever since the advent of spin dependent photonic states there is a need to have a spin filter or selector in the photonic topological insulators. The Photonic Rashba effect offers a method to engineer these spin-textured photonic states.[157] The field of spin orbit coupled photonics also utilizes the Rashba effect as its underpinning concept. Here the polarization of the photon decides the direction of propagation and overall route of the signal, hence making an intelligent pathway for optical communication.

However in case of active media, Rashba effect is of special interest because of its ability to manipulate polarization and infact isolate opposite polarizations. An example is the recent demonstration of photonic Rashba effect from Tungsten Selenide valley excitons incorporated into photonic crystal slabs with geometric phase defects. The spin dependent photoluminescence in momentum space originating from valley excitons having opposite polarizations. Another active nanophotonic device based on Rashba effect generates directional polarized photoluminescence from perovskite metasurface.[158]

2.3.2 Experiments and Results

The upconversion nanoparticles are prepared using a co precipitation method. The morphology of prepared nanoparticles is revealed in the TEM image, the nanoparticles assume uniform spherical structure with an average diameter of 27 nm. The presence of Yb^{3+} ion ensures that the nanocrystals absorb near infrared light at 980 nm. The $^2\text{F}_{5/2}$ excited state has an energy gap of around 10000 cm^{-1} with respect to the ground state. Once pumped, the Yb^{3+} ions transfer the energy to Tm^{3+} ions via conduction and upconverted photons of wavelengths 455 nm, 480 nm, 695 nm and 800 nm are produced. The concentration of Tm^{3+} is critical to our preparation

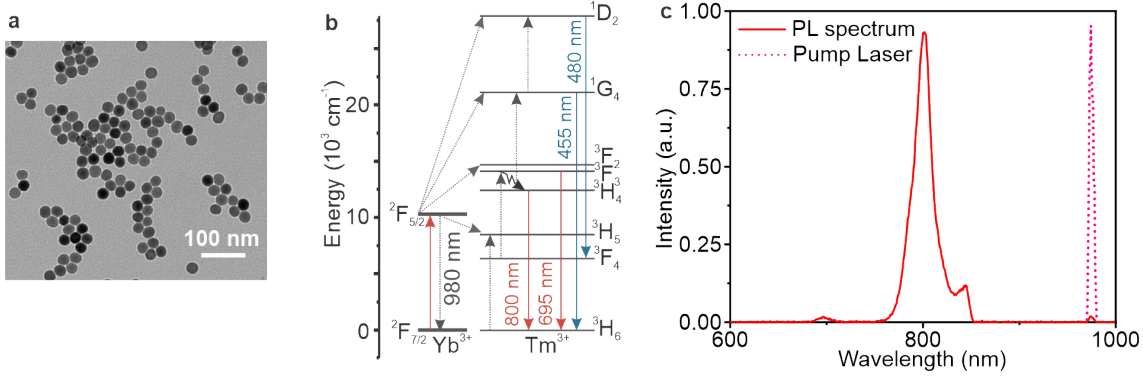


Figure 2.2: Nanocrystals with upconversion photoluminescence for coupling to a geometric-phase metasurface. (a) Electron microscope image of NaYF₄:20% Yb, 4% Tm nanocrystals. (b) Energy levels of the nanocrystals. Inset shows a core-shell structure of the crystals. (c) Spectral measurements of upconversion PL from the nanocrystals at around 800 nm (red solid) upon optical excitation at 980 nm (magenta dashed)

as it controls the upconversion wavelength.[159] This concentration was adjusted to be 4 mol to give maximum possible signal at 800 nm. The concentration of Yb³⁺ (20 mol) determines to an extent the efficiency of upconversion as it is responsible for the absorption of 980 nm pump. As a result of our synthesis, we observed upconverted photons of 800 nm wavelength at the pump intensity of 700 W/cm².

Our beam deflecting metasurface has elliptical nano cylinders tilted at a particular angle. This tilt allows them to serve the purpose of quarter wave plate. This results in a $\pi/2$ phase difference between two orthogonal polarizations. The cylinders are then arranged into 14 element super cells, each cylinder is rotated at a particular angle to create opposite phase gradients for opposite helicities of incident polarizations. This phase gradient allows for the assignment of opposite wave vectors to the photoluminescence of photons with different spins implying their splitting in the fourier space. Such phased array metasurfaces help in achieving control on the direction and phase of electromagnetic beams. This element-by-element engineering of beam profile across the area of the metasurface can also help in generating hybrid light beams and excitation of non-trivial modes in other samples.

The phased metasurface is designed using CST Studio Suite. Compositionally, this phased array is an arrangement of elliptical silicon nano pillars on a fused silica substrate. The height of each pillar is 434 nm and the side of the square unit cell is 427 nm. The major and minor axes of the elliptical cross-section are 191 nm and 106 nm respectively. The main reason behind choosing an elliptical cylinder is its ability to introduce birefringence, or simply scattering light differently for different polarizations with respect to the ellipse axes. The super cell consisting of 14 pillars

whose angle of rotation increases from $[0-\pi]$ creates a 2π phase gradient within the supercell. This is done to implement the Pancharatnam-Berry phase concept where the local phase depends on the pillars' orientations. The dimensions of the unit cells and the rotation of the ellipses is engineered to work at the upconversion wavelength of 800 nm. Due to our design, the metasurface has highest transmission and the highest percentage of beam deflection at our wavelength of interest.

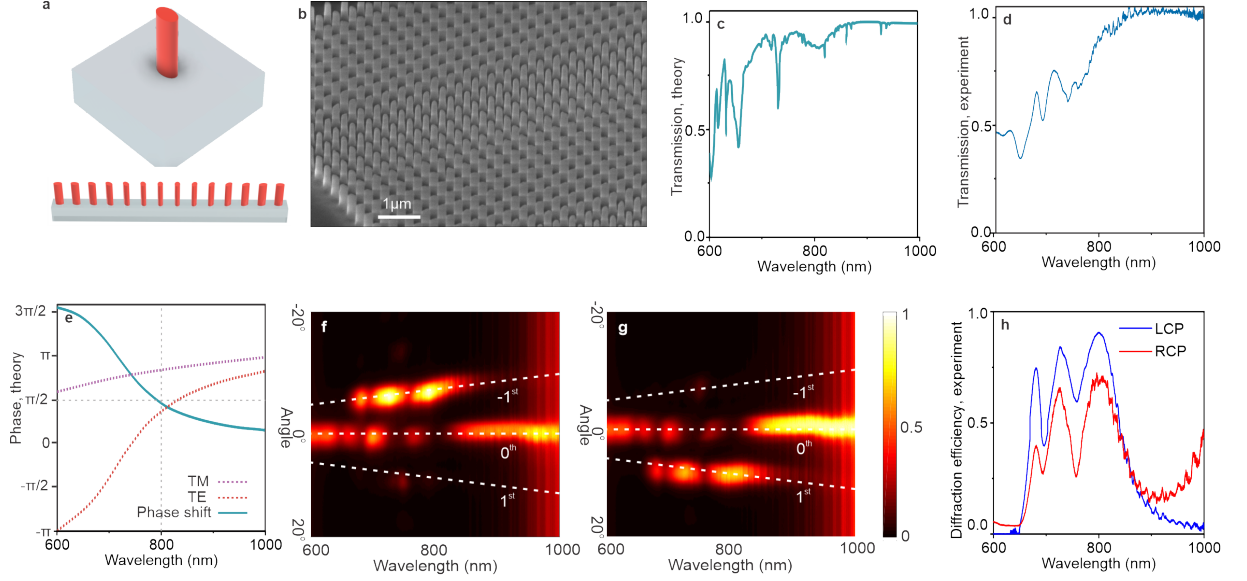


Figure 2.3: Dielectric metasurface with a geometric-phase gradient. (a) Top: schematic of a unit element pillar. Bottom: super-cell consisting of 14 pillars continuously rotated within π range of angles. (b) Electron microscope image of the fabricated metasurface. (c) Transmission spectra with 0th and $\pm 1^{\text{st}}$ diffraction orders calculated theoretically. (d) Experimentally measured total forward transmission. (e) Theoretically calculated phases and phase differences in transmission of two principal linear polarizations oriented along two elliptical axes of the unit cell. (f,g) Experimentally measured angle-resolved white-light transmission spectrum of the metasurface filtered at (f) right-circular and (g) left-circular polarization. Dashed lines mark positions of 0th and $\pm 1^{\text{st}}$ diffraction orders. (h) Experimentally measured diffraction efficiency of the metasurface (a portion of transmitted light that is directed towards +1st diffraction order (for RCP) and the -1st order (for LCP)).

The fabrication of metasurface was done using the standard electron beam lithography process just like the topological nanostructure created in the previous section.

Then we proceeded with the testing of our metasurface using angle resolved transmission spectroscopy using a tungsten halogen white light source, where the incoming light was projected normally on the metasurface. The fourier plane was the one we actually sent to the entrance slit of our monochromator equipped with an Andor

iXon 888EMCCD-camera. There is almost no beam deflection at 980 nm, however we do find diffraction of circularly polarized light at 800 nm. This is verified by filtering the left and right circular polarizations using waveplate and polarizer before the monochromator slit. This means that there is accumulation of some in plane momentum for circularly polarized light upon passing through the metasurface, confirming the Rashba effect. We can quantify the diffraction efficiency by normalizing the intensity of the diffracted light with respect to the transmitted light in the 0th diffraction order. We report diffraction efficiency of 90% for left and 70% for right circularly polarized light at 800 nm respectively. Our preliminary measurements prove that the fabricated metasurface is suitable for coupling with the nanoparticles that produce upconverted light at 800 nm.

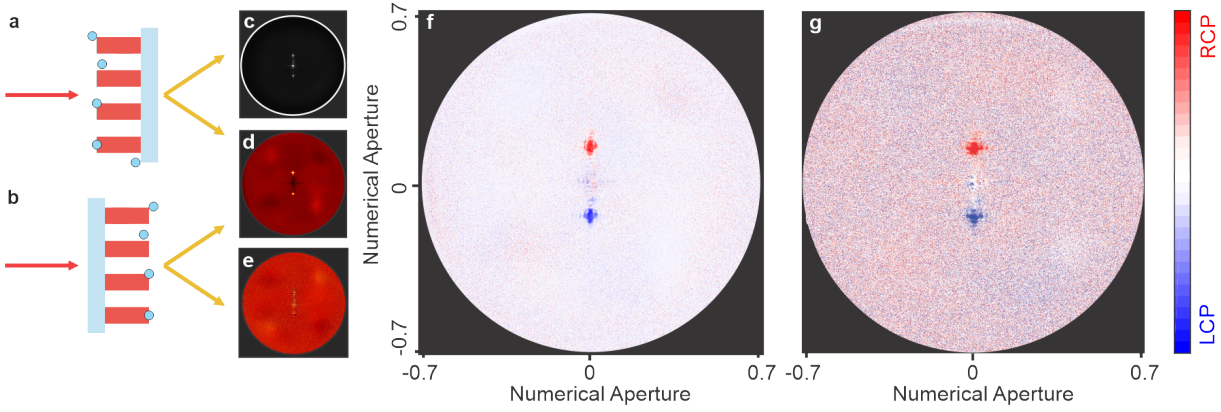


Figure 2.4: Experimental demonstration of optical Rashba effect in upconversion photoluminescence. (a,b) Schematics of an experimental method for the observation of Rashba effect. (c-f) Stokes vector polarimetry performed over the directionality diagram of the observed PL in configuration depicted in (a). (c) Total intensity. (d) Degree of polarization. (e) Polarization inclination. (f) Ellipticity. (g) Directionality diagram of the ellipticity measured in (b) configuration. Both directionality diagrams (f) and (g) of the experimentally measured PL in the momentum space show separation of the spins for both sample orientations corresponding to optical Rashba effect.

We proceed by dropcasting the prepared upconversion nanoparticles on the metasurface and measure the resulting PL signal. To measure upconversion PL from the sample, we pump the nanoparticles using 980 nm CW diode laser just like we did in the previous section. We verify the observation of upconverted 800 nm PL signal for the sample. However, this time the spectral and microscopic measurement was carried out in transmission instead of reflection because of the need to observe diffraction orders due to the inherent Rashba effect. The pump signal was filtered out using a 925 nm short pass and a 980 nm notch filter.

In the main experiment, we study the momentum space image or the directionality

diagram of the obtained upconversion PL. We set up our near infrared microscope in transmission mode using the 980 nm CW diode laser as our pump. We use a lens of ($f = 100$ mm) and place it in the back focal plane of the collection objective to obtain the fourier image of the sample. We image the directionality diagram using a Starlight Trius Pro-694 camera.

After obtaining the directionality diagram, we perform the rotating polarizer stokes polarimetry on the momentum space upconversion PL signal. We obtain the complete set of stokes parameters in the momentum space. The total intensity shows the presence of three spots, the 1st and the 0th orders respectively. The degree of polarization plot reveals that the 0th order is completely unpolarized, however both the first orders are completely polarized. The ellipticity reveals that the both the first diffraction orders are left and right circularly polarized respectively, corresponding to the spin dependent splitting of emitted photons.

2.3.3 Conclusion

We repeat our set of the angle-resolved polarimetry measurements for two sample orientations: sample side with nanocrystals facing the excitation source, and sample side facing the detector.

We report momentum-space splitting of photon spins for both orientations of the sample thereby confirming observation of Rashba effect in upconversion PL. We isolated both left and right circular polarizations in upconversion PL with almost 90% efficiency. This experiment paves the road for use active nanophotonics for PL manipulation and further application in the field of nanoscale light sources.

Lasing

Luminescence is the most basic and crucial capability in the development of the most fundamental optical device: the light source. Once control over all the parameters of luminescence is achieved, we strive to improve the quality of illumination and increase the power output.

The natural improvement in a luminous light source is the development of laser. Laser provides highly directional, coherent and narrow line emission. Laser, as an acronym was coined in the 1960s.[160] It stands for Light Amplification by Stimulated Emission of Radiation. As the name suggests, the stimulated emission process ensures that the generation of photons is triggered by pre-existing photons. This is inherently different from the luminescence discussed in the previous chapter as that was based on spontaneous emission.

In this chapter, we further raise the bar of the problem statement that we attempt to solve. After the development of controlled light sources, we strive for the development of nanolasers.[22] The development of nanolasers involved not only the use of a gain material, but also a high-quality factor feedback mechanism to ensure overall optical gain due to stimulated emission. Any functioning laser generally has three components: Gain medium, optical cavity and a pumping mechanism.[160]

As previously discussed, various materials like semiconductor quantum wells, quantum dots and many other active materials can serve the purpose of providing optical gain at the nanoscale. The optical cavity, or simply a space where the light is trapped usually consists of a tube enclosed by two mirrors one of which is partially reflecting. At the nanoscale, this purpose is served by metasurfaces having high Q near field hotspots. The near field hotspots can trap most of the incident energy in very small volumes, only allowing a little bit to leak to the far field. Hence making amplification of stimulated emission possible. We can use commercially available pulsed lasers or use electrodes to provide electrical pumping.

In the development of nanolaser, we must first choose an optimal gain material. The

optimal gain material must be chosen so that compatibility with the designed high Q metasurface is maximised.[161, 162] In our research we choose the commercial semiconductor quantum wells as our active media. This is because they have the best optical gain amongst all the available materials.

Next, we choose the nanophotonics that support the desired high Q resonances. Here we have two good options: Topological corner states and Anapole states. We try both the structures to demonstrate lasing at the nanoscale. We implement both the concepts in a membranous structure, which means that we pattern quantum wells as a membrane to ensure that the maximum field in the near field hotspots overlaps with the quantum wells.

In the first section of this chapter, we describe the use of topological corner states as the nanolasing mechanism. Topological corner states are known for their robustness against any disorder or fabrication imperfections.[162] The quality factor however cannot fully controlled, although still high quality factors can be obtained.

In the second section, we show lasing from anapole metasurfaces. The anapole states are dark scattering modes that are combination of electric dipole and toroidal dipole modes. The electric dipole and toroidal dipole modes interfere destructively in the far field.[163] In this case, we can get extremely high quality factors but they are strongly dependent on the geometry and fabrication quality.

3.1 Topological Lasing

As discussed previously, topological photonics is a lucrative division of nanophotonics owing to their resistance against disorder and imperfections. Of particular interest are the edge states that occur at the interface of two topologically distinct media. Combination of these edge states with optically active media offers both unravelling of new physics and potential applications like topological lasers.

Topologically protected lasing action has been demonstrated in the recent years in micro resonator arrays, unidirectional outcoupling of laser emission from magnetic field biased photonic crystal cavities having arbitrary shapes.[162, 164] It is of paramount importance to control the lasing threshold because the high near fields can lead to unwanted nonlinear effects like absorption and the Kerr effect.[165, 166]

As described in the previous chapter, research has been carried out in the direction of bringing topological photonics to the nanoscale.[167] Nanostructures made of high index dielectric materials and specialized lattice arrangements open up possibilities

to develop improvised photonic band diagrams and hence allow control over topological invariants.[168, 169] Intelligent design of such nanostructures can enable the creation of topological edge states[170] favourable for nanophotonics.

We already studied the implementation and application of 1D topological structures based on the SSH model. In this section we focus our attention towards two dimensional topological structures. Materials like graphene and MoS2 are two topologically non trivial materials having the ability to possess distinct topological invariants owing to their structure.[171, 172] The two dimensional hexagonal honeycomb like structure of graphene generates dirac cones in the band diagram. The dirac cones occur at the corners of the Brillouin zone where the conduction and the valence band touch each other (signifying massless dirac fermions).

Topologically, the honeycomb lattice owns a number of non trivial topological properties due to its three fold symmetry and dirac cones.[173] The edges of Graphene or MoS2 sheets are known to possess edge states or surface states that are topologically protected. This happens because the edges or the surface is topologically different to the bulk. The dirac cones at the band edges signify topologically protected edge states giving rise to unique properties. Interestingly, Topological insulators were awarded the Nobel prize in 2005 and the discovery of graphene was awarded the same in 2010. The generality of the concept of topology inspired the design of honeycomb lattices for photonic applications. Topological edge states in honeycomb lattices have been shown to host high quality modes in microscale and millimetre scale photonics.[85, 174] We try to implement these lattices on the nanoscale on InGaAsP platform which also hosts our quantum wells.

This way we combine active media with topological photonics to demonstrate low threshold lasing at room temperature. We design topological cavity relying on a closed valley-hall domain wall created by inversion of a staggered sublattice potential in a honeycomb lattice. The triangle shaped cavity is implemented in a dielectric membranous structure. The two triangular holes in each unit cell of the bipartite lattice have different dimensions that leads to the opening of bandgaps near the Dirac points of the Brillouin Zone.

3.1.1 Topology

Our system can be modelled using a 2D tight binding model. The Hamiltonian can be expressed as:

Where a and b are the creation and annihilation operators respectively for the i^{th} unit cell, $\sum_{\langle i,j \rangle}$ indicate summations over the nearest neighbours, $t > 0$ is the hopping

amplitude , and M is a parameter that destroys the inversion symmetry by detuning of sublattice. The tight binding model offers a system of coupled equations for normal magnetic dipole moments having nearest neighbour electromagnetic coupling. The electric field is in the plane of the metasurface and TE polarized, the magnetic field around each hole has a $\mathbf{z}$ component. A negative sign in front of the coupling constant t implies the presence of transverse dipolar interaction.

The linearly dispersive chiral edge states crossing the bandgap can be interpreted in the continuum limit from the Hamiltonian applicable at $|t| \gg |M|$ in the corners of the Brillouin zone.

$$\hat{H} = - \sum_{\langle i,j \rangle} t a_j^\dagger \hat{a}_i + -M \sum_i t (-1)^i \hat{a}_j \hat{a}_i^\dagger$$

$$\hat{H}_D = \nu_D (\hat{\sigma}_x \hat{\tau}_z \delta k_x + \hat{\sigma}_y \hat{\tau}_0 \delta k_y) - \hat{\sigma}_z \hat{\tau}_0 M$$

The Pauli matrices $\hat{\sigma}$ and $\hat{\tau}$ are operators for the sublattice and valley degrees of freedom, respectively.

This effective electromagnetic Hamiltonian can also be obtained for the 2D photonic crystal directly from the Maxwells equation via planar wave expansion method. In case of our structure we use the assumption $M > 0$, the case $M=0$ refers to the dirac cone situation of graphene implying the presence of massless dirac fermions. Time-reversal and parity symmetries prevent the lifting of degeneracies. The breaking of parity symmetry via a staggered sublattice potential introduces a bandgap. The following expression represents the dispersion relations for two bulk bands.

$$\omega_{\pm}(\delta k) = \pm \sqrt{\nu_D^2 (\delta k_x^2 + \delta k_y^2) + M^2}$$

Due to opposite parities of Berry curvatures, we witness a reduction of the winding number to 0. However the domain walls do support chiral edge states owing to opposite parities on either sides of the walls.

The strongly localized Berry curvatures:

$$\mathbf{F}_{\pm} = \mp \tau_z \frac{\nu_D^2 M}{2(M^2 + \nu_D^2) \delta k_x^2}$$

Here are the indices for the two valleys. We compute the winding integral to calculate the chern number in the K space in the region around the $K_{\pm}$ valleys. We obtain the quantized values of $\pm\pi$ in case of each band and hence the winding number.

The parity of Berry curvature also flips with the sign of the Parity breaking. Hence crossing over the domain wall from one valley lattice onto another results in a ± 1 difference between valley winding numbers. Due to the principle of smoothening of transition between two different topological invariants we observe the generation topologically protected edge states.

3.1.2 Results

We numerically calculate the band diagram for our membranous metasurface. The material used in the simulation was InGaAsP having a fixed thickness of 250 nm, the lattice constant was 460 nm and the triangulated holes had sides measuring 276 nm and 184 nm respectively.

The domain wall or simply the interface guides the chiral edge states and the small cavity hosts the 8 quantized modes within the bandgap region. These results agree with the analytical calculations from the tight binding model. The red and blue curves of the edge state dispersion correspond to the two possible configurations of the zigzag domain wall with smaller and larger holes respectively. Among the gap states, as a candidate for lasing, we specifically distinguish the triade cavity mode, which confines light at the three corners in-phase and exhibits a high estimated quality factor of about 4000. In its genesis, this mode is different from the corner states in quadrupolar topological insulators and distorted photonic lattices.

Despite the complexity of a real electromagnetic structure, the prerequisite of this bound-state formation at the valley-Hall domain wall is adequately captured within the nearest-neighbour tight-binding approximation. While the midgap behaviour in this system is governed by the Dirac-like Hamiltonian in two valleys, the deviation from the linear dispersion relation of the edge states closer to the upper band facilitates field localization at the corner defect.

The triade topological defect is formed at the domain wall corners. At the corners the chiral symmetry is broken, hence the parameter variations in the tight binding model shift the frequency of the state inside the gap. The corner mode however survives disorders weak compared to the gap size. We calculate that when the triangle side length is increased from seven to nine lattice constants, the shift in the corner mode frequency is only about 0.3 THz. The introduction of edge defects like a missing hole only perturbs the overall emission profile of the mode.

The fabrication is performed on a commercially available 250 nm-thick InGaAsP slab that incorporates three quantum wells. We proceed straight to electron beam lithography by coating using resist. This is followed by a dry etching process to

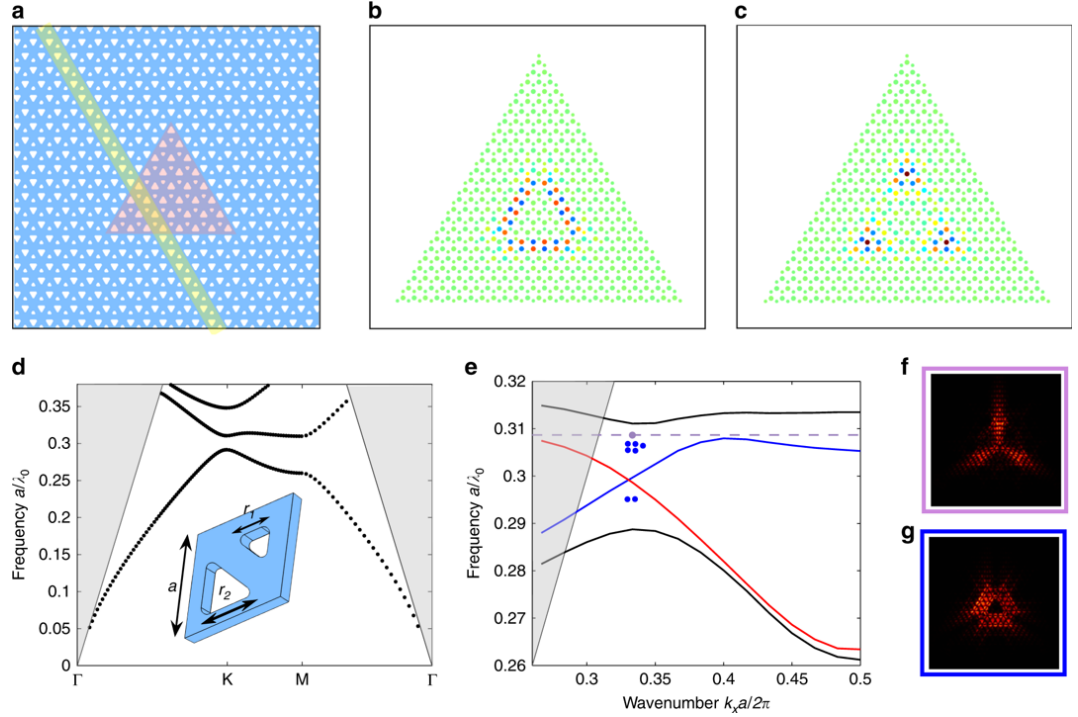


Figure 3.1: Valley-Hall topological photonic cavity. (a) A closed domain wall is created by inversion symmetry breaking in the dimerized graphene lattice and shaped in the form of an equilateral triangle (reddish area). The structure is implemented in a dielectric photonic crystal slab of a finite thickness. The shaded yellow stripe illustrates a supercell with a zigzag cut. (b), (c) Tight-binding model visualization of optical modes of the structure. (d) Numerically calculated bulk photonic band structure for TE-like modes. The grey shaded regions indicate the light cone. The inset shows the unit cell and dimensions of the structure: lattice period $a=460$ nm, sizes of triangulated holes $r_1 = 174$ nm, $r_2 = 269$ nm, and slab thickness fixed to 250 nm. A degeneracy is lifted, and the bandgap opens by dimerization between the first and second bands in the vicinity of the K point. (e) Projected bulk bands (black curves), spectra of edge states (red and blue lines) and cavity modes (dots). The dashed level marks the triade mode frequency. Characteristic distributions of the near-field intensity in the (f) triade and (g) edge cavity modes at resonant wavelengths of $1.49\ \mu\text{m}$ and $1.51\ \mu\text{m}$, respectively. Their simulated quality factors are (f) 4.3×10^3 and (g) 1.9×10^3

transfer the patterns on the slab resulting in the formation of a membranous structure (see Figure 3.2 for details). The fabrication parameters are very similar to the ones used in the theoretical calculations. The domain wall in particular has two large holes facing each other. We choose the side of our triangular topological cavity (domain wall) to be seven lattice constants or 3.22 microns.

In the optical diagnostics part, we pump the fabricated membranous metasurface using 980 nm laser having a pulse width of 8 ns and 10 KHz repetition rate. We ensure that the laser spot size on the sample is approximately the full sample size. We perform near infrared microscopy in reflection mode, just like in the case of our

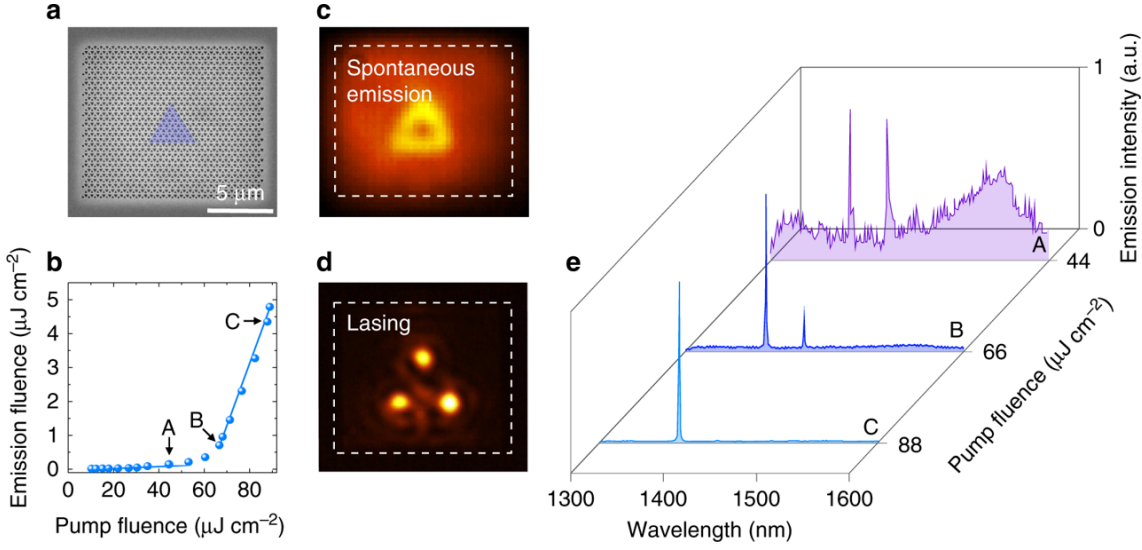


Figure 3.2: Lasing in topological cavities. (a) Scanning electron microscope image of the fabricated sample. The false-colour triangle marks the interior of the topological cavity. (b) Emission energy vs. the pump energy dependence showing a threshold transition to lasing. The sample is optically pumped at a 980 nm wavelength with 8 ns pulses at a 10 kHz repetition rate. Spatial distribution of emission for the pump intensity (c) below and (d) above the lasing threshold. e Emission spectra showing a transition to a narrow-linewidth lasing. In the lasing regime, the FWHM (full width at half maximum) does not exceed 2 nm and is resolution limited by the spectrometer

experiments with the topological zigzags in the previous chapter.

We study the relationship between the pump intensity and the emission intensity. The curve shows a clear lasing threshold like behaviour which indicates the pump intensity where the optical gain overpowers the losses. Further increment of pump power shows the saturation of emission. We also visualize the spatial mode profile of the corner triade modes. The modal profile also shows the case of spontaneous emission where the entire domain wall radiates luminescence via the edge states. Upon the increase of pump power, we observe that the emission from the corner triades dominates due to its high quality factor and small modal volume. We also perform spectroscopy at three different levels of pump intensity, showing the spontaneous emission, threshold case and stimulated emission respectively.

We proceed to the verification of coherence of emission from the corner states by studying its directionality diagram. For this purpose we perform back focal plane imaging of the lasing triade mode. Then we isolate two of the three emitting corners in the real space using an aperture to observe interference pattern in the Fourier space. The presence of fringe like pattern in the fourier space indicates some degree of coherence.

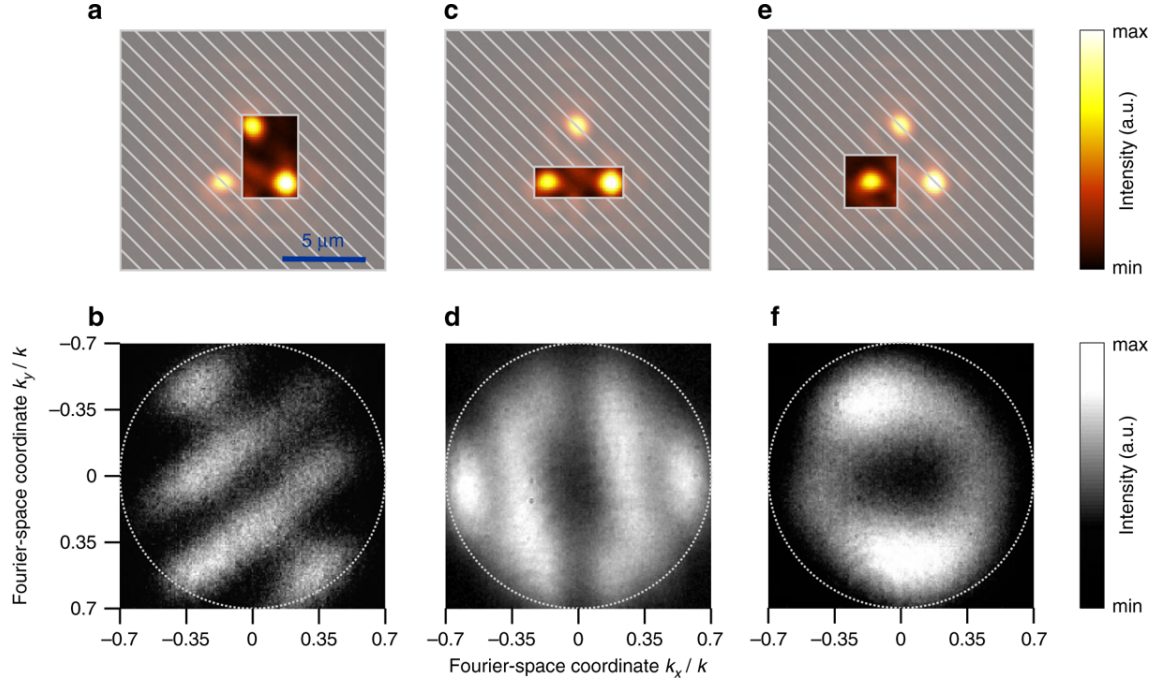


Figure 3.3: Coherence and far-field patterns of the laser emission. (a–d) Experimental study of the coherence of the emission observed in Fig. 3.2d. (a), (c) When two emission spots are isolated, (b), (d) interference fringes are observed in Fourier space. (e), (f) Real space emission distribution and Fourier-space emission directionality from an isolated cavity corner. An experimentally observed donut-like directionality pattern suggests an optical singularity hosted by the laser beam

Finally, we verify the directionality to establish lasing. For this purpose, in the back focal plane imaging measurement described previously, we isolate a single corner emitter instead of two. We observe a donut shape in the fourier space indicating directional emission. The donut like pattern in fourier space can be modelled by a particular distribution of magnetic dipoles in the corner.

We calculate the Poynting vector and flux for a single corner of the triade state. The simulated far field diagram indicates the topological charge of +1. This singularity index is found from the line pattern of the far-field polarization around the dark direction. We can also selectively excite the corners using directional evanescent coupling with a spin polarized light source.

In this way, the directional coupling stems from the chirality of the evanescent edge states excited by the source operating at frequencies within the minigap. As a result, sources of opposite handedness evanescently couple to and excite the opposite corners of the cavity. In turn, individual corner modes emit singular beams in the far field, as demonstrated.

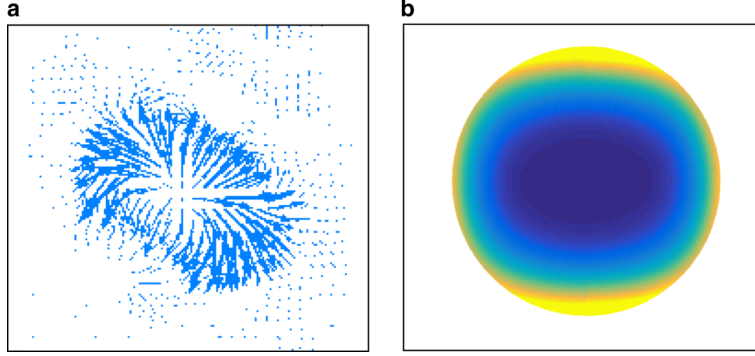


Figure 3.4: Singular emission from the domain wall corner. (a) Distribution of the Poynting vector, plotted as arrows around an isolated lasing cavity corner, indicates a singularity point. Numerically calculated with the 3D finite-element method for the triade eigenmode depicted in Fig. 3.1f. (b) Modelled far-field intensity pattern of the top corner

3.1.3 Conclusion

We demonstrate nanolasing from active two dimensional topological cavities at room temperature. The purpose of optical gain is served by the semiconductor quantum wells. We overcome the challenge of integration of active media with nanophotonic structure by using a membranous structure which actually patterns the active media itself. This solves any problems regarding coupling and incompatibility as it eliminates the need of another nanophotonic structure. Our demonstration shows that we can obtain lasing even with fabrication imperfections and disorders unlike the case in most nanophotonic lasers. We develop all dielectric platform desirable for application in integrated photonic applications.

3.2 Anapole Lasing

We developed nanolaser from active topological metasurfaces. We achieve robustness against disorder and fabrication imperfections; however, we also observe that the quality factor of mode confinement was not fully under the control of design parameters. This implied that the lasing threshold and the overall loss of the system couldn't be controlled either. Also, the phenomenon of lasing took place from a defect mode, where we cannot precisely control the lasing wavelength by geometrical or material changes.[175]

To overcome the aforementioned challenges, we must change one ingredient in our recipe of designing active nanophotonic devices which is the choice of nanophotonic structure. We have found from the previous section that the quantum wells are a great optical gain medium and can easily work with structures of quality factor above 1000. Now, what remains is to choose a resonant nanophotonic structure

whose quality factor and the resonant wavelength can be controlled by geometric parameter alteration.

The bound states in the continuum are one such nanophotonic tool that specialized in unique ways to trap light through weakly radiating modes.[176, 177] Unlike other resonances,[178] they are known to possess infinite lifetimes. Usually, the BICs are a result of peculiar destructive interferences between the leaky or outgoing waves. This peculiar destructive interference can be enforced by symmetries like the mirror symmetry, the time reversal symmetry and rotational symmetry or by special geometric conditions resulting due to periodicity of the lattice and consequential phase matching. Depending on the reason of destructive interference, we classify the BICs into two major types: Symmetry protected, and Geometry induced. The BICs unfortunately cannot be used for a lot of applications due to their imperviousness to loss. For all practical purposes, we use quasi-BICs which are structures that almost resemble BICs, and due to a slight perturbation there is a very weak leakage. The quasi BICs have finite lifetimes unlike the regular BICs.

Further research in the field of dielectric scattering revealed a new class of scattering modes, the Toroidal dipole modes. The toroidal dipole modes refer to the fields produced due to toroidal flow of currents in the dielectric nanoparticles.[179, 180] This discovery revealed that the toroidal dipole mode was actually very similar to the electric dipole mode in the far field. This gave birth to another method to ensure confinement of energy due to destructive interference of outgoing waves which was different to symmetry or geometry protection. This method involved the destructive interference of two scattering modes (electric dipole and the toroidal dipole)[181] of the same nanoparticle. This destructive interference resulted in the generation of a new ‘dark’ mode also known as the anapole mode which had only near field and no far field.[163]

The anapole state if combined with geometrical protection method can completely eradicate the presence of any other higher order mode,[182, 183, 184] hence giving the highest possible quality factor amongst all the bound states.

Owing to their strong field confinement, the anapoles offer exclusive opportunities to study strong field optics and nonlinear effects of nonperturbative regime.[185] Due to this ability, we can dictate various nonlinear processes [186, 187] like second harmonic or higher harmonic generation, sum frequency generation and four wave mixing.

Recent works have demonstrated engineering of anapole states into dielectric metasurfaces for demonstration of high harmonic generation, enhancement of Raman scattering and enhancement of plasmonic or excitonic coupling. From the perspec-

tive of futuristic technologies, switchable anapole states have also been demonstrated in phase changing materials like Ge-Sb-Te.[188, 189] Further theoretical research has also shown the evidence of higher order anapole states that are produced by the destructive interference of other complex scattering modes.

In this research, we take another shot at the demonstration of a working nanolaser. As mentioned in the beginning of this thesis, we increase the complexity of the problems we attempt to solve using active nanophotonics. In the previous nanolaser, we simply wanted to verify the possibility of nanolasing lasing through our quantum wells hence we chose an easier to fabricate and disorder robust topological system. This time however we base our nanolaser on the anapole states for precise wavelength and quality control. Unlike the topological counterpart, here the dimensions of every constituent nanoparticle are of critical importance, thereby posing a greater design challenge.

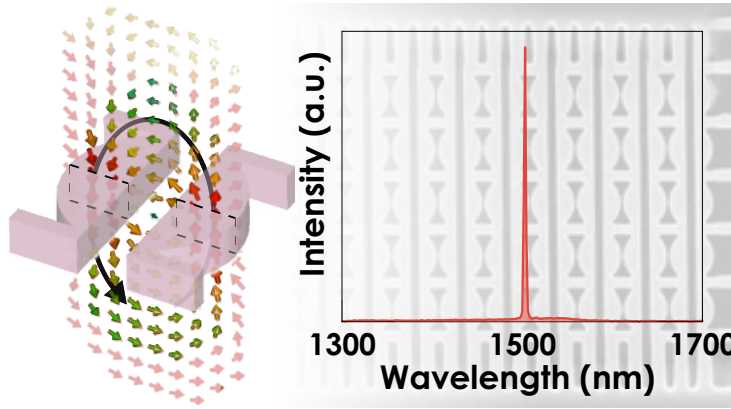


Figure 3.1: Concept image for anapole based nanolaser.

3.2.1 Results

To achieve efficient lasing action we design metasurfaces where individual nanoresonators take the shape of a flattened disk of diameter 925 nm and height 200 nm. The disks are made of InGaAsP which is also the host material for our quantum wells. The disks are so designed to host magnetic quadrupole mode and associated anapole modes. The magnetic quadrupole modes are chosen here instead of the toroidal dipoles because the toroidal dipole modes are only understood in cartesian decomposition not in spherical harmonic decomposition. Radiation leakage in side-way directions is prevented by arranging the disks in a square lattice having a period $P = 1010$ nm. This results in the leakage of radiation only in a particular set of directions also known as the diffraction modes. However, for the lower frequencies or higher wavelengths only the zeroth order mode or the normal direction is the

direction of propagation of the scattered outgoing light.

As discussed previously, the anapole mode is a result of destructive interference between the two interfering modes. Using a simple disk doesn't give the ability to tune the phase between these two modes. The ability to manipulate the mutual phase between the interfering modes is provided by the introduction of a diametrical split gap in the individual nanodisk, a strategy also used in [182]. Due to the existence of mirror symmetry in the metasurface plane, the design doesn't allow for any substrate near the nanodisk. For overcoming this design challenge, we added a very thin ladder (width is $L = 176$ nm) to provide mechanical support to the disks in the membranous structure. The design and schematics of the metasurface and the unit cell are shown in Figs. 3.2a and 3.2b.

We employ multipolar decompositions (described in chapter 1) to analyse the scattering from our designed metasurfaces. The dyadic Greens function is expanded into Taylor series, which helps in the consideration of several dominant multipoles. Magnetic quadrupole couples to the electric dipole via an electric toroidal contribution, which can be treated as a virtual magnetic current loop generated by a solenoid rolled into a ring [179, 183]. Due the lattice arrangement, the scattering in all directions other than the normal direction is nullified and only the anapole type eigenmodes that are dependent on destructive interference due to toroidal momentum patterns survive.

The magnetic quadrupole can be visualized as a pair of oscillating magnetic dipoles or as a sectional toroidal current circulating around the split gap. The scattering direction condition as discussed before does not allow for the magnetic dipolar field pattern, however the toroidal field pattern does coincide with the electric dipolar field pattern which allows normal scattering. Adjustment of the split gap leads to destructive interference between these two modes resulting in the formation of a high-Q dark anapole mode suitable for lasing. The mechanically supportive ladder has negligible influence because of absence of electric field penetration.

The metasurface quality factor is optimised by performing a parameter sweep for the split gap size. The energy density seemed to resemble a Lorentzian and the quality factor was calculated by fitting to the Lorentzian function. We plot the quality factor as a function of the split gap size and observe an inverse square dependence. $\propto (G - G_0)^2$, with the maximum at $G_0 = 228$ nm exceeds $Q = 4 \cdot 10^8$ in our calculations. This sharp rise in the Q factor is similar to that observed in supercavities and BICs. This observation indicates that the anapole mode we observe is dependent on the geometry of single nano resonator rather than any geometric effect or symmetry mismatch.

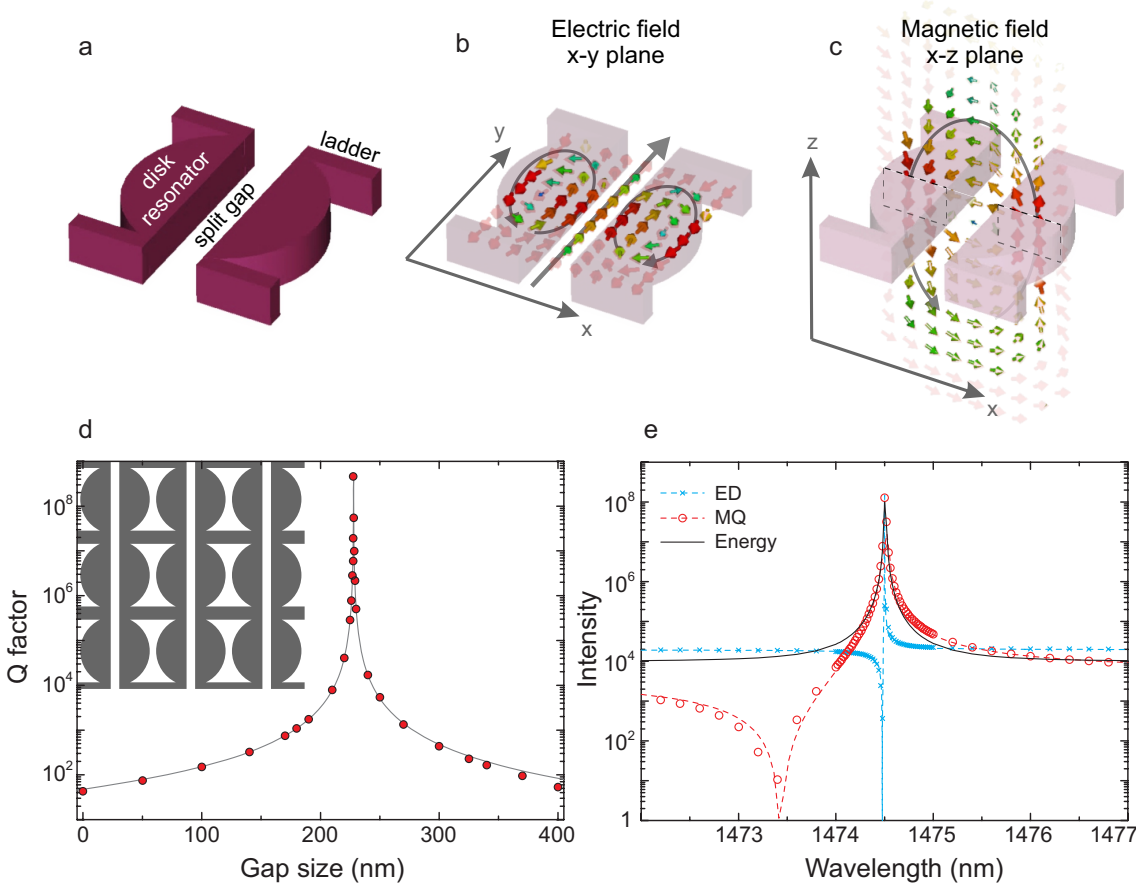


Figure 3.2: Design of the anapole-empowered metasurface. (a) Schematic of the metasurface unit cell composed of a high-index disk supporting an anapole-type mode. (b,c) Numerically calculated electric and magnetic field cross-sections in the vicinity of the anapole resonance for the split gap size of 230 nm. Black arrows are guides for an eye. The electric field distribution shows an electric dipole-like oscillation in the gap as well as a pair of counter-rotating vortices in the bulk of the disk resonator within the x-y plane (the metasurface plane). Magnetic field demonstrates a loop in the x-z plane. (d) Simulated Q factor of the anapole-type mode as a function of the split gap size. The inset shows a schematic of top view of the metasurface. (e) Multipole decomposition around the anapole-type mode and energy stored by the system. Spectra of the electric dipole moment, the magnetic quadrupole moment, and the stored energy are indicated by blue crosses, red open circles, and black solid curve, respectively. The gap size is 230 nm.

The multipolar decomposition performed for the fields generated in the case where the split gap size is 230 nm is shown in the figure 3.2e. The contributions of electric dipole and magnetic quadrupole are revealed to be asymmetric fano resonances [178]. The combination of these sharp fano resonances leads to a sharper combined resonance or simply the anapole resonance.

The metasurface is fabricated using the same recipe (Figure 3.3) as in the topological lasers. The sample is fabricated out of commercially available quantum wells (same

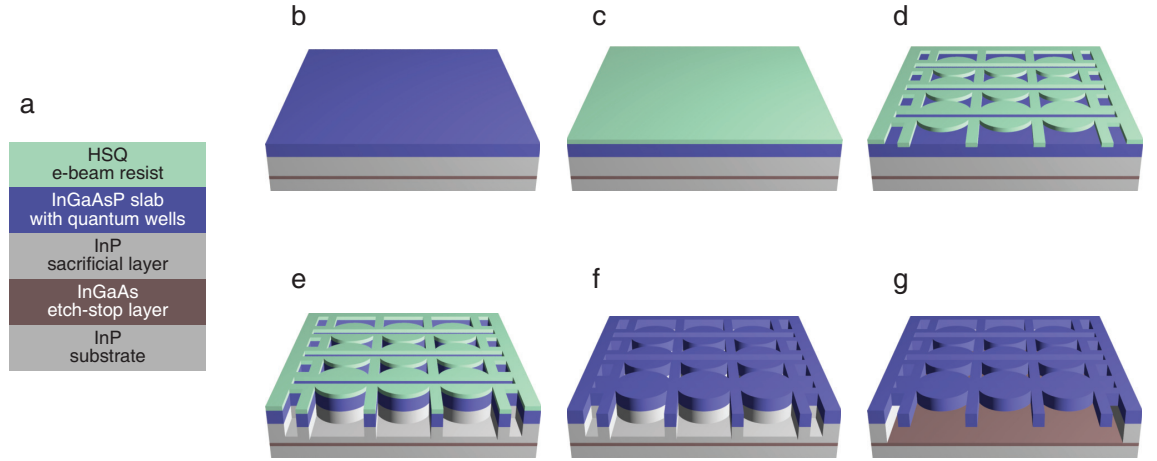


Figure 3.3: Sample fabrication steps. (a) Schematic of the layer sequence. (b) Initial sample grown by molecular-beam epitaxy. (c) HSQ resist layer coated on top of the sample. (d) Nanopatterns defined by electron-beam lithography. (e) Dry etching. (f) Removal of HSQ by O₂ plasma. (g) Wet etching of the sacrificial layer.

as the ones used in topological lasers). On the top of the indium phosphide substrate a thin InGaAs layer of 100 nm was grown covered by 800 nm thick InP sacrificial layer. An InGaAsP slab includes three quantum wells with an emission around 0.82 eV (1.5 μm). A thin hydrogen silses quioxane (HSQ) layer was spin-coated on the top and was used as a resist mask patterned by electron-beam lithography. Chemically-assisted ion beam etching was used to produce resonators from InGaAsP slab. After that the HSQ mask was removed by oxygen plasma and diluted chlorhydric acid was used to wet-etch the sacrificial InP layer. As a result the free-standing metasurfaces composed of active resonators of desired geometry were produced (see SEM image in Fig. 3.4a). By using this procedure we fabricated a number of samples with different geometry parameters and number of unit cells [ranging from 75 (15 by 5) to 225 (15 by 15) disks].

The lasing experiments are exactly the same as the ones performed in the case of topological laser using the same microscopy and pumping conditions. The threshold was calculated to be 10 $\mu\text{J}/\text{cm}^2$ for the optimal anapole metasurface. At pumps of lower fluence, we observe a broadband photoluminescence in the near infrared band around 1.5 microns. In the lasing regime (Figure 3.4b,d), we see the presence of a sharp 1503 nm line having 2.2 nm full width at half maximum. We plot the lasing threshold curve in Fig. 3.4c.

Finally, we measure the coherence of the system using backfocal plane imaging just like in case of topological laser. Here also two areas of the emitting metasurface were isolated using an aperture. The interference pattern was observed in the fourier

space (Figure 3.4e), cementing the evidence for the quality of coherence.

To establish the dependence of lasing parameters and the quality factor on geometrical parameters of the individual nanoresonator, we compared the lasing characteristics for all the samples. We used the following equation to calculate the mean square geometric deviation from the optimal sample. $\xi = \frac{\Delta D}{|\Delta D|} \cdot \frac{\Delta L}{|\Delta L|} \cdot \frac{\Delta G}{|\Delta G|} \sqrt{(\Delta D^2 + \Delta L^2 + \Delta G^2)}$, where ΔD is the deviation of the disk diameter, ΔL is the deviation of the ladder width and ΔG is the deviation of the split-gap size. We use curve fitting to calculate lasing thresholds for all the lasing samples. The lasing threshold is qualitatively a very accurate experimental indicator for the quality factor, as the greater is the quality factor the lesser is the lasing threshold. The relationship between lasing thresholds and the geometrical deviation is presented in the Fig. 3.4f.

The threshold spectra can be used to measure the spectral width of the lasing mode and to calculate the line width of the laser radiation via the the Schawlow-Townes equation [190]: $\Delta\omega_{\text{laser}} \propto P_{\text{out}}/Q^2$, where $\Delta\omega_{\text{laser}}$ is the FWHM of the laser line, and P_{out} is the laser output power. This relation shows that the laser spectral width is narrower than the spectral width of the cavity mode and additionally depends on the generated power. The line narrowing process is limited due to the saturation of optical gain.

Due to $\Delta\omega_{\text{laser}} \propto (Q^{-2})$ and $Q \propto \xi^{-2}$, we expect the dependence $\Delta\omega_{\text{laser}} \propto \xi^4$. A rough theoretical estimate of the quality factor is as follows: $Q = Q_{\text{rad}}^{-1} + Q_{\text{sca}}^{-1}$.

The experimentally calculated Q factor in this case is $2 \cdot 10^2$. We observe that there is strong dependence of Q_{rad} on the gap size. Hence establishing the strong dependence of lasing parameters on the geometry unlike the case of topological laser.

3.2.2 Conclusion

In the case of a single dielectric disc that supports the anapole mode, the radiation suppression is sustained due to the existence of external field. In case of the nanodisk array, the anapole state becomes an eigenmode of the system. As a result of the lattice arrangement, two modes a BIC and the anapole mode are locked as high Q modes leading to lasing.

We design another laser, using the same active material as the topological laser, but having a different modus operandi. The topological laser on one hand is robust against disorders and fabrication imperfections but there is no control over the Q factor and the lasing wavelengths, the anapole laser on the other hand is strongly dependent on geometry and needs perfect fabrication but provides precise control over

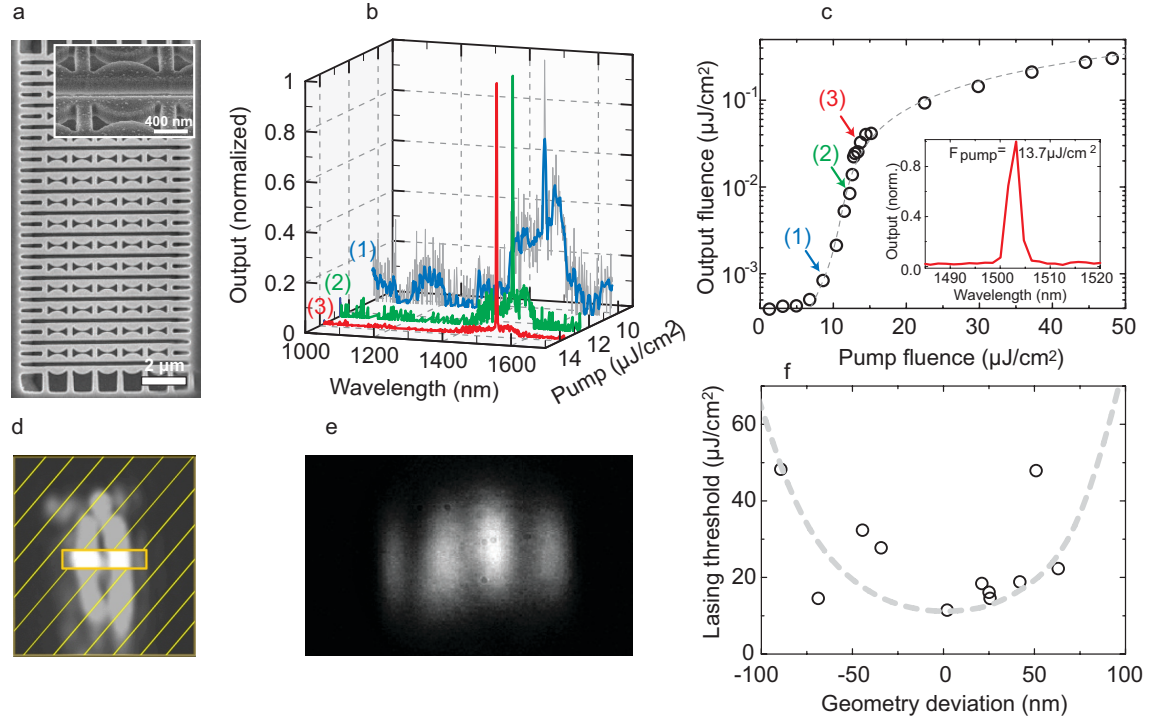


Figure 3.4: Lasing empowered by the anapole metasurface. (a) SEM image of the fabricated metasurface. (Cross-section view in the inset) (b) Evolution of the normalized output power as a function of the pump fluence and wavelength. For the spectrum (1), the gray curve is a raw spectral data, and the blue curve is post-processed plot smoothed with quadratic Savitzky-Golay averaging. The blue curve therefore serves as a guide for an eye. (c) Measured output fluence at lasing wavelength as a function of the pump fluence. Gray dashed curve is a theoretical result. The inset demonstrates the lasing peak with a wavelength of 1503 nm and a linewidth of about 2.2 nm. Numbers (1) to (3) denote the spectra shown in (b). (d) Far field radiation pattern of the anapole nanolaser. The small yellow box indicates aperture window isolating two bright spots for coherence measurement. (e) Interference pattern in Fourier space obtained by back-focal plane imaging of two spots isolated in (d) confirming coherence of lasing radiation from the anapole. (f) Dependence of the lasing threshold on the geometry deviation from the parameters of the best sample. Gray dashed curve is a theoretical result obtained from Eqs. (2) and (3).

lasing parameters and the Q factor. We develop another active nanophotonic device demonstrating intelligent combination of active media and nanophotonic structures. This research will develop further interest in solving more complex problems of integrated photonics using active nanophotonics.

Tunable and Nonreciprocal metasurfaces

After the demonstration of basic optoelectronic functionalities like lasing and luminescence, we move forward to more difficult problem statements like achieving active control over nanophotonic devices.[149] Active control here essentially means that the device can be controlled by external means or stimuli and has two or more distinct functionalities.[191] Active devices play a vital role in logic, memory, and communications in applications like routers, logic gates and multiplexers.

Active devices depend on external stimuli for switching action. In case of electronic devices, the external stimulus is provided by a bias current or a bias voltage. For optical and nanophotonic devices, the stimuli can be either electrical, thermal, mechanical, or even optical. In nanophotonic devices, mechanical and electrical stimuli are extremely difficult to implement owing to the fabrication intricacies at the nanoscale. This leaves us with the options of thermal or optical stimuli only. In nanophotonic devices, the capability to switch actively can be added by the introduction of active media that can undergo a transition under the application of stimuli. Such an active media can be a phase change material or a highly nonlinear material.[192, 193] The phase change material can respond to almost any kind of stimulus (optical, thermal, electrical, or mechanical) and a highly nonlinear material can be activated upon the application of strong optical fields.[194, 195]

The field of nonreciprocal nanophotonic devices however is somewhat close to Switchable devices yet different. Here, the impetus is on breaking the Lorentz reciprocity theorem or the time reversal symmetry. This is done by using external stimulus like magnetic fields or acoustic signals for temporal variations or by the use of nonlinear effects. However, at the nanoscale only the use of nonlinearity seems to be the feasible option because of the size constraints. This need for nonlinearity can be catered using phase change materials or using highly nonlinear materials just like in case of Switchable devices.

Because of similar design requirements, both the Switchable and nonreciprocal optical devices require the use of similar active materials: the phase change materials and highly nonlinear materials. The phase change materials like vanadium dioxide are capable of a sudden transition in their lattice structure which also leads to a sudden change in the refractive index values. The nonlinear materials like Indium Tin Oxide (ITO) on the other hand can provide a pump intensity dependent change in the refractive index values. Both however have a ‘nonlinear’ change in their respective refractive indices and a significant absorption loss. The absorption loss is so high that it can kill almost any near or far field scattering feature like energy hotspots and the multipolar resonances. Just like friction in mechanics, the absorption in these materials is also a necessary evil. If the absorption is too low or insignificant, it will be impossible to absorb energy needed for the phase transition or the nonlinear change of refractive index. The constraint of absorption loss in these materials makes the design of nanophotonic devices based on these materials extremely challenging, infact it is prudent to say that the design of Switchable and nonreciprocal optical devices is the engineering of absorption loss.

The key technique used to address this challenge is engineering the volume and the position of the active materials. In the design of the nanostructure, it is often ensured that the active material sits close to the field, but not so close that it completely eats up the near fields. An example of this principle is demonstrated by the optical amplitude modulators designed by Howes et al [196]. Where vanadium dioxide sits on top of individual dielectric resonators as caps to ensure that only a portion of scattered fields is used up in the phase transition. Other techniques include doping and alloying with other materials to obtain the desired absorption parameters. This however isn’t easy as it involves heavy research into chemistry and materials science. Hence, we stick to the former approach for the design of our devices.

4.1 The problem statement

In the next section of this chapter, we approach the problem of designing a Switchable or reconfigurable meta surface to enable dual functionality from the same platform. Reconfigurable or Switchable nanophotonics usually deals with tunability in basic properties like transmission, reflection, emission, or absorption.[192, 193] This ability is achieved by imparting reversible changes in the device materials in real time. At the millimeter and the microscale, liquid crystals and electro optic cells have been used to achieve this effect. At the nanoscale, Phase change materials and highly nonlinear materials [194, 195] that are capable of imparting reversible

changes via change in their refractive index are often bistable in nature and capable of generating hysteresis like transformers.

Majorly, there are three types of reconfigurable nanophotonic devices namely: Optical modulators, Optical Switches and Tunable filters [192, 193, 194, 195]. The optical modulators can alter the phase, amplitude or polarization of the incident light in different ways like tunable spectra and spatial light modulation. Potentially they can be used in optical signal processing and information encoding. The Optical switches can perform an on and off type of operation as well as directing the signal in one of the multiple available paths to enable optical routing, on demand signal processing and multiplexing of signals. Finally, tunable filters selectively allow or block certain wavelengths to provide dynamic spectral shaping in optics. Basically, the three types on reconfigurable devices mentioned above are classifications on the basis of domain of their functionality. The optical modulators have purely qualitative functionality, optical switches perform spatial operations to direct light on a route or block it off completely and the tunable filters perform spectral operations.

In the first section, we try to approach the problems of developing a nanoscale optical modulator and a nanoscale tunable filter. We rely on the phase change of vanadium dioxide to provide the switching action in our designs.[197]

The subsequent sections deal with a more complicated problem: the design and demonstration of nonreciprocal nanophotonic devices. Nonreciprocal optical devices as the name suggests can show asymmetric propagation properties like asymmetric transmission, asymmetric phase generation or asymmetric spatial distributions. These devices break the Lorentz reciprocity condition to demonstrate ornate functionalities like nonreciprocal phase shifting, polarization tuning and unidirectional propagation of light.

The effect of optical nonreciprocity is achieved by breaking of time reversal symmetry. There are three pathways for breaking of time reversal symmetry: magnetic fields, time varying structures (acoustics) and nonlinearity. The application of magnetic field to generate faraday rotation requires the presence of external magnets which occupy the minimum size of a few millimeters. Acousto-optic structures need the application of sound as a modulating force, implying that the dimensions of the device must be of the order of the shortest wavelength of ultrasound usable i.e. a few microns. Optical nonlinearity on the other hand is an intrinsic material property triggered only by incident light intensity, not the wavelength. Hence it doesn't depend so much on the material dimensions and therefore is the only feasible option at the nanoscale.

By nonlinearity we mean an optical transition preferably in the refractive index

that is nonlinear in nature. Phase change materials like Vanadium Dioxide and highly nonlinear materials like ITO meet these requirements. Vanadium Dioxide changes its phase at 68 degrees Celsius and suddenly behaves like a metal instead of the original dielectric. This phase change results in a step function like change in refractive index. Similarly, ITO in its near epsilon zero region shows very sharp enhancement of Kerr coefficient and nonlinear absorption. Upon the exposure to high intensity of light, the extremely high Kerr coefficient helps in generating a sharp change in the values of refractive index like a delta function.

In our research, we try to demonstrate unidirectional transmission or simply non-reciprocity in transmission of light. This effect is also known as optical isolation in the field of photonics. It is similar in nature to the function of a diode in electronics and the function of valve in fluid mechanics. At the nanoscale, optical isolation can potentially open up new possibilities for all optical logic in integrated photonics and optical computing. We try to develop nonreciprocal metasurfaces based on Vanadium Dioxide as well as based on ITO. Both of these metasurfaces rely on the asymmetric distribution of near fields upon excitation from either direction. Due to this, the phase transition or simply the change of refractive index occurs at different intensities resulting in a nonreciprocal regime.

4.2 Switchable Metasurface

Vanadium Dioxide is known for its unique metal to insulator phase transition that occurs at 68 degrees celsius. This first order phase transition in vanadium dioxide is caused by its inherent strong correlation between the lattice and electronic degrees of freedom.[197, 198] When this transition temperature is achieved, the electron phonon coupling destabilizes the conducting state. This results in the generation of localized electronic states and an eventual bandgap generation. This generation of bandgap evolves the conductor to an insulator.

Structurally, the phase transition that occurs at 68°C is nothing but a change in the lattice arrangement from one bravais lattice to another. At room temperatures, Vanadium Dioxide has a monoclinic structure, where we see insulating or dielectric behaviour. In this situation, Vanadium exists in +4 oxidation state with an electronic configuration of $[Ar]3d^1$. Implying that a single electron can occupy the d orbitals leading to degeneracy. The degeneracy is lifted by lowering the overall system energy by the means of structural distortion also known as the Jahn-Teller effect [199]. Due to this effect, the octahedral structure of VO_2 is distorted and non-centrosymmetric.

However, as the temperature is increased, observe a rearrangement into rutile structure which has a tetragonal shape and centrosymmetric arrangement of vanadium atoms.

The standout features of this phase transition or in other words the reason for choosing Vanadium Dioxide over other phase change materials are the low activation temperature and the crystalline-to-crystalline nature.[200, 201] The low activation temperature makes the transition more easily accessible, and it becomes an overall faster process as it nearly eliminates the requirement of complex temperature controls. And the crystalline-to-crystalline nature makes the transition highly repeatable for an active nanophotonic device.[202, 203] Which is ensured due to the fact this way there is no creation of defects at the interfaces due to the lack of crystallinity.

From the point of view of photonics, the large change in refractive index due to vanadium dioxide phase transition is the key factor for the development of multiple reconfigurable and memory based devices.[204, 205, 206] So far works on VO_2 based metasurfaces have reported the use of metallic antennas. The efficiencies of these systems are however quite questionable due to the ohmic losses. The dielectric metasurfaces offer a way around this challenge and are the next generation of reconfigurable nanophotonics.[207, 208] As promising as reconfigurable dielectric metasurfaces appear, the nanofabrication techniques for realizing them remain premature. Howes et al had recently demonstrated bilayer Si-VO_2 disks to realize tunable metasurface.[196] The design parameters were emulating those of Silicon based Huygens metasurfaces and then the parameters were optimized using a parameter sweep. This approach is very similar to our approach for the design of active nanophotonic device.

Instead of a bilayer Si-VO_2 , we use a thin underlying film of VO_2 with Si nano resonators on the top to control the amount of field sitting inside Vanadium dioxide. The nanophotonic structure we choose for the design of our reconfigurable optical modulator and tunable filter had to ensure that the individual scattering modes are altered but not destroyed.[209, 210] A structure with sharp resonances is almost useless because the sharp resonance will be almost nullified by the material losses. But a structure having a component of both electric dipole and magnetic dipole resonances is ideal for this purpose as the electric dipole mode mostly sits on top of the nanoresonator and hence will not be depleted due to material loss. This fact makes the implementation of a Huygens metasurface based design an ideal nanophotonic structure for our reconfigurable metasurface.

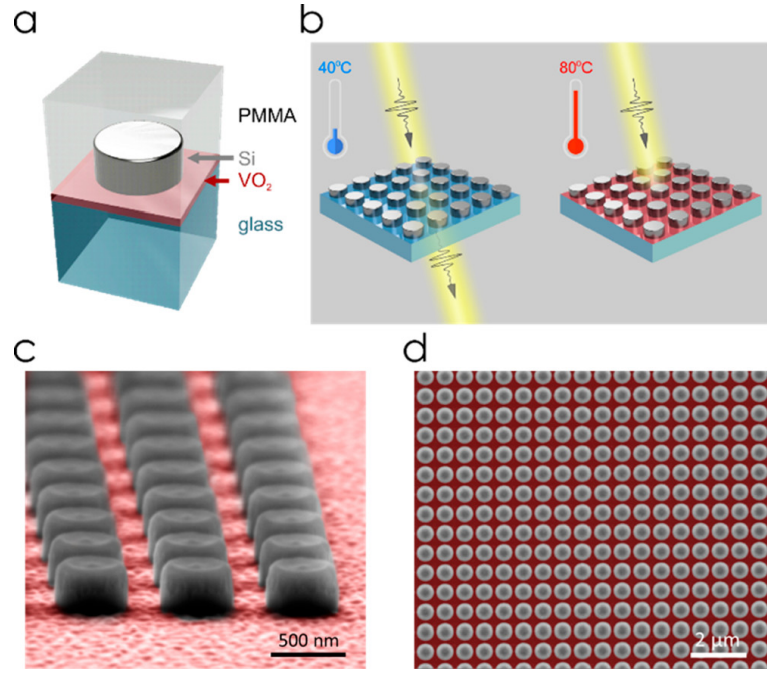


Figure 4.1: Tunable dielectric metasurfaces. (a) Design concept: a Silicon meta-atom is embedded into glass/PMMA surrounding with a buried thin VO₂ layer of 25 nm. Si disk height: 240 nm, disk radius: 255 nm, square lattice period: 700 nm, PMMA layer thickness: 500 nm. (b) Functionality of the metasurface featuring tunable extinction with temperature variation. (c, d) Electron microscope images of the fabricated Si-VO₂ metasurface prior to the PMMA coating (VO₂ layer in false red color).

4.2.1 Results

We proceed by applying the idea of coherent perfect absorption to account for the overall absorption loss of VO₂ film.

Coherent Perfect Absorption is observed when two or more coherent waves interfere in a way that transmittance and reflectance are eliminated. In other words, it means that all the incident energy is channelized into absorption. Mathematically, it implies that the product of scattering matrix and the input beam vector must be a zero vector. This indeed can be achieved with the right choice of amplitude, phase and polarization.

In metasurfaces, this phenomenon is achieved by mechanisms like impedance matching, temporal coupling, parity time symmetry, bound states in the continuum and the critical coupling condition [211]. Amongst these, the underpinning mechanism for our metasurface is the critical coupling condition. The critical coupling condition allows for coherent perfect absorption provided the material absorption losses are equal to the radiative losses. This allows us to exploit the Switchable absorption of VO₂ layer and switch it to enable or disable perfect absorption. Mathematically, the

critical coupling condition gives us two equations for transmittance and reflectance (see equations 4.1,4.2) going down to zero in a special case also known as the Kerker conditions.

With these two conditions serving as a quantitative check for the radiation and absorption, we can verify that the tunability of optical characteristics is an interplay of the two.

The original design is intended to cater the wavelength range 1530-1565 nm also known as the C band wavelengths of the optical fiber communication. The individual nanoparticles are so designed to host Electric Dipole and the Magnetic Dipole scattering modes. The spectral range and modulation levels dictate the geometric parameters. As a rule of thumb, we scale all the geometrical parameters according to the wavelength range and modulation levels are adjusted using a parameter space. VO₂ layer has a two-fold purpose, the first is causing sudden change of refractive index and the second is manipulation of absorption. The phase of VO₂ layer serves as the deciding factor for the interaction and the profile of the interfering electric dipole and magnetic dipole modes. The electric dipole modes undergo frequency shifting with about 50 percent attenuation and the magnetic dipoles don't undergo as high frequency shift but get more significantly attenuated. For the development of an optical modulator, we use Huygen's metasurface having a coincidence of Electric dipole and Magnetic dipole resonances. And for the development of spectrally tunable near perfect absorber we employ a dominant electric dipole resonance and a weaker magnetic dipole moment.

The Huygens metasurface based on Coherent perfect absorption relies on the accurate interplay of ED and MD modes. A high index material like Silicon is used to make the nanodisks that host these modes. The parameters of this disk are then tuned to achieve overlapping ED and MD modes to achieve coherent perfect absorption. This condition is also known as the Kerker condition. The spectral performance of Huygens' metasurfaces can be described phenomenologically within an approximation of the dipoles as Lorentzian oscillators.

Complete or 100 percent transparency requires the perfect overlap of resonance frequencies $\omega_{ED} = \omega_{MD}$ and the equity of radiative losses $\gamma_{ED} = \gamma_{MD}$. Both the qualitative and quantitative natures of the dipolar resonances are determined by the geometry of the individual nanoparticle and the relative refractive index of the disk. Integration of phase changing Vanadium dioxide in the structure would hence alter both the relative refractive index and the overall characteristics of the transparency condition.

A similar simplistic model may be used to describe absorption in such systems once

absorption losses δ_{ED} and δ_{MD} are incorporated in the Lorentz oscillator model, and transmission correspondingly reads

$$t = 1 + \frac{2i\gamma_{ED}\omega}{\omega_{ED}^2 - \omega^2 - 2i(\gamma_{ED} + \delta_{ED})\omega} + \frac{2i\gamma_{MD}\omega}{\omega_{MD}^2 - \omega^2 - 2i(\gamma_{MD} + \delta_{MD})\omega} \quad (4.1)$$

$$r = \frac{2i\gamma_{ED}\omega}{\omega_{ED}^2 - \omega^2 - 2i(\gamma_{ED} + \delta_{ED})\omega} - \frac{2i\gamma_{MD}\omega}{\omega_{MD}^2 - \omega^2 - 2i(\gamma_{MD} + \delta_{MD})\omega} \quad (4.2)$$

The above equations are inclusive of material absorption. The fractions containing the subscript ED represent electric dipolar contributions and those containing the subscript MD stand for magnetic dipolar contributions. Substituting the conditions where the ED and MD frequencies, radiative losses and material absorption losses are equal, we find that transmittance and reflectance go down to zero.

In the transmittance equation (forward direction), both the electric dipole and magnetic dipole components retain their signs which means that they don't undergo a phase change. However, in the reflection case (backward direction) the electric dipole component retains its sign (no phase change) but the magnetic dipole component becomes negative (phase change upon reflection).

Disk parameters in the range of 200 to 650 nm (see Figure 2(a,b)) are scanned and the multipolar decomposition of their respective optical responses is performed. We discover that the ED and MD modes overlap for the 510 nm diameter (see Figure 2(e)). Figure 2(f:i) show examples of normalized electric field distribution inside the samples for both insulating and metallic phases of the VO₂ layer for a disk diameter of 550 nm at several different wavelengths ((f) 1410 nm, (g) 1470 nm, (h) 1380 nm, and (i) 1450 nm), corresponding to maxima of the electric and the magnetic dipole contributions, respectively.

VO₂ is a challenging material to integrate into patterned heterostructures, owing to the complexity of high deposition temperatures necessary to form the phase and to the multiple phases that are present in the VO system. This requires a careful optimization of the device processing parameters to ensure film quality and preserving the desirable insulator metal transition properties after the fabrication steps are completed.

The fabrication of the metasurface is performed by depositing a 25 nm thin VO₂ layer atop a quartz substrate by the means of magnetron sputtering from V₂O₅ target in the partial pressure of Ar/O₂. In the deposition process, the chamber is kept at 5 mTorr with a flow rate of 49 sccm Ar and 1 sccm 10 percent Ar balanced O₂. The oxygen partial pressure during deposition was therefore 2.67×10^{-4}

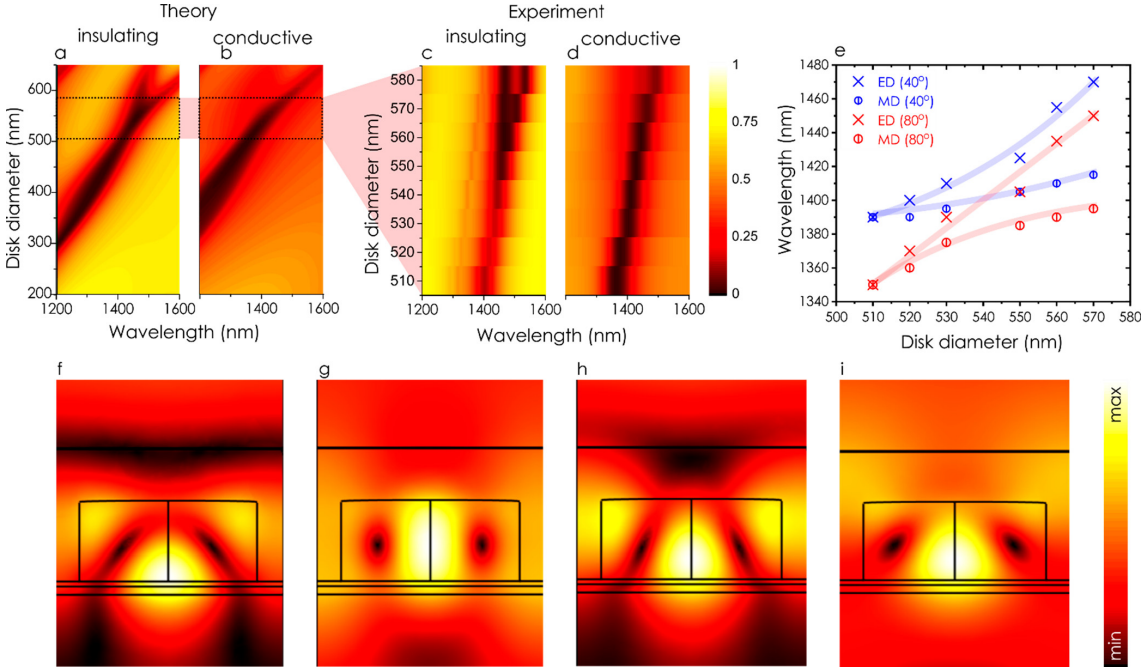


Figure 4.2: Experimental and theoretical optimization of the metasurfaces. (a, b) Calculated and (c, d) experimentally measured transmission spectra of the metasurfaces with varying disk diameter for the (a, c) insulating and (b, d) conductive phases of the VO₂ layer. (e) Theoretically calculated maxima of the electric dipole and the magnetic dipole Mie resonances for metasurfaces with varying disk diameters in (blue) insulating and (red) conductive phases. Lines are guides for the eye. (f–i) Normalized electric field distribution in the cross sections of the hybrid a-Si/VO₂ nanodisks 550 nm in diameter when VO₂ is insulating (f, g) and metallic (h, i) for wavelengths 1410 nm (f) and 1380 nm (h) at the magnetic dipole maximum as per (e) and for wavelengths 1470 nm (g) and 1450 nm (i) at the electric dipole maximum as per (e).

Pa. As the utilized substrate (quartz) is not lattice matched to VO₂, the deposition temperature has to be carefully adjusted in order to optimize the resulting crystallization and insulator to metal transition property of VO₂. The optimized deposition temperature found and used throughout this work was 750 °C. To protect the phase transitioning ability of VO₂ from the forthcoming steps, we deposit a 13 nm thick SiO₂ layer on top of the VO₂ layer. Next, we deposit a 240 nm thick layer of amorphous Silicon (a Si) by plasma enhanced chemical vapor deposition using a SiH₄ precursor at a temperature of 300 °C. A hydrogen silsesquioxane (HSQ) resist is then spun on the sample and patterned using e beam lithography. After development in tetramethylammonium hydroxide (TMAH), the HSQ resist is used as a hard mask to transfer the patterns into a Si via ICP RIE dry etching with Cl₂/Ar. Finally, a 500 nm thick layer of PMMA is spun on the sample. We systematically measured the optical properties of VO₂ by temperature-controlled ellipsometry after

each technological process to verify that the insulator to metal properties remained unaffected. Each of the various fabrication steps listed above was optimized through a trial-and-error approach in order to preserve a fully functional VO_2 thin film in the final devices.

The optical properties of Vanadium Dioxide based reconfigurable metasurface are using a heat cell that allows temperature control of $\pm 0.1^\circ\text{C}$. We then focus a broad-band light source on the devices using a 50x objective lens with 0.42 NA and the transmitted light is collected using the same objective lens using an iris diaphragm at the backfocal plane to contain the angles of collection. We then perform spectrometry of the transmitted light. The setup is calibrated using an unfabricated portion of the sample whose reflection and transmission are already known. The extinction is calculated as $E=1-T$. To get the idea of hysteresis effect, the measurements are performed over both the ascending and the descending cycles. We also get an idea of the repeatability as we see that the sample performance remains unaltered even after 10 heating-cooling cycles.

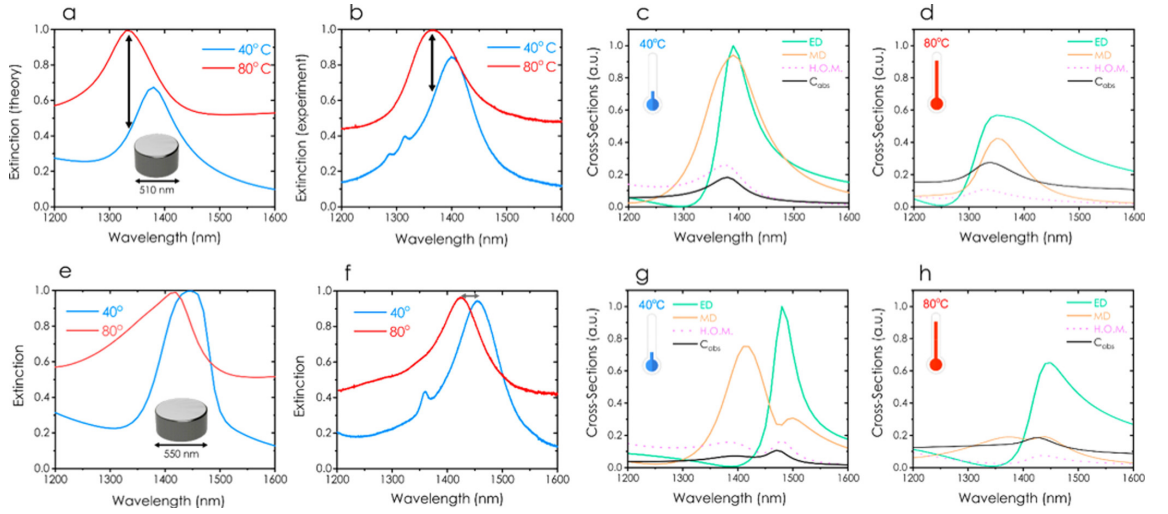


Figure 4.3: Experimental vs theoretical results on tunability of the Si- VO_2 metasurface. (a-d) Theoretical extinction (a), experimental extinction (b), and absorption and scattering characteristics (c, d) of optical resonances for disks with a diameter of 510 nm in the two states of VO_2 : insulating (c) and metal (d). The scattering component of the extinction is presented as a decomposition of Mie multipoles. (e-h) Theoretical extinction (e), experimental extinction (f), and absorption and scattering (g, h) of optical resonances for disks with a diameter of 550 nm in the two states of VO_2 : insulating (g) and metal (h). H.O.M.: higher-order multipoles.

Upon observing the behaviour of all the fabricated samples, we choose to focus particularly on two: one with disk diameter of 510 nm and the other with disk diameter of 550 nm. For the diameter of 510 nm, we observe amplitude modulation

and at 550 nm, we observe spectral modulation. In Figure 3(a,b) we compare both the theoretically evaluated and experimentally measured extinction spectra for both the conducting and insulating phases for the samples having disk diameter of 510 nm. We note that at the optimal wavelength of 1370 nm, we observe a strong extinction contrast. Here the extinction reaches almost 1 implying near zero transmission. This meant that the contrast in transmission could be as high as 2 orders of magnitude (Fig 4a).

The corresponding data for the samples with disk diameter 550 nm is visualized in Fig 3ef. We see here that there is a high extinction regime in both hot and cold states of the sample. However in the hot state, the spectrum is blue shifted from 1450 nm to 1420 nm. We also checked the unpatterned areas and found no special features in transmission or extinction that were effected by the phase transition of VO_2 . Thereby confirming that the observed effects are due to interaction between the nanostructures and the active media (VO_2). We also observe that there is a very good agreement between theoretical and experimental measurements of spectra.

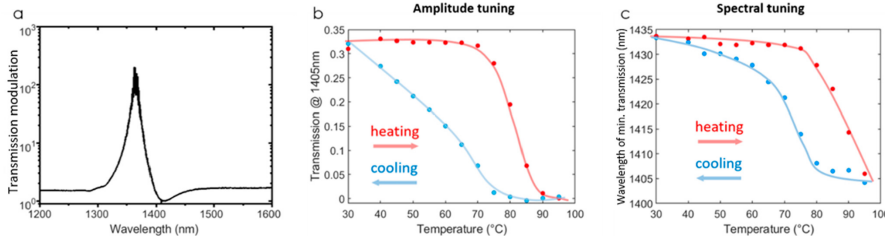


Figure 4.4: Experimental large transmission modulation, spectral tuning, and hysteretic memory effect of the Si- VO_2 metasurface. (a) Two orders of magnitude experimentally measured transmission contrast ($T_{25^\circ\text{C}}/T_{100^\circ\text{C}}$) induced by the VO_2 insulator to metal transition on the metasurface with a disk diameter of 510 nm. (b) Reversible tuning of the amplitude of transmission featuring a hysteresis behavior of the same device. (c) Reversible spectral tuning of the metasurface transmission showing a continuously reconfigurable near-perfect absorber (metasurface with a disk diameter of 550 nm)

The observed optical effects in our reconfigurable metasurfaces are because of the redistribution of energy in the multipoles of the nanodisk and increased or decreased absorption of the VO_2 film (Figure 3(c,d) and (g,h)). In case of amplitude modulation (samples with disk size 510 nm), the Kerker conditions are met in the cold or insulating phase, and hence the device operates like a Huygens metasurface even though the exact resonance conditions aren't satisfied. After the phase transition into metallic or the hot state, the ED and MD amplitudes are highly reduced and the overlap is killed. This violates the Kerker conditions, but due to increased ab-

sorption, the critical coupling conditions for coherent absorption are met where the radiative loss and the absorptive loss are equal. This leads to near zero transmission in the metallic state, and hence we observe amplitude modulation.

In case of spectral modulation (samples with disk size 550 nm), we already meet the conditions for Coherent perfect absorber in the insulating state resulting in extinction higher than 0.95. After switching to the metallic state, the ED and MD modes get blue shifted. Along with the blue shift, the metallic state oversees a steep increase in absorption leading to a blue shifted near perfect absorber. This result is showing the proof of concept of one of the very few dynamically tunable optical filters at the nanoscale.

The switching of vanadium in the metasurface is thermal in nature. So the switching speed for our system is the fastest time required to change the temperature of the metasurface from 65 to 70 °C which is about 3 seconds. However if the heating mechanism is faster, the switching can be achieved at a rate of a few picoseconds as discussed in the next subsection.

4.2.2 Conclusion

From the perspective of our goal of developing active nanophotonic devices, we have demonstrated two different reconfigurable devices using the same design principles. The first device serves the purpose of dynamic amplitude modulation at the nanoscale, and the second performs spectral tuning. We successfully combine the qualities of Vanadium dioxide with nanophotonic structures without hampering the functionalities of either. This research provides hope for the development of more reconfigurable nanophotonic devices by demonstrating a plausible working methodology for overcoming the design and fabrication challenges.

4.3 Nonreciprocal metasurface based on VO₂

As discussed previously, a nonreciprocal system can have dissimilar received-transmitted fields upon the exchange of sources with detectors. In other words, it simply means that the action in a nonreciprocal system if the direction of light propagation is reversed, we observe a distinct behaviour. Such systems are distinct from the standard linear time invariant systems because they don't obey the time reversal symmetry. In optics, the first time such systems were brought to light was in 1845 when Michael Faraday discovered the Faraday rotation principle. The first theoretical studies were proposed by Stokes and Helmholtz in 1840 and 1856 respectively. Nonreciprocity is the necessary and sufficient condition for the demonstration of

unidirectional propagation of light or optical isolation/circulation. Analogous non-reciprocal systems in electronics are the common p-n junction diodes and in fluid mechanics they are the valves. For electrodynamics, Lorentz formulated a mathematical condition to distinguish between reciprocity and nonreciprocity.[212, 213] Today this mathematical condition also known as the Lorentz reciprocity theorem is the standard method to determine nonreciprocity in most of the systems.

One must note however that there are certain systems that seem to be nonreciprocal, but they are actually only asymmetric. For example, let's say we have a system N. We send a beam A from the left, having an amplitude and modal profile and receive a beam B on the right having a different amplitude and modal profile. If we send the same beam A from the right in the system and receive a third beam C on the left side, we cannot call our system nonreciprocal.[62] Such systems might be asymmetric but not nonreciprocal.[214] For the system N to be called nonreciprocal, we must send in the beam B from the right side in the system and receive a beam D which is distinct from beam A in terms of modal profile, phase distribution or relative amplitudes.

In the field of photonics, most of the processes such as reflection, refraction, transmission, interference, and many more are fundamentally reciprocal processes because of their inherent time reversal symmetry [145, 215]. This leaves us with three options: (i) magnetized materials exhibiting asymmetric permittivity/permeability tensors, (ii) time-varying systems, and (iii) non-linear effects. As of date, the most used approach is the use of magnetic fields and magnetized materials. The modern optical isolators based on the principle of faraday rotation is this approach. The only major drawback of this approach is the need of high field magnets or superconductive coils to provide the sufficient magnetic fields. Such systems are obviously not suitable for nanoscale implementations. Time-varying systems have allowed the miniaturization of non-reciprocal systems to micrometer scale due to their reliance on acoustic signals to provide stimulus to induce the needed time variation.[216, 217, 218] This technology has not yet matured fully as issues such as inefficiency and modulation complexity need to be addressed.

The third approach: nonlinear light matter interactions is the only plausible mechanism for breaking time-reversal symmetry and demonstrating nonreciprocal optical at the nanoscale. Nonlinear interactions are inherent properties of the materials used in building the samples, and hence they don't require any external action or stimulus to provide nonreciprocal action. Also, considering the advent of high quality sub-wavelength nanoresonators capable of generating fields high enough to induce the generation of high harmonics in the non-perturbative regime, nonlinear symmetry

breaking seems to be particularly easily achievable[32] Such effects have already been demonstrated in systems having optical harmonics and topological edge states. Non-linear systems are mostly self-acting,[219] hence they don't require the application of any external stimulus or fields and can enable nonreciprocal action in fully passive systems.[220, 221] This makes fabrication much easier and the miniaturization to the nanoscale a natural process.

So far there have been theoretical studies that suggest the use of nonlinear nanoscale platforms for asymmetric and nonreciprocal light control. Asymmetric fields have also been demonstrated experimentally at the nanoscale using plasmonic metasurfaces [222]. One approach claiming nonreciprocal action using Kerr Nonlinearity in Silicon Nitride metasurfaces has been demonstrated so far.[223]

In this section we describe our efforts in achieving nonreciprocity at the nanoscale. We use our principles of design for active nanophotonic devices to demonstrate non-reciprocal action. As previously discussed, the only suitable approach for breaking time reversal symmetry at the nanoscale is the use of nonlinear light matter interactions. Kerr nonlinearity requires extremely high optical powers to show any noticeable symmetry breaking, hence we exploit the nonlinear phase transition of vanadium dioxide to demonstrate nonreciprocity. This phase transition occurs at 68 degrees Celsius and hence doesn't require extremely high optical powers to induce. We combine vanadium dioxide with broadly resonant dielectric metasurfaces to obtain distinct transmissions for both excitation directions.

4.3.1 Results

We design and practically demonstrate a half micron thick nonreciprocal metasurface having different forward/backward transmissions (see Fig. 4.1). The metasurface possesses nanoresonators made of Silicon arranged atop a thin film of Vanadium dioxide. Vanadium dioxide film is deposited on a glass substrate and covered with PMMA (Fig 4.1b). In the previous section we discussed that Vanadium dioxide is a phase change material whose crystalline structure is altered at 68 °C. This results in an insulator to metal phase transition resulting in a steep change in optical properties. The reversible nature of this phase transition makes it ideal for use in nonreciprocal devices and reconfigurable metasurfaces. However, unlike the thermal induction of phase transition like in the previous case, we optically trigger the phase transition using the power of our exciting laser.

The metasurface is designed to have a high transmission at 1470 nm wavelength when the vanadium dioxide is in insulating phase (Fig. 4.2c, black line). We performed the numerical simulations on COMSOL using the Floquet periodic boundary conditions

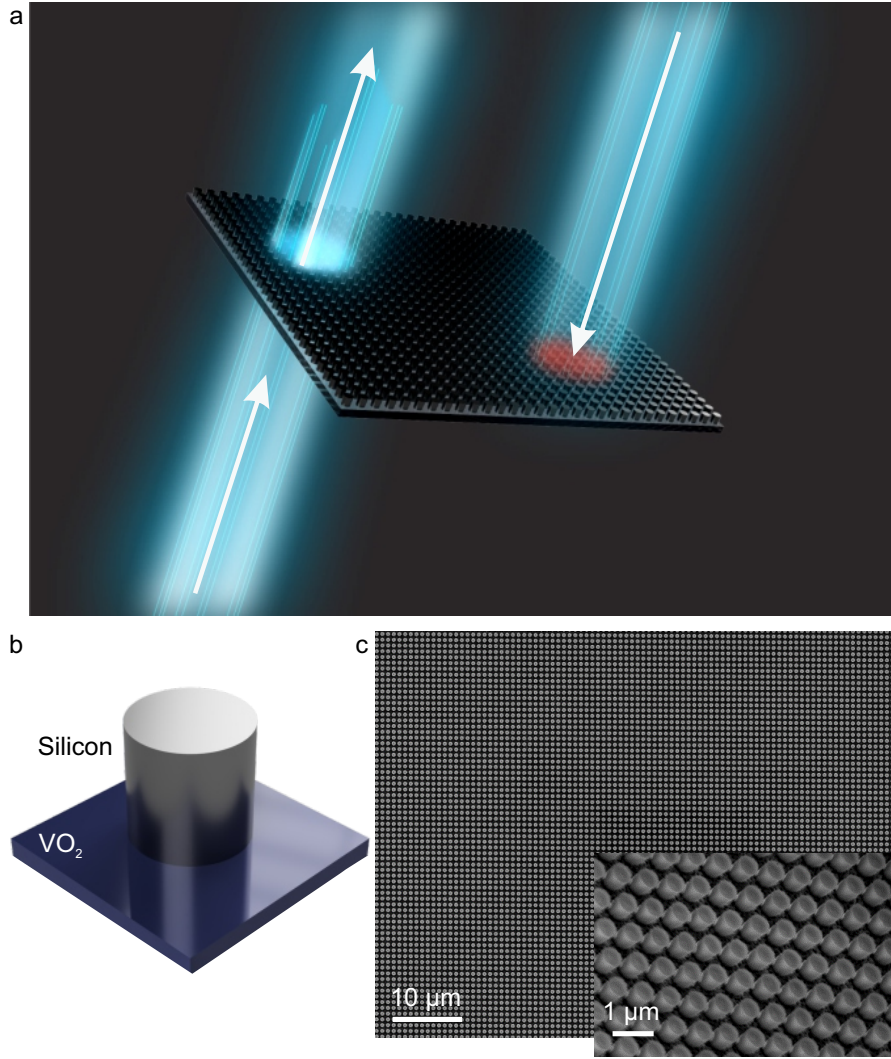


Figure 4.1: Nonreciprocal transmission of light through a hybrid Si–VO₂ metasurface. (a) Concept image of one-way transmission through a metasurface. (b) Schematics of a subwavelength resonator (metasurface unit cell): silicon disk placed on top of vanadium dioxide (VO₂) film. The metasurface resides on glass and is covered with polymethyl methacrylate (PMMA) with a refractive index similar to glass. This creates an environment close to homogeneous and isotropic for the Si-VO₂ nanostructures. (c) Electron microscope images of the fabricated Si-VO₂ metasurface.

with a unit cell depicted in Fig 4.1b. The phase transition of VO₂ to the metallic state comes with a jump in the material absorption and hence there is a significant loss in transmission (see Fig.4.2c, red dashed line). Due to asymmetry with respect to the direction of propagation of light, there is a difference in the near fields and the absorption see Fig. 4.2a. And since the absorption is different, the power needed to induce the phase change for both the incidences is also different. Therefore, we see a range of optical powers for which we get a metasurface with conducting VO₂ for one excitation and a metasurface with insulating VO₂ for the other which

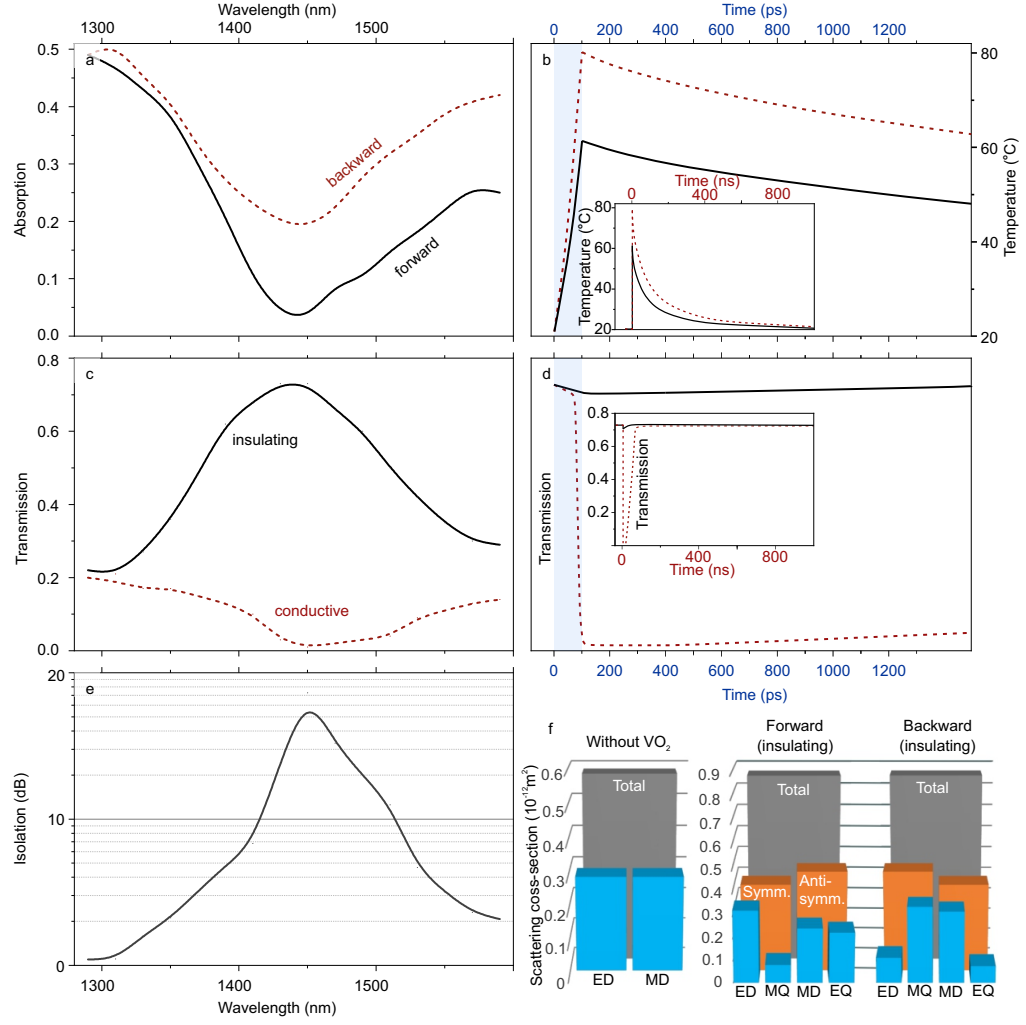


Figure 4.2: Theoretical study of spectral and temporal response of the nonreciprocal metasurface. (a) Absorption spectra of a Si-VO₂ metasurface for the insulating VO₂ phase and two directions of incidence (forward/backward). (b) Temporal dynamics of VO₂ temperature for the two opposite directions of excitation at 1450 nm wavelength under an excitation with 100 ps pulse. (c) Transmission spectra of the metasurface for insulating (black, solid) and conductive (red, dashed) VO₂ phases. (d) Temporal dynamics of the metasurface transmission for the two opposite directions of excitation at 1450 nm wavelength and under an excitation with 100 ps pulse. (e) Contrast between transmission in insulating and conductive phases. (f) Multipolar composition of the unit cell scattering: (left) metasurface without the VO₂ film featuring identical scattering for the forward and backward directions, (right) with the VO₂ film for the two opposite directions of excitation. Here ED – electric dipole, MD – magnetic dipole, EQ – electric quadrupole, MQ – magnetic quadrupole.

also implies different transmissions (high for the insulating case and low for the conducting case).

Like our previous works, we use multipolar decompositions to reveal the physics of

our metasurfaces. We decompose the scattered fields of individual unit cell having periodic boundary conditions into a series of spherical harmonic multipoles as described in chapter 1. In the spectral region of interest, scattering is dominated by the first four multipoles: electric dipole (ED), magnetic dipole (MD), electric quadrupole (EQ), and magnetic quadrupole (MQ) with negligible contributions from higher-order multipoles (see Fig 3d). High transmission of the metasurface is caused by the balance of symmetric (ED, MQ) and anti symmetric (MD, EQ) multipoles which interfere constructively in the far field in forward direction and destructively in the backward direction (see figure 4.2f) like in the case of Huygens metasurface described previously. Like in the previous section, there is presence of higher order multipoles due to the presence of VO_2 film. We observe an interplay of energy between these four scattering modes in our metasurface, which leads to distinct near fields and absorptions that we observe. This effect is also known as the magneto-electric coupling of Mie modes. Literature suggests that metasurfaces having this property can demonstrate peculiar photonic functionalities like polarization transformations, asymmetric transmission, asymmetric reflection and transverse kerker effect.

To fabricate the metasurface, 35 nm of VO_2 films were grown on a fused silica wafer and annealed in 250 mTorr of oxygen at 450 °C. 10 nm of aluminum oxide (Al_2O_3) serving as a spacer and stopper layer was deposited on the VO_2 via e-beam evaporation. 540 nm thick amorphous silicon was grown on the $\text{Al}_2\text{O}_3 - \text{VO}_2$ layered structure. The resonator structure was created by a standard electron beam lithography process with a PMMA photoresists and a 1:3 MIBK/IPA developer. An Al_2O_3 hard etch mask was prepared by electron beam evaporation, and the undeveloped resist was successfully lifted off in an acetone bath. The samples underwent reactive ion etching (RIE) to create silicon nanopillars. Finally, a top layer of PMMA was spun onto the sample to create an index matched layer. Electron microscope image of the fabricated metasurface is shown in Fig 4.1c.

We use nonreciprocal optical diagnostics (see chapter 1) to measure the optical characteristics of the fabricated VO_2 based metasurfaces. We use a bias temperature of 40 degrees Celsius which is monitored by Thorlabs TC300 temperature controller. The metasurface is then illuminated with a tunable continuous wave diode laser with power less than 10 mW. We deflect a small portion of the incident beam using a microscope slide placed at 45 degrees angle. This led to deflection of about 2 percent of the incident power onto the power meter for measurement of incident power. The power incident on the metasurface is controlled using a wire grid polarizer and a rotational stage. We weakly focus the metasurface using a long focal distance

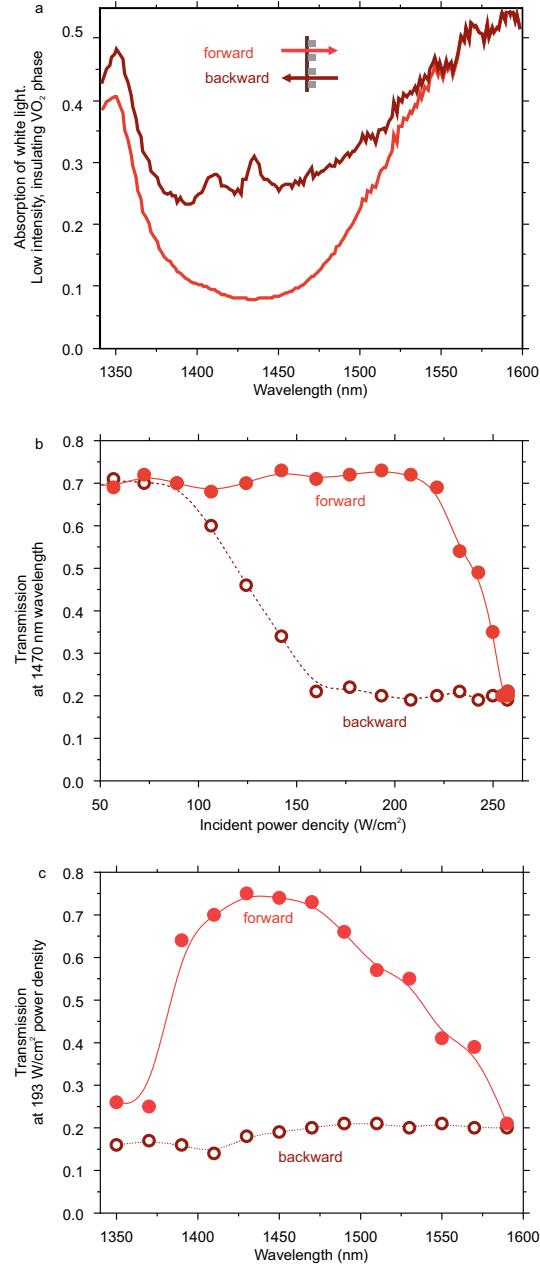


Figure 4.3: Experimental demonstration of nonreciprocal transmission of light with a hybrid Si-VO₂ metasurface. (a) Experimental white-light absorption for two opposite directions of illumination. (b) Experimental transmission for two opposite directions of illumination at 1470 nm wavelength as a function of an increasing intensity of light. For power densities in the range 150-250 W/cm², the metasurface demonstrates pronounced contrast between forward and backward transmissions. (c) Transmission for two opposite directions of illumination vs. wavelength at 193 W/cm² power density.

lens ($f=200$ mm achromatic doublet) making the illumination a near plane wave illumination (spot size: 1mm and $NA=0.005$). After illumination, we collect the light using the same lens in an identical reciprocal setting as on the excitation side. We measure this transmitted light using another power meter and reference it with

respect to the PMMA coated glass substrate. For the reverse (backward) excitation we flip the sample in the setup using additional magnetic stage.

We measure the transmission by calculating the ratio of transmitted power to the incident power for all the incident powers and wavelengths. We perform this measurement in two cycles ascending power and descending power (heating and cooling) to reveal the hysteresis behaviour of the sample and its optical bistability. Here since the transmission is dependent on the previously applied power. Together, bistability and nonreciprocity provide four different transmission dependencies (forward/backward and increasing/decreasing) see figure 4.4a. In either of the cycles (increasing or decreasing) we observe a range of wavelengths and intensities where we find prominent nonreciprocity (figure 4.3b,c).

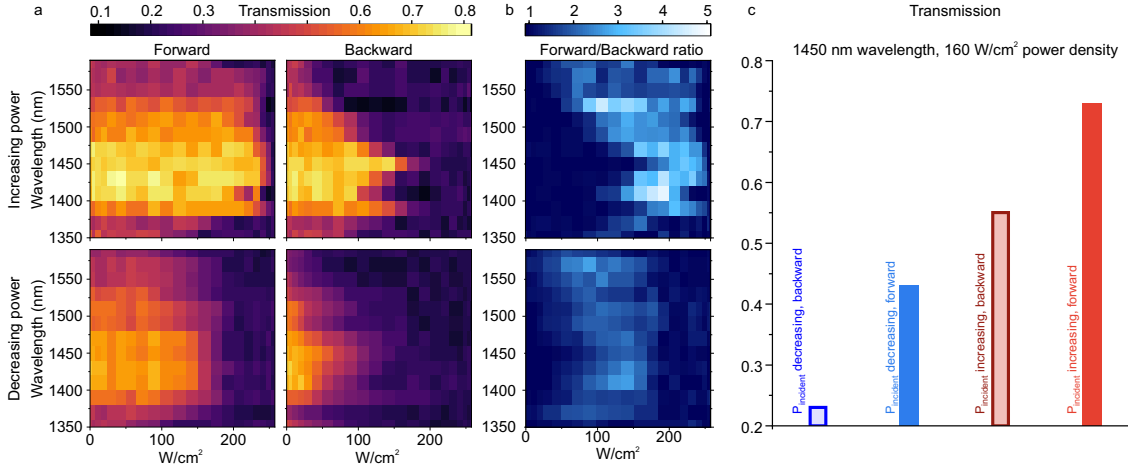


Figure 4.4: Experimental demonstration of the interplay between nonreciprocity and optical bistability. (a) Experimentally measured maps of transmission as functions of the intensity and wavelength of incident light. Top row: light intensity is increasing. Bottom row: light intensity is decreasing. First column: for the forward illumination; second column: for the backward illumination. Differences in transmission between the rows are caused by the bistability and differences between the first two columns are caused by nonreciprocity. (b) Forward-to-backward ratio showing nonreciprocity over a range of wavelengths and intensities for both cases of the increasing and decreasing light intensity. (c) Four distinct transmission levels for the same incident wavelength and power (P_{incident}) enabled by the interplay of nonreciprocity and bistability.

To quantify the degree of nonreciprocity, we calculate the observed contrast between forward and backward transmission. We choose our metric to be the ratio of forward transmission to that of the backward transmission. We see that the contrast reaches a factor of 5 in case of increasing optical power and it remains at around 3-3.5 for decreasing powers (4.4b), hence revealing that the sample shows a greater range and degree of nonreciprocity for the increasing excitation.

We throw some more light on the bistable nature of transmission in our sample. In figure 4.4c we visualize four distinct levels of transmission at the same wavelength and the same intensity depending on the direction of excitation (forward/backward) and the type of cycle or the previous intensity level (increasing or decreasing). We achieve optical bistability at comparatively low powers compared to saturable absorption or optical nonlinearity.

4.3.2 Conclusion

Just like our previous work on reconfigurable metasurfaces, we demonstrate two functionalities: nonreciprocity and bistability from one device. We show high contrast between transmission resulting from forward and backward illuminations of VO₂ based Silicon metasurfaces. This nonreciprocity in transmission is observed because of the nonlinear phase transition of VO₂ that causes a sudden change in the refractive index. The phase transition ensures that the operating powers needed for the device are pretty low compared to the devices based on Kerr nonlinearity, also this device can operate on both CW and pulsed incidence contrary to its other counterpart which can only work with pulsed inputs. Our nonreciprocal nanophotonic device works in a wavelength range of over 100 nm in the communication band region. The observed optical bistability is also highly consistent and repeatable and requires low operational powers. This device is one of the first demonstrations of optical nonreciprocity in nanophotonics. Further research can improve the range and contrast of the device and develop new nanoscale optical isolators. Such nonreciprocal devices also hold a promise for the upcoming LiDAR industry. The observed bistability also indicates towards a nanophotonic memory storage for potential applications in integrated photonics.

Using the fabrication techniques developed in the previous section, we fabricated another device where we combine the capabilities of phase change materials with those of nanophotonics to demonstrate a working nonreciprocal nanophotonic device.

4.4 Nonreciprocal metasurface based on ITO

In the recent past there have been various theoretical studies on usage of nonlinear nanoptics to demonstrate asymmetry and nonreciprocity. In the previous section VO₂ based metasurfaces were used to demonstrate nonreciprocity in transmission by exploiting the insulator to metal phase transition. Silicon metasurfaces hosting high Q resonances were also demonstrated experimentally to exhibit nonreciprocal

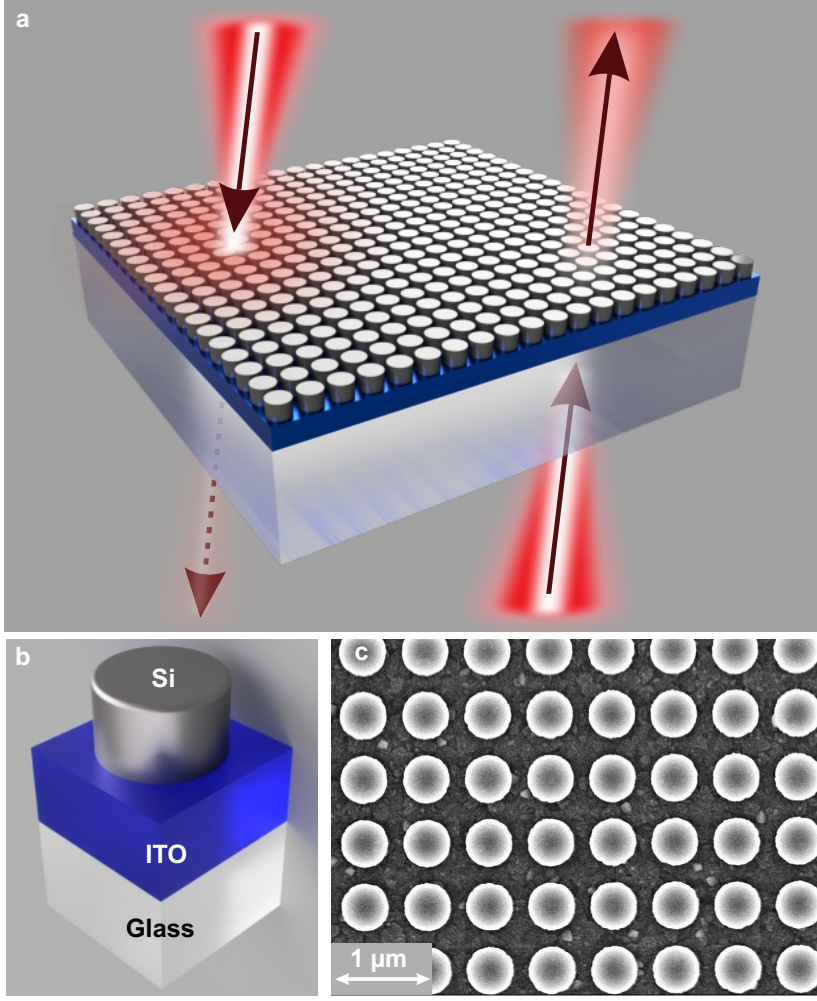


Figure 4.1: Nonreciprocal transmission of light through a hybrid Si-ITO metasurface. (a) Concept image of nonreciprocal transmission through a metasurface. (b) Schematics of a subwavelength resonator (metasurface unit cell): silicon disk placed on top of ITO and Glass film. (c) Electron microscope image of the fabricated Si-ITO metasurface.

transmission using Kerr nonlinearity. However, the Silicon based metasurfaces were faced with a major drawback that they required very high optical powers for the demonstration of their thermal nonlinear nonreciprocity. This shortcoming is in fact a characteristic of most nonlinear materials. The nonlinear change in refractive index in the perturbative regime requires the use of extreme optical powers to incite nonreciprocal action.

In this section, we overcome this roadblock by using a highly nonlinear material ITO. In 2016, it was demonstrated that the real permittivity of ITO reaches zero at around 1243 nm. Differential calculus reveals that in this region where permittivity tends to zero, the variation in refractive index is huge (permittivity tending to zero in the equation $dn = \frac{1}{2} \frac{d\epsilon'}{\sqrt{\epsilon'}}$) [224]. The equation implies that, at the near epsilon

zero wavelength, a sudden change in permittivity can trigger a very strong change in refractive index. This sudden change in permittivity and consequential huge change in refractive index at the near epsilon zero wavelength can only be induced by nonlinear action. And since this change in refractive index due to nonlinearity is huge, we exploit this phenomenon to demonstrate nonreciprocal transmission.

4.4.1 Results

We experimentally demonstrate a 500-nm thick non-reciprocal metasurface with different forward and backward transmissions for identical incidence illumination (see Figure 4.1). The metasurface is composed of square unit cells of Silicon nanodisk resonators placed on a 310 nm thick ITO over a glass substrate, as shown in Figure 1b. ITO is an Epsilon near zero (ENZ) material which has extreme nonlinearity in the wavelength region of around 1230 nm where the real part of its permittivity reaches absolute zero. This extreme nonlinearity results in very high coefficients of kerr nonlinearity and two photon absorption around the ENZ region.[224] It has been demonstrated that this giant nonlinearity can lead to extreme changes in refractive indices at high incident powers of around 100 GW/cm². ITO is particularly useful for symmetry breaking as the nonlinear transition is sharp and fully reversible being a third order nonlinear effect itself. This large complex refractive index variation produced by cranking up incident power in the ENZ wavelength regime of this material made it an attractive choice for metasurfaces reconfigurable thermally or electrically. Here we trigger the nonlinear transition of ITO by the self-action of the incident laser pulse enabling symmetry breaking and hence nonreciprocity induced by nonlinearity.

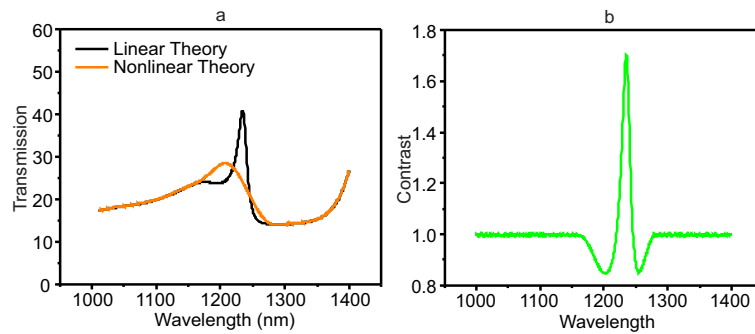


Figure 4.2: Numerical results for Si-ITO metasurface. (a) Linear (Black) and Nonlinear (red) transmission. (b) Calculated Forward to backward ratio.

We engineer the metasurface parameters to have a resonance at around 1230 nm wavelength when the ITO is in its linear state (Fig. 4.2a, black line). The numerical simulations were performed in COMSOL using the Floquet periodic boundary

conditions with a unit cell depicted in Fig. 4.1b. The transition of ITO to its nonlinear state kills off the resonance due to change in refractive indices and hence the transmission is reduced (see Fig.4.2a, red line). In the unit cell, symmetry is broken by the ITO layer which results in non-identical near fields and multipoles for the opposite sides of illumination (Fig 4.3). However the nonidentical near fields as well as multipoles do not change the transmission for both illuminations implying that until now the metasurface is reciprocal. In the linear regime, the differences in absorptions are compensated by differences in reflections.

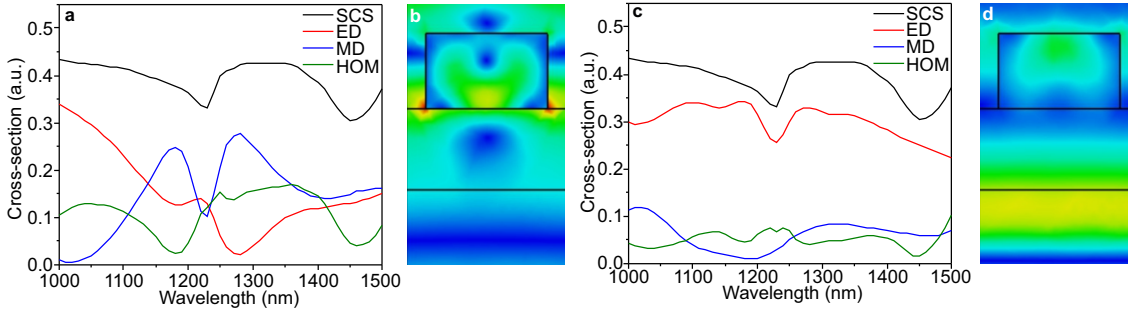


Figure 4.3: Multipolar and field analysis of metasurface. Near-field electric field amplitude distributions at the ENZ wavelength of 1230 nm for (a,b) front and (c,d) back illumination. The legend: all - all multipole contributions (black); ED - electric dipole (red), MD - magnetic dipole (blue), HOM - higher order multipoles (green).

To fully understand the generation of nonreciprocity in the metasurface we perform analysis of scattered fields from individual unit cell with floquet boundaries and perform their decomposition into a series of Mie multipoles. The multipolar decompositions were evaluated as described in ref . In the wavelength regime of interest, the scattering is primarily dominated by two modes electric dipole (ED) and magnetic dipole (MD), with negligible contributions from higher-order multipoles (see multipolar spectra and near fields in figure 4.3). The transmission resonance results due to a dip in either ED or MD scattering at incident wavelength of 1230 nm.

This results in completely different near fields for both illuminations. The cases where ED component is greater, have higher field concentration near the air-Si interface and for the cases where MD component is higher we see that higher fields reside near the Si-ITO interface. High near fields near the Si-ITO interface imply that the nonlinear transition will take place at lower incident powers than in case where high field is concentrated near Si-air interface. Hence in the incident power range between the power required for ENZ transition for both the illuminations, we observe difference in transmission or nonreciprocal transmission. The presence of the ITO film breaks the geometrical symmetry of the design enabling

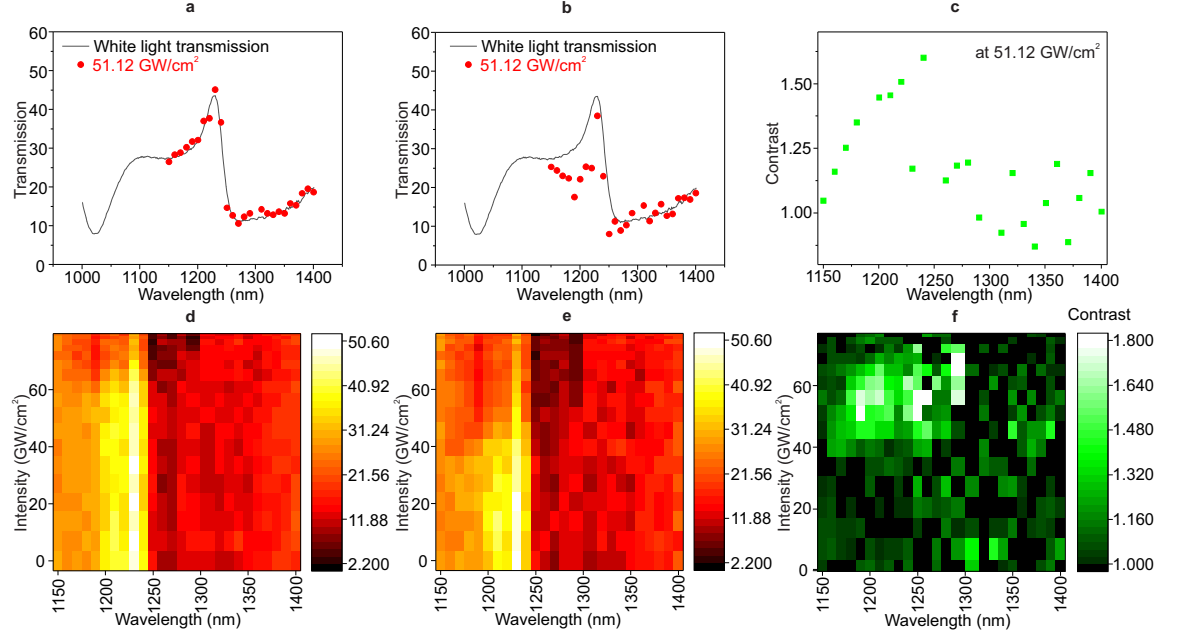


Figure 4.4: Experimental results. White light transmission and nonlinear transmission at incident power of 51.12 GW/cm^2 for (a) front (b) back illuminations. (c) Front to back ratio at incident power of 51.12 GW/cm^2 . Wavelength and Power dependence of nonlinear transmission for (d) Front and (e) Back illuminations. (f) Wavelength and Power dependence of Forward to Backward ratio

magneto-electric coupling between the Mie multipoles. Metasurfaces that exhibit magneto-electric coupling exhibit peculiar photonic functionalities such as polarization transformations [149], including asymmetric transmission [214], asymmetric reflection [225], transverse Kerker effect [226], photonic analogues of spin-Hall effects [227], photonic Jackiw-Rebbi states [228], nontrivial topological phases [147].

We proceed with optical diagnostics of the fabricated metasurfaces. We illuminate the metasurface with an Optical parametric amplifier (OPA) with pulse duration of 230 fs. A small portion of the laser beam is reflected onto a thorlabs power meter which measures the incident intensity level. The power is attenuated with a set of polarizers. The laser beam is then tightly focused onto the metasurface with a Mitutoyo planapo 100x NIR objective. This is followed by measuring the transmitted signal using another identical 100x NIR objective. We detect light transmitted through the metasurface with a second Thorlabs power meter and reference it to the transmission through the glass substrate. For forward/backward experiments we flip the sample inside the setup. Figure 4.4 shows transmission through the metasurface for forward and backward scenarios of excitation.

4.4.2 Conclusion

We show difference between forward and backward transmissions of Si-ITO metasurfaces of a half wavelength thickness. Nonreciprocity in transmission caused due to transition of linear to nonlinear ITO which behaves like an ultrafast self acting medium. The metasurface requires high peak power (100 GW/cm^2), ultrafast laser pulses to ensure the triggering of nonlinear transition and the safety from burning due to material absorption. In contrast to VO_2 based nonreciprocal metasurfaces, these require higher powers but are almost as fast. Our ITO based metasurfaces has an operating wavelength range of about $1230 \pm 15 \text{ nm}$, and power range of $40\text{-}80 \text{ GW/cm}^2$. Such a nonlinear nonreciprocal metasurface can create new inroads into ultrafast nonreciprocal nano-optical components for optical information channeling, LiDAR machine vision , switching and even logic.

Conclusions

In this work, we developed two types of active nanophotonic devices, light control devices and light generating devices. The light generation devices like fiber amplifiers, lasers and light emitting diodes produce light with specific properties suited for applications in the field of fiber communications, spectroscopy, biophotonics and quantum optics. These devices can precisely control polarization, amplitude, coherence, wavelengths and even entanglement of photons at nanoscale.

We develop both spontaneous light emitters and nanolasers, spontaneous light emitters use a relatively new active material namely the rare earth doped nanoparticles capable of providing spectrally tunable photoluminescence and upconversion. These rare earth doped nanoparticles are interesting from the viewpoint of biomarking and biophotonics and most of their emission properties like wavelength and intensity can be altered by playing with their chemistry, morphology and doping. We overcome the challenge of manipulating emission polarization which is impossible using chemical methods. Our works like photonic rashba effect in upconversion photoluminescence enable efficient polarization control by not only increasing upconversion efficiency but also generating circularly polarized light at the source instead of traditional conversion. Through our works on luminescence we developed an understanding that generation of polarized light at the source via optical coupling with nanoresonators is the best way to produce polarized light at nanoscale.

The main challenge in nanolasing sector is posed by losses in the geometries approaching subdiffraction limit. We use the commercially available semiconductor quantum wells as our active material and etch nanostructures on them that are designed to support high quality factor resonances. Depending on the type of nanostructure we etch, we demonstrate two distinct lasing capabilities: resistance against disorders and fabrication imperfections and Q factor and lasing wavelength control. Our work on topological lasers showcases robustness against disorder and the work on anapole lasers demonstrates precise wavelength and quality factor control which can dictate lasing features like linewidth and lasing threshold. Through these

works we bring quantum well lasers to the nanoscale, eliminating the need of temperature control and voltage bias. From the viewpoint of optical data processing, our nanolasers provide the optical analogue of current in an electronic logical operation. The laser light once generated at the nanoscale can be guided using waveguiding channels (interconnects) all around the circuit board.

On the other hand, light control devices like optical modulators, routers, isolators, filters and multiplexers are designed to process input light in different ways like altering phase, amplitude, polarization and direction mainly for applications in optical signal processing, quantum information processing and experimental optical diagnostics. Control devices add the capability of generating complex signals and operations on light produced from various sources. Their functionality is like electronic circuit components like multiplexers, switches and logic gates.

Amongst all light manipulation operations, the phenomena of optical isolation and dynamic tuning are difficult to realise on the nanoscale because of extra space needed to add external stimuli like magnetic fields, electric fields or acoustic signals. We use phase change materials and epsilon near zero materials like Vanadium dioxide and Indium Tin oxide respectively in our Silicon metasurfaces and provide a thermal or optical stimulus to demonstrate isolation. The paper on tunable metasurfaces shows that the phase change of vanadium dioxide enables the metasurface to have two modes of operation thereby adding switchable features. Our works on nonreciprocal metasurfaces based on VO_2 and ITO exploit the nonlinear nature of this phase transition. The extreme nonlinearity breaks the Lorentz reciprocity condition thereby enabling nonreciprocal action and optical isolation. We learn that the introduction of materials that can induce a sudden change in refractive index in a nanophotonic device can drastically change its optical properties if this refractive index change is triggered.

Through this work, we developed and tested some design principles for active nanophotonics devices that will further the work in this field. The first of them is the principle of choosing a good active material suited for our desired functionality. For example, for making light sources we need media with high optical gain like quantum wells or quantum dots. The exact type of material is chosen based on ease in fabrication procedure, for example we chose Vanadium Dioxide instead of other phase change materials because its transition is accessible, and it is easier to incorporate in Silicon metasurfaces compared to others.

Secondly, we choose the working physics of the system. The working physics dictates to what degree our desired functionality is achieved. In case of nanolaser design, the lasers based on topological metasurfaces are robust against fabrication imperfec-

tions and the ones that are based on anapoles are extremely geometry sensitive but they offer precise control over lasing linewidth, threshold and quality factors. The working physics decides the type of nanophotonic structure which is to be used in the device.

And lastly, the positioning of active materials with respect to the metasurface elements determines the overall efficacy of the device. Upconversion nanoparticles when spin coated atop the Berry phased metasurface produce more intense and polarized photoluminescence. However, if not positioned properly, there is almost no effect. Similarly, the position of Vanadium Dioxide film in our work on nonreciprocity has a vital role. It is sandwiched in between Silicon and glass, making the absorption of magnetic dipole mode feasible. Overall, the design process is a mix and match between a pool of active media and nanophotonic structures. Once this match is performed, an ideal geometrical structure is designed keeping in mind all the constraints and limitations.

The major roadblocks in the field of active nanophotonics are mainly fabrication costs, fabrication challenges, and integration with other nanophotonic devices. The fabrication costs of a single active metasurface are not economical for consideration for production. Different active media have different chemistry, making it difficult to find a standardized procedure for fabrication. Eventually, achieving a common platform on which all the designed devices could work together harmoniously could be the last nail in the coffin toward bringing active nanophotonics to the market. In this work we overcome the aforementioned challenges to some extent. The use of a relatively new active material: Rare earth doped nanoparticles solved the issue of complicated fabrication as it only used the method of drop casting or spin coating for integration with the metasurface. The use of membrane based topological structures has also been vital in solving the issue of planar waveguiding of light generated at the nanoscale.

We believe that this work is one of the first works in the field of active nanophotonics that encompasses practical demonstrations in almost all the major directions the field of nanophotonics has to offer. Solutions to some of the challenges in the field have also been found. We develop new design fundamentals and show the proof-of-concept demonstrations of diverse functionalities ranging from light generation to light control.

Future work in this field can provide answers to the major questions posed by device integration. There is lack of a common platform where all the active devices can be built on, in other words there is lack of a breadboard where all the components could work together. In development of the product, there are performance limitations and

manufacturing costs which need to be thoroughly researched. Further work in the field can open up pathways in the field of optical and quantum technologies.

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