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Khaidarov, Egor

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**NANYANG
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SINGAPORE

**CONTROL OF LED RADIATION WITH
FUNCTIONAL DIELECTRIC
METASURFACES**

EGOR KHAIDAROV

**SCHOOL OF ELECTRICAL AND ELECTRONIC
ENGINEERING**

2020

Control of LED radiation with functional dielectric metasurfaces

EGOR KHAIDAROV

**SCHOOL OF ELECTRICAL AND ELECTRONIC
ENGINEERING**

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

2020

Statement of originality

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

7 March 2020

Date

Egor Khaidarov

Supervisor Declaration Statement

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

7 March 2020

Date

Prof. Hilmi Volkan Demir

Authorship Attribution Statement

This thesis contains material from 4 papers published in the following peer-reviewed journals in which I am listed as an author.

Chapter 2 is partially published as:

R. Paniagua-Domínguez, Y. F. Yu, E. Khaidarov, S. Choi, V. Leong, R. M. Bakker, X. Liang, Y. H. Fu, V. Valuckas, L. A. Krivitsky, A. I. Kuznetsov, “A Metalens with a Near-Unity Numerical Aperture”, *Nano Letters*, 18 (3), 2124–2132, 2018. DOI:10.1021/acs.nanolett.8b00368

The contributions of the co-authors are as follows:

R.P.-D. and A.I.K. conceived the idea. E.K. designed the nanoantennas and R.P.-D. designed the flat lens. E.K. and R.P.- D. performed the numerical simulations. Y.F.Y. fabricated the samples and performed the optical measurements. S.C., V.L., and L.A.K. built the confocal setup and performed fluorescence measurements. Y.H.F. and X.L. helped with the optical measurements. V.V. performed the SEM imaging and *Cr* hole fabrication. R.M.B. performed NSOM measurements to corroborate the measured focal spot. R.P.-D. wrote the first draft of the manuscript. A.I.K. coordinated the work. All authors discussed the results and reviewed the manuscript. R.P.- D., Y.F.Y., and E.K. contributed equally to this work.

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E.K. designed the structures; E.K. and R.P.D. performed the numerical simulations; H.H. and J.N.S.K. performed the *TiO₂* deposition; H.H., S.Y.L.K., and Y.T.T. fabricated the

samples; H.H., Y.H.F., and Y.F.Y. performed the optical characterization of the samples; V.V. performed SEM imaging; A.I.K. and R.P.D. developed the concept and coordinated the work; E.K., R.P.D., and A.I.K. prepared the manuscript. All authors discussed the results and commented on the manuscript.

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N.K.E and E.K. fabricated the samples, E.K. and R.P.-D. designed and numerically simulated the structure. Y.H.F. did the optical characterization. S.L., X. Z. and S.T.T. did the *GaN* wafer fabrication. V.V. did the SEM imaging. H.V.D. and A.I.K. conceived the idea and coordinated the work. N.K.E. prepared the first draft of the manuscript. All authors discussed the results and reviewed the manuscript.

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The contributions of the co-authors are as follows:

E.K. and Z.L. contributed equally to this work. Z.L., Y.A., H.V.D., and A.I.K. conceived the idea; E.K., Z.L., and Y.A. designed the metasurface and the FP cavity; Z.L., Y.A., E.K., and R.P.-D. performed the numerical simulations; E.K. fabricated the FP cavity and the metasurfaces on top of the LED; S.T.H. performed the optical measurements; X.L. measured the vortex beam emission; V.V. performed the SEM imaging; E.K. and Z.L. wrote the first draft of the manuscript. A.I.K., H.V.D., C.E.P,

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Abstract

Light-emitting diodes (LEDs) are excellent candidates to replace the widespread conventional fluorescent light sources. This stems from their higher compactness and brightness, as well as their better performance in terms of energy efficiency, long lifetime and high color rendering qualities. Their potential use in a broad range of applications has attracted enormous interest to the LED research and development. This has translated to a rapid improvement of the LED characteristics as well as to their commercialization in past years. Further integration and miniaturization of these devices, for applications such as optical communications, requires a higher level of control in terms of LED emission characteristics and a compact solution for any desired functionality on the output. Metasurfaces (structured surfaces with engineered characteristics) grant an unprecedented control over the wavefront of light, while retaining a subwavelength-thickness feature (relative to the excitation light wavelength). Moreover, in some cases, they also offer opportunities for large-scale industrial fabrication. Among different types of the metasurfaces, those based on dielectric and semiconductor materials typically exhibit lower losses comparing to metallic counterparts, which potentially enhances the efficiency of their integrated devices.

The main focus of this dissertation is to develop and demonstrate the compact and novel LED platforms, with light emission characteristics on demand, enabled by means of dielectric metasurfaces. For this purpose, highly efficient metasurfaces based on dielectric and semiconductor materials (*Si*, *TiO₂*, *GaN*)

have been designed and fabricated. The functionalities realized with these include beam deflectors, polarization beam splitters, complex light field generators and lenses. For the latter, a novel class of metasurfaces with asymmetric radiation profile was engineered. With them, light channeling into a single desired diffractive order has been achieved with efficiencies reaching 99%. Ultra-high angle beam deflection metasurfaces operating in the visible regime were demonstrated using TiO_2 for the blue and green and Si for the red part of the spectrum. Based on this concept, a near unity numerical aperture (NA) lens was designed and fabricated, far exceeding any previously reported, both commercial and laboratory experimental models.

Moving towards direct metasurface integration on conventional LED platforms, GaN metasurfaces were etched directly into optically thick GaN slabs. Despite the low index contrast between the patterned metasurface and its substrate (both GaN), the metasurfaces exhibited high efficiencies across the operational wavelength range of the LED emission. However, when excited directly with the photoluminescence from the LED, the desired functionality was lost due to the low spatial coherence (Lambertian shape of radiation pattern) of the LED. Hence, the approach for direct integration of the metasurface on top of LED was found to be inefficient.

To solve this issue, external and internal cavity design solutions for the LED and metasurface integration were proposed and engineered. The efficient transformation of the Lambertian radiation pattern into plane-wave-like radiation, suitable for the metasurface to work, was experimentally demonstrated. Further integration with the designed metasurfaces allowed to obtain advanced functionalities for the LED emitted light, namely beam deflection and vortex beam generation. Those were realized in a GaP LED architecture via optical pumping. The electrically-driven GaN LED device with a beam deflection functionality was demonstrated using the external cavity method.

The results presented in this dissertation constitute a step forward towards

compact, advanced, efficient and integrated optical devices, by leveraging on the attractive platform of LEDs and the emerging field of metasurfaces, which enables unprecedented control of light. The method proposed can be applied to any other incoherent light source beyond LEDs, and may find broad applications in optical communications, Li-Fi, displays, sensing, smart lighting and many more.

Keywords: Light-emitting diode, dielectric metasurfaces, resonant cavity light-emitting diode, high angle beam deflection, high numerical aperture lens, vortex beam, orbital angular momentum, silicon, titanium dioxide, gallium phosphide, gallium nitride.

Dissertation organization

This dissertation starts with the introduction and motivation of this thesis research in Chapter 1, followed by a detailed literature review on the light-emitting diode (LED) structure, properties and modification influence on the LED radiation. Next part of the literature review is devoted to metasurfaces, their development from metamaterials, comprehensive description and comparison of plasmonic and dielectric metasurfaces, their peculiarities, advantages and drawbacks. At the end of Chapter 1 the goals of this dissertation are put forth and the main accomplishments in the pursuit of fulfilling those goals are described.

Chapter 2 is focused on development of the advanced metasurface designs that outperform the existing conventional bulky and even previously reported flat optical components. The novel concept behind ultra-high angle beam deflecting metasurfaces is explained and demonstrated using *Si* and *TiO₂* platforms. At the end of Chapter 2 the apogee of this concept is presented - the state-of-the-art ultra high numerical aperture lens outperforming any of the known analogues.

Chapter 3 is a step towards integration of the LEDs and metasurfaces, featuring the most straightforward approach - direct patterning of the LED surface with nanoantenna structures. The initial metasurfaces are designed with regularly used plane wave excitation, patterned directly into the LED platform. The concept underlying the effective metasurfaces with low refractive index contrast with the substrate is explained and experimentally demonstrated on a GaN platform. However, high efficiencies in that case are possible only with plane wave excitation. At the the beginning of Chapter 4 the challenge of LED

and metasurface integration is fully revealed, underlining the impracticality and functional limitations of the direct patterning of the LED surface approach.

Chapter 4 and 5 propose the solution for full wavefront control of the LED radiation using the collimating cavity and metasurfaces. External and internal cavity methods are demonstrated on the *GaP* and *GaN* LED platforms efficiently enhancing the spatial coherence of the LED. The working principle of the concept is shown employing the optically pumped *GaP* LED platform with a beam deflecting metasurface. The full potential of the technology is unveiled in Chapter 5 with electrically-driven *GaN* external cavity platform highlighting the beam deflecting functionality.

Chapter 6 concludes the work in this dissertation and describes the plans for future work.

Acronyms

AR Augmented Reality

BIC Bound State in the Continuum

CCD Charged Coupled Device

CMOS Complementary Metal-Oxide Semiconductor

DBR Distributed Bragg Reflector

DC Direct Current

EBL Electron Beam Lithography

EL Electroluminescence

ENZ Epsilon Near Zero

EQE External Quantum Efficiency

FP Fabry-Pérot

FWHM Full Width at Half Maximum

GAN Generative Adversarial Networks

IAD Ion Assisted Deposition

ICP Inductively Coupled Plasma

IQE Internal Quantum Efficiency

LCDs Liquid Crystal Displays

LDOS Local Density-of States

LED Light-Emitting Diode

Li-Fi Light-Fidelity

LLO Laser Lift-Off

LSPR Localized Surface Plasmon Resonance

μ LED Micro Light-Emitting Diode

MOCVD Metal-Organic Chemical Vapour Deposition

MQW Multiple Quantum Wells

NA Numerical Aperture

ND Nanodiamond

NV Nitrogen-Vacancy

NZI Near Zero Index

OAM Orbital Angular Momentum

OLEDs Organic Light-Emitting Diodes

PBS Polarization Beam Splitter

PhC Photonic Crystal

PECVD Plasma-Enhanced Chemical Vapour Deposition

PL Photoluminescence

PSS Patterned Sapphire Substrate

QD Quantum Dot

RCLED Resonant Cavity Light-Emitting Diode

RGB Red, Green, Blue

RIE Reactive Ion Etching

SEM Scanning Electron Microscope

SERS Surface Enhanced Raman Spectroscopy

SHG Second Harmonic Generation

SOM Spin Orbital Momentum

SPP Surface Plasmon Polariton

SRR Split Ring Resonator

SSL Solid-State Lighting

TFT Thin-Film Transistor

TIR Total Internal Reflection

THG Third Harmonic Generation

UV Ultra Violet

VR Virtual Reality

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Chapter 1

Introduction

1.1 Background and motivation

Photonics, being a successor and a complement to electronics and optics fields, inherited an abundance of existing practical applications as well as the means and colossal enthusiasm for new research and development. Successful research resulted in the emergence of unique photonics devices and applications, one cannot imagine the current everyday life without. This includes consumer products (gadgets, displays, laser printers, Blu-Ray devices), telecommunications and information processing (optical fibers, data centers), biology and medicine (laser surgery, endoscopy, chemical synthesis and analysis), construction and metrology (range measurement, leveling), sensors and detectors (charged coupled devices), entertainment (holograms, laser shows, virtual reality) and computing to name a few. These revolutionary breakthroughs are not neglected by the society - international recognition and support are evident on the highest scientific rewards level: Nobel prize in Physics in 2018 was awarded for development of the optical tweezers and high intensity ultra short pulses lasers using the chirped pulse amplification method. In 2014 Nobel prizes in two categories were awarded for the photonics research: invention of blue LED and laser diodes in physics, super-resolution microscopy in chemistry. In fact, around

30% of physics Nobel prize winners are strongly related to or benefited from the photonics and optical technologies [1].

Technical progress sparked by those significant inventions is directed towards further efficiency enhancement of the devices, integration and compactness, customization and broader set of functionalities. In the same manner as LEDs outperform and replace fluorescent and incandescent lamps, the emerging field of metasurfaces (patterned surfaces with engineered electromagnetic properties of light at the output) indicates the potential to go beyond and substitute the bulky conventional optical components. We envision future confluence of those research directions and integration of top-notch LEDs and novel metasurfaces into the efficient, multifunctional and compact devices.

1.2 Literature review

1.2.1 LEDs

LEDs as one of the most widely-spread light sources play a major role in a vast variety of applications. In particular, III-V group semiconductor LEDs stand out from the other solid-state lighting devices due to many advantages. The outstanding simplicity is one of the main advantages (plain p-n semiconductor junction) providing higher robustness and less fragility compared to conventional incandescent and fluorescent light sources. Another reason is the high efficiency [2] that makes them a priority choice for illumination purposes not neglecting the ecological issues (mercury free, reduction of CO_2 and other greenhouse gases pollution).

Inorganic LEDs are frequently used as backlight in liquid crystal displays (LCDs), solid-state lighting (SSL), imaging and electronic devices by virtue of their long lifetime (can reach $>100\,000$ hours) and small power consumption [3]. Longer lifetime and material stability are the main advantages of the inorganic

LEDs over the organic light-emitting diodes (OLEDs) [4]: blue OLED pixels in displays degrade faster than red and green leading to distorted color balance. Additionally, the wavelength of an inorganic LED can be tuned by alloying its materials, for example, one of the most widely-spread GaN (Gallium Nitride) based LEDs can be alloyed with Al and In to cover almost all of the ultra violet (UV) (230 - 400 nm) [5] and visible (400 - 700 nm) range [6], respectively. Furthermore, LED spectral bandwidth can be also relatively narrow (~ 50 nm), leading to rich and wide color gamut and, hence, realistic color rendering of the illuminated object.

Some of the applications require white light sources for operation. In particular, thin-film transistor (TFT) LCDs employ the white light LED as a backlight, followed by specific color filters for red, green and blue pixels implementation. White color is typically generated by blue GaN based LED combined with downconverted light from phosphors (e.g. garnets (YAG:Ce), silicates, nitrides) [7]. When the LED is combined with colloidal quantum dots (QDs) the emission bandwidth of the corresponding color peaks can be narrowed even further (< 30 nm) [8]. Individual inorganic micro light-emitting diode (μ LED) can, in principle, act as emissive pixels in displays without any additional liquid crystals [9] in a similar way as the active matrix OLEDs, however the packaging process still remains a challenge today.

The brightness of an LED is another benefit, beating the majority of the other SSL competitors and conceding only to lasers. However, LEDs are not constrained by the threshold current limitations, which makes LEDs a superior and more suitable alternative to lasers for those practical applications where temporal and/or spatial coherence with ultra-fast operation is not a concern. Low heat, light weight, small footprint, and rapid on/off switching with minuscule power consumption [10] take LEDs beyond lighting applications market, underlining the future potential as signal sources in optical circuits, data centers and supercomputers.

Hence, current research on LED focuses on further enhancement of the competitive advantages and reduction of the drawbacks of the devices. Nonetheless, researchers also look into new directions, in particular, to introduce additional functional features to the device: full polarization control, highly directional emission, complex beam formation. Despite the opulence in design variations of GaN LED (conventional lateral, vertical, flip-chip, double side, thin film etc.) [6], [11], the conceptual building blocks of the device remain constant. Figure 1.1 shows the widespread conventional lateral (a) and vertical (b) LED chip configurations, correspondingly.

Figure 1.1: LED chip configurations: lateral (a) and vertical (b).

A typical growth process using metal-organic chemical vapour deposition (MOCVD) includes p- and n-doped GaN layers grown on insulating substrate (sapphire) with several alternating layers of multiple quantum wells (MQW) in between. The electrical contacts on p- and n- doped layers inject the charges into the p-n junction. Recombination of the electron and hole pair produces a photon emitted into the environment. The material of electrical contacts may

vary depending on the design from copper or aluminum to transparent conductive oxide (indium tin oxide, ITO) on one or both sides of the LED device.

Deeper refinement of the LED architecture serves the aim of enhancement of the key LED parameters. One of the most crucial among them is the external quantum efficiency (EQE) [12] characterizing the exact number of photons extracted from the device into surrounding medium divided by the quantity of injected electrons. EQE can be decomposed into the more basic injection efficiency, internal quantum efficiency (IQE) and extraction efficiency.

$$\eta_{EQE} = \eta_{Injection} \times \eta_{IQE} \times \eta_{Extraction} \quad (1.1)$$

All the constituents of EQE have a strong influence on the performance of the LED device: injection efficiency defines the number of electron-hole pairs injected in the active layer (MQW) normalized to the overall number of electrons passing through the device. Considering that electron-hole pair can undergo either radiative or nonradiative recombination, the IQE specifically determines the radiative part. Finally, the extraction efficiency describes how many of the photons generated in the LED can escape into the environment.

A broad, thorough review on the injection enhancement and IQE is presented in [13]. Particularly, modifications to the LED design such as additional electron blockade layers and hole blockade layers lead to a higher charge injection and improved electron-hole confinement. The introduced modifications have to be precisely engineered in pursuance of the high hole injection efficiency and current uniformity. Subsequent MQW rearrangement combined with a suitable set of parameters variation (doping percentage, layer thickness, chemical composition) decrease the IQE deteriorating effects e.g. polarization, current crowding and Auger recombination.

Aside from the IQE improvement, the state-of-the-art LED research is concurrently engaged in boosting the extraction efficiency of LEDs. The total

internal reflection (TIR) limits the extraction cone: directions within a solid angle for which the photons can escape the LED and not reflected back, trapped and absorbed in the device (Figure 1.2 a). TIR occurs when light is incident on the boundary of two media - from higher refractive index medium (GaN) into the lower refractive index (air).

Figure 1.2: (a), Total internal reflection and extraction cone in LEDs. (b), Patterning of LED surfaces and influence on light extraction.

Fortunately, the relative simplicity of an LED system offers a huge space for design variations with almost imperceptible constraints. For instance, by enclosing an LED within a resonant cavity [12] the electric field inside the LED is modified in consequence of the interference effects. A resonant cavity may be constructed from metallic or dielectric mirrors (distributed bragg reflector (DBR), photonic crystal (PhC)). The first impact of the cavity is the higher electromagnetic local density-of states (LDOS) that amplifies the spontaneous emission [14], [15] (Purcell effect [16]). The second effect is the reshaping of the emission pattern [17]: the resulting directivity pattern is concentrated within the extraction cone, which enhances the extraction efficiency.

Surface roughening and patterning of structures in the sapphire substrate not

only improves the quality of the GaN growth, but positively influences extraction efficiency as well (Figure 1.2 b). The pattern acts as additional light scatterers that increase the extraction by generating additional wave-vector component spread [18]. Modification and tweaking of the LED top and bottom surfaces and slab thicknesses (with respect to the MQW) suppresses and/or enhances some of the excited modes. Several cases with PhC incorporation into top [19], [20] (Figure 1.3 a) or bottom [21] slab have been examined; the highest efficiency exhibited the case of the low number of guided modes operation (i.e. higher order modes were eliminated). A detailed analysis of the modes existing in the PhC modified LED structures coupled with reflectors (metallic and dielectric) is shown in [22]. The effect of surface roughening on both top and bottom slabs is meticulously analyzed in [23], concluding a distinct positive influence on light extraction. Artificial quasi-random porosity introduced by nanoimprint and etching techniques into the p-doped layer of GaN is compared with regular periodic PhC patterning in [24]; extracted light efficiencies for both structures display similar values, marginally higher for the porous design. Scatterers in surfaces of standard lateral or flip-chip LED configurations can be etched in different geometrical shapes: cylinders [25], (truncated) cones [26], hexagons [27], pyramids [28], blocks. All those have been shown to slightly improve the extraction efficiency, via different electromagnetic effect (distribution of charges, electric fields, guided modes, resonances, if any) occurring in the fabricated patterns.

Figure 1.3: LED modifications for extraction efficiency and directivity enhancement. (a) Extraction enhancement via coupling of the trapped guided modes of the LED slab to diffractive modes in a PhC. Insets show the microscope image of PhC and the experimental far field emission diagram; reproduced with permission from [20]. (b), Directional emission by combination of SPP with asymmetric particles (triangular prisms) on top of the GaN LED, inset is the experimental emission pattern; reproduced with permission from [29]. (c) Extraction enhancement and directivity via coupling to a specific mode of a hexagonal PhC. Colors correspond to various periods of PhC. SEM image shows the pillars patterned into the quantum wells structure (emission layer); reproduced with permission from [30]. (d), Directional emission from the asymmetric grooves in the metallic contact on top of the QD LED with the microscope image of the patterned structure; reproduced with permission from [31].

Although the scatterers have been shown to improve the light extraction

efficiency, the effect on the composing it carrier injection efficiency can be quite the opposite. Carrier injection and I-V characteristic of the device depend among many factors on the LED heterostructure, device design and electrical conductance. If the scatterers affect the current distribution, ohmic resistance or uniformity of spreading (e.g. patterning of the p-GaN and the current spreading layer of the lateral LED, Figure 1.1 a), the resulting current injection efficiency might experience an efficiency drop with a slope deterioration in the I-V curve [32]. However, in some LED modifications (e.g. patterned sapphire substrate in the lateral LED) scatterers are isolated from the charge injection and current distribution and, hence, have negligible effect on the injection efficiency and applied electric field [33].

Bringing nanostructuring a level deeper (into the LED device, i.e. through the MQW layers [30], Figure 1.3 c) induces a more complex phenomena coaction [34], [35]. First, this arrangement may introduce well-pronounced directional emission ([36] regular planar LED Lambertian angular profile is transformed), boost the extraction efficiency and diminish the charge polarization issues in quantum wells. However, on the downside, the etching process and lateral confinement might damage the MQW inherent architecture and therefore cause a decline in the IQE.

Manipulation of the LED emission pattern is attractive not only as an approach for higher light extraction but as a separate research direction on its own. Redistribution of emitted light and focusing of the radiated energy into a narrow solid angle is vital in many real world applications: light-fidelity (Li-Fi) devices, augmented reality (AR), virtual reality (VR), holography or efficient optical communications. One of the methods to obtain directional emission is to create a PhC array in the LED slab [22], [37]. However, it is still a complex challenge to reach high contrast between the desired emission direction and secondary unwanted lobes. Furthermore, the resulting emission pattern typically possesses partial or complete symmetries resembling those of the PhC, which are

often undesired or fully incompatible with some particular applications.

Metallic nanoparticles also have a high potential to alter the directivity. In contrast to dielectrics, some metals may exhibit surface plasmon polariton (SPP) resonances due to the negative value of the electric permittivity. These SPPs are surface propagating or localized waves (localized surface plasmon resonance (LSPR)) that occur as a result of collective oscillations of free electrons in the plasmonic material, coupled with light photons. By periodic patterning of the metallic structure with a period longer than the wavelength of light, the structure will support the diffractive modes. Using the interplay of the SPP and the periodic lattice resonances one can modify the LED radiation pattern from regular Lambertian into relatively narrow light beams [38]. The deflection angles of those are determined by the diffraction orders of the periodic structure. Moreover, by breaking the plasmonic particle symmetry one can focus the light into the specific diffractive orders. Hence, by using the asymmetric nanoparticle shapes (e.g. silver triangular prisms [29] or rectangular grooves [31], Figure 1.3 b and d correspondingly) one may concentrate the emission into a single narrow peak. On top of that, plasmonic particles can be considered as resonators, enhancing the LDOS and, therefore, boosting the Purcell factor [39] of the system, which leads to higher operation speed of the LED. While possessing so many benefits, metals in visible range have to deal with a large amount of ohmic losses (hence, the problem of heating). This is a major drawback which tremendously reduces their performance, and as a consequence leads to similar value or even lower efficiency [40] as compared to dielectric materials [41]. As a result of these constraints, dielectric materials display a high potential as an alternative to metals for LED integration, especially with the rapid development of the metasurfaces field which allow for unprecedented control of the light characteristics.

1.2.2 Metasurfaces

Technological capacities to fabricate nanoscale patterns and measure electromagnetic fields distributions with resolution much higher than the light wavelength sparked the development of nanophotonics and, in particular, metamaterials. Metamaterial (from Greek $\mu\epsilon\tau\alpha$ - beyond) is an artificially engineered material overcoming the limitations and highly outperforming the naturally occurring analogues. Optical metamaterials were largely influenced by the PhC research, however, the building block of a metamaterial is much smaller than the light wavelength. Precise engineering of these metamaterial "atoms" [42] plays a crucial role in defining the metamaterial optical properties, granting almost unlimited freedom in tailoring electromagnetic response of the system and turning any desired light properties on the output into a reality.

Historically, metamaterials, in particular optical, started to thrive with development of plasmonic nanostructures. Noble metals such as gold and silver have their plasmonic frequency in the visible range and possess high negative values of electric permittivity (ϵ). The idea to have a material with negative magnetic permeability (μ) [43] and, moreover, with negative refractive index [44] (product of electric and magnetic responses of the material), initially proposed more than 50 years ago [45], fascinated many researchers. The negative refractive index material does not naturally occur and it was a definite challenge to be solved by the emerging field of metamaterials. Initial solution was demonstrated in the microwave regime with a copper split ring resonator (SRR) [46]; later the result was transferred to the terahertz and the optical range with the use of silver double wire [47] and fishnet layered [48] structures. A possible application for this metamaterial might be the flat super-resolution lens, exploiting the evanescent fields to obtain resolution beyond the diffraction limit [44]. Another intriguing material regime is epsilon near zero (ENZ), i.e. value of electrical permittivity is close to zero [49]. This regime has several peculiarities: an almost uniform phase of optical modes inside the material,

supercoupling and weak dependence on geometry of the resonances [50]. Those effects have high perspectives for real applications in optical interconnects and communications industry. In general, calculating the inverse problem of the material parameters distribution that creates the desired light propagation is the core of the transformation optics [51]. However, control of permittivity and permeability distribution must be independent and precise, different from the conventional natural materials, making metamaterials a suitable candidate. With an engineered gradual distribution of material parameters in space (ϵ , μ), it is possible to create regions that light bypasses without perceptible wavefront or amplitude distortions and hide any desired objects inside (cloaking [51], [52]).

In many cases, where light-matter interaction is strong (generally resonant), one patterned layer of metamaterial (metasurface) is sufficient for full control of the light characteristics. A metasurface is basically a two-dimensional version of the metamaterial. The use of plasmonic resonances enable concentration of light in volumes much smaller than the light wavelength ($\sim \lambda^3 \times 10^{-7}$ [53]). In fact, those plasmonic hotspots also tremendously intensify the electric field and hence the LDOS, which can be used in a vast variety of applications. The photoluminescence signal of a single molecule can be enhanced by more than 1000 times with a bowtie gold nanoantenna [54]. Nonlinear processes have a strong dependence on the electric field density as well, hence, such applications as surface enhanced Raman spectroscopy (SERS) [55], [56], sensing and nonlinear harmonic generation [57], [58] highly benefit from the electric field enhancement in the plasmonic metasurfaces. Selective chemical reactions can be triggered or accelerated with the hot electrons transport [59] (consequence of the plasmon excitation). Polarization anisotropy can be introduced by breaking the symmetry of the unit elements in metasurface, e.g. linear polarizers and quarter waveplates formed with silver nanoslits [60]. Chiral response (different response for left and right circularly polarized light excitation) can be observed for the metasurfaces with special unit cell geometries where their mirror image cannot

be superimposed with the original [61].

It was shown that using the metasurface building blocks to locally control the phase enables formation of virtually any wavefront [62] which is restricted only to a designers' imagination. Hence, the problem formulation switched to what kind of metasurface elements will provide the sufficient phase variation for a desired wavefront map. This phase variation in the metasurface elements has to be tunable gradually and continuously for precise mapping, and moreover, it needs to cover all values from 0 to 2π for the full control of the phase. For example, a V-shaped antenna [63] (Figure 1.4) made of plasmonic materials (gold, silver) satisfied those requirements. It employed the localized surface plasmon resonance (LSPR) for phase accumulation; the V-shape was due to the fact that the electric dipole resonance produced only $0 - \pi$ phase variation (as any other resonance) and angular variation between the arms of the V-shape adds the supplementary $0 - \pi$ for the full phase coverage.

Figure 1.4: Wavefront formation in the beam deflecting metasurface supercell consisting of eight V-shaped antennas. Reproduced with permission from [63].

Another approach is to exploit the Pancharatnam-Berry geometrical phase [64], [65], [66]. Using the Jones matrix formalism [67], anisotropic scatterer can

be described by:

$$\hat{J} = \hat{R}(-\theta) \begin{pmatrix} t_o & 0 \\ 0 & t_e \end{pmatrix} \hat{R}(\theta) \quad (1.2)$$

where t_o and t_e are scattering coefficients for ordinary and extraordinary directions, $\hat{R}(-\theta) = \begin{pmatrix} \cos(\theta) & \sin(\theta) \\ -\sin(\theta) & \cos(\theta) \end{pmatrix}$ is the rotation matrix. In case of the incident light having right (left) circular polarization, the light on the output [68]:

$$E_{out}^{R(L)} = \hat{J} E_{in}^{R(L)} = \frac{t_o + t_e}{2} E_{in}^{R(L)} + \frac{t_o - t_e}{2} E_{in}^{L(R)} e^{2il\theta} \quad (1.3)$$

First term are scattered waves with same handedness as the incident light, second corresponds to the opposite with an additional (Pancharatnam-Berry) geometrical phase term. $l = -1$ for right and $l = 1$ for left circular polarization of the incident light. Typically, this approach uses some resonance of the structure (dipolar, quadrupolar, etc.) and complements the remaining $0 - \pi$ phase variation with geometrical rotation of the antenna orientation in space. In the case of the circularly polarized light incidence, the antenna rotation allows to change the relative phase shift (from the initial $\pm \frac{\pi}{2}$ in circularly polarized beam) and, hence, transforms the polarization state between two orthogonal components [69], [70] (Figure 1.5). Effectively, the phase change of the resulting output beam produced by interfering components will cover the remaining $0 - \pi$. So, equipped with means to form any wavefront, for any given application, one simply needs to define the phase distribution and map it with the metasurface.

Figure 1.5: Phase accumulation the circularly polarized beam using the Pancharatnam-Berry geometrical phase (rotation of the wave plate). Reproduced with permission from [70].

A linear phase gradient produces the beam deflection functionality, the slope of the gradient defines the deflection angle. Practically, the beam deflection functionality is implemented as a supercell with a linear phase gradient periodically repeated along the metasurface [63], [71], [72]. For the case of lens the wavefront output of the metasurface is required to have a parabolic shape in order to constructively interfere the light waves in the focus from all parts of the lens [73], [58]. A continuous uniform azimuthal phase variation from 0 to 2π in a metasurface endows a light beam with a nonzero orbital angular momentum (OAM), in particular, a topological charge of ± 1 [69], [74], depending on the rotation (left or right, i.e. clockwise or counterclockwise) of phase variation. Higher charges and more complex beams can be formed by multiple 0 to 2π azimuthal phase alteration and/or with an additional radial phase dependence. By precise engineering of the scattering response of the individual antennas (pixels) and their interference in the far field, it is possible to create a desired light distribution of intensities (holograms [75], [76]) at a specific distance from a metasurface. Spatial separation of the pixels responsible for red, green and blue image formation in the metasurface and off-axis illumination can be used for

color multiplexing and formation of the multicolor complex 3D holograms [77].

For all the wealth of potential applications plasmonic metasurfaces possess a major drawback. High value of the imaginary part of electric permittivity translates into a large amount of ohmic losses and significant heating. There are few exceptions where those losses can be used for a benefit e.g. as an absorber [78], a source of hot electrons [59] or heat [79]; however, in general, high losses result into low transmission and, hence, low efficiency of the system.

Dielectrics and semiconductors highly surpass plasmonic materials in terms of ohmic losses in the visible range. The imaginary part of the refractive index of a typical dielectric and semiconductor material is typically negligible comparing to those of plasmonic counterparts. On the contrary, the real part of refractive index for dielectrics is much higher and is shown to be sufficient for full wavefront and amplitude control of light [80]. Nonetheless, it is clear, that a higher value of refractive index allows for stronger light concentration and effortless light manipulation. Hence, high refractive index materials in visible [81], [82] such as silicon (*Si*, $n > 3.4$), titanium dioxide (*TiO₂*, $n > 2.4$), gallium nitride (*GaN*, $n > 2.4$), gallium phosphide (*GaP*, $n > 3$) become very appealing for nanophotonics and metasurfaces research. Moreover, in terms of fabrication, some of those materials (e.g. *Si*) are complementary metal-oxide semiconductor (CMOS) process compatible which most of plasmonic materials are lacking. CMOS processes are well developed in electronics industry, therefore, compatibility is a strong advantage for a real future product and mass production.

Naturally occurring materials do not exhibit a magnetic response at optical frequencies ($\mu \sim 1$). To obtain it, artificially, plasmonic materials need to be patterned into split rings (SRR) [43], [46] or other complicated shapes. Dielectric nanoparticles, on the other side, according to Mie theory [83] describing scattering properties of the spherical particles, can exhibit magnetic resonance behaviour even in subwavelength sizes. This artificial magnetic response can

be even observed implicitly from scattering of the dielectric particles [84], [85]. The essence of the optical magnetism is explained by the nature of modes excited in dielectric particles: besides the electric type of resonances (observed in plasmonics), incident light also excites an equally large family of magnetic resonances (dipole, quadrupole, octupole, etc.). Consequently, all the artificial magnetic effects observed with plasmonic nanostructures can be translated to a dielectric platform. Moreover, with magnetic type of resonances the design freedom is much larger as compared to the plasmonics and prospects of novel effects are highly promising.

Dielectric particles can enhance both electric field [86] and magnetic field [87] (>100 times). As the field enhancement directly boosts the Purcell factor [16], dielectrics acquire a broad range of potential applications. Optical transition rates of molecules and quantum dots are influenced by Purcell factor. High magnetic Purcell factor enhances magnetic transitions [88] (which commonly occur with low probability), with a potential for shaping the emission spectrum by making magnetic transitions even more probable than conventional electric transitions. Fluorescence spectroscopy and Raman spectroscopy (SERS) signals are significantly improved with the use of dielectric dimer (*Si*, *GaP*) [89], [90]. Low heat generation and temperature gradient is another advantage for sensitive organic dyes or molecules using this approach. Other nonlinear processes such as second harmonic generation (SHG) and third harmonic generation (THG) also benefit from strong electromagnetic field concentrations. Resonant pillars of GaAs exhibited 10^4 fold enhancement of SHG signal [91] and disks of Si >100 times THG enhancement [92] relative to plain, unstructured films. Sharp and well-pronounced resonances can also be used for color printing at nanoscale with a pixel size close to a light wavelength [93].

Interaction of multipoles within the nanostructures introduces many conceptually novel effects. Metasurfaces have been recently shown to exhibit polarization properties similar to classical Brewster angle effect where reflected

light becomes linearly polarized. Classical Brewster effect happens due to electric dipole excitation in the media that cannot emit along the direction of that dipole. In dielectric nanoantennas one can excite plenty of multipoles and affect the interaction between them by design in a way that some directions become strongly suppressed and polarized by destructive interference of those multipoles (electromagnetic Brewster effect [94]). Furthermore, excitation of the toroidal family of modes (displacement current flowing along the surface of the torus) may induce anapole states at specific conditions. Peculiarity of the anapole state is that it does not radiate in the far field. Initially discovered in particle physics [95] its properties are similar to the dark matter (up to date undetectable with conventional methods, however is supposed to be a major mass constituent of the universe). In optics anapole state occurs when electric and toroidal dipoles interfere destructively in the nanoantenna and can be implicitly observed as a sharp spectral drop in scattering [96], [97]. Due to strong nearfield enhancement and high quality factor the anapole mode can be used for Raman scattering enhancement [98] and lasing [99].

The bound state in the continuum (BIC) is another example of precise band or mode engineering in the metasurfaces. Light coupled to BICs cannot escape into environment as it is topology, symmetry or otherwise protected [100]. BICs have incredibly large quality factors reaching infinity in theory, however even in experiments it can be effectively above 10^6 [101]. Applications of BICs are remarkably broad: lasing [102], [103], [104], polarization conversion [105], polarization vortex generation [106], sensing and spectroscopy [107]. By overlapping two photonic bands of opposite curvature in the band structure at the Γ point (Dirac cone) it is possible to obtain the near zero index (NZI) material properties [108] [109]. NZI material has similar properties to ENZ, however due to impedance matching condition (both ϵ and μ are near zero) reflection from this material is strongly suppressed which is highly desirable for many applications. Practical implementation of the NZI material was demonstrated with layered

Si wires structure in glass matrix [110]. NZI material enhanced the Quantum dot (QD) emission and introduced strong directivity in the radiation pattern.

The interference of the electric and magnetic dipole resonances can lead to strong directional scattering, demonstrated on silicon spheres [111] and GaAs pillars [112]. If the electric dipole amplitude value equals the magnetic dipole amplitude in the nanoparticle with dipoles oscillating in-phase (constructive interference in forward direction and destructive in backward), the backwards scattering becomes negligible [113]. This situation is called the first Kerker condition [114]. Whereas the out-of-phase interference of electric and magnetic dipoles may almost fully suppress the forward scattering (the second Kerker condition). In the vicinity of the first Kerker condition, additionally, when electric and magnetic resonances spectrally overlap (their maxima coincide), the resonances interaction can be used for phase accumulation in metasurfaces. In fact, each of the resonances (magnetic and electric) provides a $0 - \pi$ phase coverage, with both of them involved any phase front can be generated. This approach corresponds to the concept of Huygens' metasurfaces [115], [116] where reflection is strongly suppressed due to metasurface elements (low back scattering). The first Kerker condition can be achieved by varying diameter in cylindrical shape particles [117] and hence changing the spectral position of the excited electric and magnetic dipole modes. Based on this, metasurfaces with beam deflection functionality [118] and holograms [119] were demonstrated. Holographic images in this case are formed as a result of interference from all the pixels of the metasurface hologram at a specifically designed distance and wavelength of operation. The variation of period in parameter space might provide similar means of phase control (phase shift between multipole resonances) in the case of strong interaction between particles (i.e. interparticle gap comparable to or smaller than particle size and wavelength of excitation). Based on this approach, the vortex beam (with nonzero OAM) generating metasurface was demonstrated [120].

Figure 1.6: Phase accumulation using interacting multipoles approach (electric and magnetic dipoles). a, Numerical simulations of transmission and phase for the *Si* disk system. Top panel shows electric and magnetic dipole contributions in modes excited in *Si* disk array calculated by multipole decomposition. b, Electric and magnetic field distributions at the wavelength of maximum transmission (electric and magnetic dipoles are in phase). Reproduced with permission from [118].

The Pancharatnam-Berry phase concept is also popular to achieve phase control in dielectric metasurfaces [70]. It is much more efficient than in metals due to low losses and based on rotation of the individual elements in metasurface to gain additional phase coverage for circularly polarized excitation beam [66]. With the use of TiO_2 nanofins, research groups were able to demonstrate plenty of functional applications: metalens at red, green, blue (RGB) wavelengths [121], high numerical aperture (NA) lenses [122], converter of spin orbital momentum (SOM) to orbital angular momentum (OAM), efficient holograms [123]. Moreover, by placing two blocks (nanofins) inside one-unit cell and varying of the size of each of them it is possible to compensate the chromatic dispersion and construct achromatic metasurface devices. In experiment, the achromatic lens was presented [124], however, the efficiency of the device was

much lower than single wavelength operation ($<20\%$) and NA was around 0.2 in air, which is insufficient for most of real life applications.

Arguably, the most prominent and efficient way of phase accumulation is the waveguiding approach. Discovered and demonstrated almost twenty years ago with TiO_2 binary blazed grating, this concept used large aspect ratio ($\sim 10 : 1$) blocks as unit cell elements [125]. Unlike the other approaches this method relies on phase accumulation along the nanoparticle (Figure 1.7, [126]), without explicitly exploiting the resonant nature of the system. Furthermore, if the unit cell particle is symmetrical (cylinder), the polarization insensitive response can be effortlessly obtained. This method has got a second breath with current development of nanolithography and fabrication tools. Recent experimental demonstrations with Si and TiO_2 displayed the wealth of the practical applications of the waveguiding approach: beam deflectors, polarization beam splitters [127], high NA [128] and achromatic [129] lenses, polarimeters [130], holograms [131], OAM beam generators, endoscopes [132] etc. Efficiency of this approach is generally high and can reach above 90%. Moreover, through stacking of several metasurfaces it is possible to reduce the chromatic and other aberrations effect with the increased angle of view [133].

Figure 1.7: Phase accumulation using waveguiding approach. a, Effective refractive index as a function of dielectric ridge width, inset shows the scheme of the dielectric ridge system. b, Electric field distribution (real E_y) showing different phase accumulation in ridges of different width. Reproduced with permission from [126].

1.3 Dissertation objectives

The main focus of this dissertation is the development of novel dielectric and semiconductor metasurfaces and III-V LED devices with engineered light characteristics (radiation pattern, wavefront profile, polarization) by means of the fore-mentioned metasurfaces integrated inside the device. This includes:

- New metasurface designs for various dielectric and semiconductor materials (*Si*, *TiO₂*, *GaN*), their fabrication and optical characterization.
- Demonstration of advanced functionalities such as: high angle beam steering, polarizers, beam splitters, high NA lens, vortex beams.
- Integration of metasurfaces with LED; optical and electrical pumping of the devices.
- Study of the emission enhancement in LEDs integrated with metasurfaces.

Ultimate goal of this dissertation is a compact and novel LED device with light emission characteristics on demand by means of dielectric metasurfaces.

1.4 Major contribution of the Dissertation

1. Highly efficient metasurfaces based on dielectric and semiconductor materials (*Si*, *TiO₂*, *GaN*) were designed and fabricated. Functionalities include a beam deflector and a polarization beam splitter (PBS).
2. *GaN* metasurfaces were etched directly into the regular *GaN* slab. Despite the low index contrast between metasurface and the substrate (both *GaN*), the metasurfaces exhibited high efficiencies within the designed part of the visible range. Beam deflection and polarization beam splitting functionalities were demonstrated.

3. A novel class of metasurfaces with asymmetric radiation profile was engineered. Efficiencies of directing the light into the desired diffractive order reach 99%. Ultra-high angle beam deflection metasurfaces operating in the visible regime were demonstrated: TiO_2 for blue and green, Si for red part of the spectra.
4. Near-unity Numerical Aperture (NA) lens was designed and fabricated for Si , demonstrating one of the promising metasurface applications of the asymmetric designer antennas approach.
5. An external and internal cavity design solution for LED and metasurface compact integration was proposed and engineered. The efficient transformation of the Lambertian radiation pattern into plane wave like radiation suitable for the metasurface excitation was experimentally demonstrated.
6. Advanced functionalities by means of Si metasurfaces, viz. beam deflection and vortex beam generation, integrated with GaP LED were shown with optical pumping.
7. The dielectric bragg external cavity was integrated with a GaN LED. A beam deflection functionality was experimentally demonstrated using TiO_2 metasurfaces with electrically driven GaN -based LED.

Chapter 2

Advanced metasurfaces

The material in this chapter was in part presented in [\[134\]](#), [\[135\]](#).

2.1 High angle beam deflection

The redirection of a light beam is one of the simplest forms of beam manipulation. Nonetheless, it is also one of the most fundamental and crucial essentials required in any optical system. In conventional optics a simple prism has the capacity to direct the light at the desired angle. From a simple wavefront consideration, the incoming plane wave experiences different phase delays depending on the thickness of the prism at a specific location (Figure 2.1 a). The resulting wavefront becomes tilted, whereas the normal to the wavefront determines the new direction of the light beam propagation. The beam deflecting metasurfaces mimic the wavefront tilt in the prism, with an exception of discretization added to the procedure. The continuous tilt in the projected wavefront is approximated with a phase step function induced by individual elements of the metasurface. Figure 2.1 b illustrates this principle of the phase mapping approach [\[62\]](#) where the output desired wavefront is mapped with several metasurface unit blocks. Each of the unit cell elements imposes its own phase shift. Combined into a supercell, they can be used to form intricate wavefronts with

virtually any complexity. The individual unit blocks or metasurface particles and their periodicity are required to be subwavelength (with respect to excitation wavelength of light) to avoid diffraction on individual particles. Otherwise the phase mapping approach becomes invalid. If the operation wavelength of interest is enclosed in the visible range, the particle sizes cannot exceed several hundreds of nanometers. Hence, they can be referred to as nanoparticles. Figure 2.1 b is a schematics of a typical supercell in a beam deflecting metasurface which is periodically repeated along the surface to cover the area of excitation, using the 2π periodicity of the phase.

Figure 2.1: (a) Conventional optical element - prism tilts the wavefront of the incoming light, resulting in an optical beam deflection, red to blue color map corresponds to the phase variation from 0 to 2π ; (b) a phase mapping approach in metasurfaces: tilted phase wavefront is sampled with discrete number of elements (seven cylindrical nanoantennas as an example). Continued on the next page.

Figure 2.1: (c) a high angle beam deflection issue: when the number of mapping elements is small (only two), beam deflection becomes inefficient and symmetrical in both directions. Green frames denote unit cells deflecting the incident beam into opposite directions.

Despite the strength of the phase mapping approach, which is generally used in metasurfaces for wavefront formation, some challenges still arise, attracting more attention into the field. One of the challenges originates from the discretization step: for a metasurface to be efficient, the number of elements mapping the desired phase needs to be sufficient. In signal processing this issue has a direct analogue - the Nyquist-Shannon-Kotelnikov theorem [136], describing the conditions for the sampling of continuous-time signals (as the wavefront map) with the discrete-time signals (as the metasurface elements). Whereas in signal processing the sufficient sample rate is double the frequency of the signal, metasurfaces based on two-element supercell demonstrate very low efficiency. Further details are clarified in Figure 2.1c on the simple example of the high angle beam deflecting metasurface. The angle of light beam deflection is defined by the length (period) of the metasurface supercell periodically repeated along the surface. The precise value of the angle can be found from the basic diffraction equation (excitation is at normal incidence):

$$\sin(\phi) = \frac{m\lambda}{\text{period}} \quad (2.1)$$

where ϕ is the deflection angle, measured from the surface normal; λ is the light wavelength; period refers to size of the supercell, m is the diffraction order number. Hence, longer periods of the supercell correspond to low deflection angles and short periods, i.e. small supercells, correspond to higher angles. Typical sizes of the dielectric metasurface elements are hundreds of nanometers, in order to use any of the phase accumulation mechanisms described in the metasurface introduction section (waveguiding, Huygens, Pancharatnam-Berry).

Therefore, the wavefront profile for high angle beam deflection can only be mapped with a few nanoparticles. Figure 2.1c illustrates the critical case where the wavefront is mapped with only two elements in the supercell periodically repeated along the metasurface. Angles of deflection corresponding to this case can reach values close to 90° (depending on the wavelength-supercell period ratio, Equation 2.1). From the figure it is clear that the grating loses its asymmetry when the number of mapping elements is low: the grating is completely symmetric relative to the center. Therefore, two types of supercells marked in Figure 2.1c with green frames will bend the incoming light into the opposite directions with equal efficiency, hence the desired directivity is completely lost. Moreover, to be precise, the incoming light will be redistributed between all the existing diffraction orders (T_{-1} , T_{+1} , etc) including the zero order (T_0) in transmission and reflection (R_{-1} , R_{+1} , R_0 , etc), which makes the portion of light directed into the correct order unsatisfactory small.

2.2 Concept of a nanoantenna combined with a diffraction grating

As explained above, one of the phase mapping approach limitations in metasurfaces is associated with the discretization: if the target wavefront is mapped with a low number of metasurface nanoparticles (in the case of the high angle beam deflection), the efficiency drastically drops. Even in the limiting case of continuous mapping, it has been shown that extreme care should be taken when designing the impedance of the device, both in transmissive [137] and reflective [138], [139] configurations, in which simultaneous control of the amplitude and phase profiles is necessary to achieve high efficiencies. Based on these ideas, recent experimental works [140], [141] have shown remarkable efficiencies, even after discretization of the ideal impedance profile. Computer algorithms (particle swarm, genetic) have also been recently used to generate supercell geometries

achieving high experimental efficiencies in the near-IR spectral range [142]. An alternative concept suggested in this dissertation and concomitant journal publications [134], [135] allows for efficient, high-angle light deflection to cover any desired wavelengths in the visible range. More broadly, the concept can be applied to control the light energy redistribution in diffraction gratings and does not rely on the commonly used phase/amplitude mapping. Another advantage is the intuitiveness of the approach. It is based on engineering of scattering directivity patterns of each of the unit cells of the diffraction grating using its antenna properties. This allows for suppression and/or enhancement of the selected diffraction orders at will and, as a particular example, channelling of the incident light energy into one particular diffraction order in transmission or reflection, causing the light bending effect. Related to this approach, recent theoretical studies have shown that efficiencies close to 100% for high-angle light bending can be achieved if polarizabilities of the grating components are appropriately selected [143]. In this dissertation experimental experimental results show that nanoantenna characteristics of grating elements indeed can be used to accurately engineer energy distribution between the diffractive orders and achieve highly efficient high-angle light bending through the whole visible spectral range.

The concept is based on the combination of the diffraction grating concepts and nanoantennas with angle-suppressed, asymmetric scattering patterns. Recently, it has been shown that isolated, dielectric asymmetric antennas or dimers [144] allow to direct scattered light into a specific solid angle and may exhibit strong radiation directivity [145–148] associated with the complex interaction of modes or multipoles excited inside the constituent parts. In this chapter it will be revealed that, when these antennas are arranged in diffractive arrays, the precise tailoring of their scattering patterns allows to control the power distribution among the different diffraction orders supported by the grating (both in reflection and transmission).

Figure 2.2 schematically depicts the key concept behind the working principle of the device. The well-known grating equation 2.1 determines the specific angles of the diffractive orders for any given period and operating wavelength. It, however, does not provide any information about the power ratios. The energy distribution between the orders is specified by the scattering efficiencies into corresponding directions of the elements composing the grating, with the only exception of direct transmission that is determined by the interference between the incoming wave and the forward scattering. Under normal incidence, a symmetric scattering response would lead to an equal distribution of power into positive and negative orders from the grating, as shown in Figure 2.2a. Therefore, in order to break this symmetry, one needs inclusions producing asymmetric scattering patterns. Moreover, if one would like to concentrate the energy into a single order, the scattering pattern should further be suppressed in those directions corresponding to all the rest of the grating orders. One of the simplest systems, both from the theoretical and experimental points of view, that has been shown to produce this kind of patterns is the asymmetric dimer [144], [145], [148]. By precise engineering of the dimer geometry one can obtain scattering only into the selected angles with suppression of all the other directions.

Figure 2.2: Device working principle. (a) Allowed diffraction orders and illumination conditions of the proposed device. The power distribution between orders is related to the scattering of the unit cells and, under normal incidence, is symmetrical for gratings with symmetrical inclusions. Red arrow denotes the incidence direction. (b) Example of asymmetric inclusion (an asymmetric dimer) providing highly directional scattering toward the desired diffraction order direction and suppressed elsewhere. (c) Output energy distribution corresponding to the convolution of a single element scattering pattern and the grating diffraction directions. Reproduced with permission from [134].

In Figure 2.2b the scattering pattern of one elementary dimer in the grating array is considered, with all the interactions with the neighboring elements and the incoming wave taken into account. The scattering from the structure has

been optimized in such a way that it is maximal toward the desired grating order (T_{-1} in this case) and suppressed in the directions of all other transmitted orders (T_0 and T_{+1}) and reflection orders (R_0 , R_{-1} , R_{+1} etc.). When summing up the scattering contributions from all the dimers in the grating, most of the energy is concentrated into the T_{-1} diffraction order, as shown in Figure 2.2c. Hence, using this concept, the incoming power can be precisely distributed at will into the different diffraction directions by accurate design of the grating element scattering pattern. In the case of Figure 2.2, and in the following, 2D gratings are considered and the number of diffractive orders in transmission are restricted to 3 by choosing a diffractive period along the x -direction and subdiffractive period in the y -direction. When the constituent elements are properly engineered, these gratings then act as beam bending devices with the minimum number of elements in the grating unit cell. The proposed design principle can be applied for transmission and reflection regimes or for a combination of both. Furthermore, although the consideration involves only the case of three diffractive orders for simplicity, the proposed concept can be applied to a higher number of diffraction orders.

2.3 Experimental demonstration of high angle beam deflection with Si nanoantennas

To demonstrate the feasibility of this approach, we designed several gratings for beam bending based on asymmetric nanoparticles, supported by a silica substrate. *Si* asymmetric dimer was chosen as one of the first examples due to the simplicity of the system and high development of the *Si* fabrication process. *Si* also possesses high refractive index ($n \sim 3.66$ at 715 nm wavelength) and relatively low loss in the red part of the visible spectrum ($\kappa \sim 0.01$ at 715 nm wavelength). The schematics of the supercell in the array under study is shown in Figure 2.3A. It was optimized to deflect normally incident light from the

glass substrate (715 nm wavelength) at an angle of 82° . Full wave numerical simulations of diffraction efficiencies in periodic arrays of nanoantennas were performed using a finite difference time domain (FDTD)-based commercial software (Lumerical FDTD). The simulation domain consisted of a single unit cell with in-plane Bloch boundary conditions, numerically representing infinite periodic metasurface array. SiO_2 substrate was modeled as a medium with a constant refractive index of 1.46, the nanoantennas were modeled as cylinders with the appropriate dimensions with material parameters taken from experimental ellipsometric measurements. A power monitor was applied in the vertical direction (z -direction) to collect the diffracted orders in transmission. Plane wave excitation was used to excite the system with an appropriate polarization. The injection angle was set to zero to simulate a normally incident wave. All FDTD simulations were also checked with a finite element method (FEM)-based commercial software (Comsol Multiphysics version 5.2).

Figure 2.3: (A) Schematic description of the unit cell of the array of asymmetric dimers producing energy concentration into the T_{+1} diffraction order leading to light bending at 82° for plane waves normally incident from the substrate side at a wavelength of 715 nm. Simulated out-of-plane components of the (B) electric and (C) magnetic field when the array is illuminated by a normally incident plane wave with the magnetic (B) or electric (C) field along the axis of the dimer. Reproduced with permission from [135].

The supercell consists of an asymmetric dimer nanoantenna made of silicon (Si) nanodisks of height $H = 250$ nm placed on top of a silicon dioxide (SiO_2) substrate and surrounded by air. The nanodisk diameters are $D_1 = 150$ nm and $D_2 = 190$ nm, and the gap is $g = 50$ nm. The diffractive and nondiffractive periods

of the rectangular lattice are $P_d = 721$ nm and $P_{nd} = 260$ nm, respectively. The former is selected in such a way to provide the first diffractive order going to air at 82° to the normal, and thus, it is fixed for a given wavelength and angle of diffraction. The latter is chosen to be small enough to avoid diffraction in air in this direction and keep a high array density.

Panels B and C of Figure 2.3 show the simulated field distribution when the array is illuminated by s- or p-polarized plane waves normally incident from the substrate side at the operating wavelength. We define the s(p)-polarization as that with the magnetic (electric) field directed along the axis of the dimer. As seen from the figures, the energy is mainly concentrated into the T_{+1} order. This is evidenced by the plane wave emerging from the array tilted at 82° with respect to its normal with minimal distortion. In the simulations, more than 88% of the transmitted energy is concentrated into the correct diffraction order for s-polarized light and more than 82% for p-polarized light.

Metasurface fabrication procedure is described in Methods. The simulated and measured spectral dependencies of diffraction efficiencies of this array in Figure 2.4. Panels A and B show the numerical simulations of the absolute power directed into the desired diffraction order normalized to the incidence power (absolute diffraction efficiencies) for p- and s-polarized light, correspondingly.

For measuring large-angle bending samples, for which the diffraction angles were larger than the maximum acceptance angle of our collection microscope objectives (NA=0.95), a home-built free-space microscopy setup was used as described in Methods. The measured results of the absolute diffraction efficiencies, in good agreement with the numerical simulations, are given in Figure 2.4 C and D. For the wavelength of interest (715 nm) the simulated absolute diffraction efficiencies are 42% and 18% for p- and s-polarization; experimental measurements give 35% and 32%, correspondingly. Panels E and F demonstrate the relative efficiencies (absolute power into desired diffraction order, normalized to the transmitted power) for numerical simulations (82%

for p- and 88% for s-polarization) and the experiment (75% and 70%). While these values are still far from the ideal 100% efficiency, it should be mentioned that there is no fundamental limitation to the efficiencies that can be achieved using the method described here. We foresee that further optimization of the nanoantenna scattering patterns could lead to designs beating the one reported here, mainly constrained by polarization insensitivity requirements, as has been recently theoretically reported using polarizability considerations [[143](#)].

Figure 2.4: Numerical and experimental characterization of silicon dimer arrays bending light at 82° . Continued on the following page.

Figure 2.4: Simulated absolute diffraction efficiencies under normally incident light illumination with (A) p- or (B) s-polarization as defined in the text. (E) Simulated relative bending efficiency of the array, representing the amount of power into the desired order relative to the total transmitted one. Measured absolute diffraction efficiencies under normally incident light illumination with (C) p- or (D) s-polarization. The solid lines correspond to back-focal plane measurements, while those with symbols correspond to measurements taken using a free-space microscopy setup. (F) Measured relative bending efficiency of the array. Scanning electron microscopy images of the fabricated sample under (G) top and (H) tilted views. The scale bars correspond to 500 nm. The scale bars correspond to 500 nm. Reproduced with permission from [135].

2.4 Near-unity numerical aperture lens

On the basis of the concept presented above, a spherical lens is designed to have a NA of ~ 0.99 at 715 nm wavelength. This corresponds to a maximum bending angle of 82° , realized with the system described above. The lens has a total diameter d of $600 \mu m$ and a focal distance f of $\sim 42 \mu m$. The nanoantenna inclusions and unit cells on the different areas of the lens are designed to generate different bending angles corresponding to their radial positions within the lens, thus approximating the ideal parabolic phase profile to a piece-wise linear one, sampled in 1° steps. The central region of the lens, with a diameter of $13.5 \mu m$, is kept empty in this proof-of-concept demonstration.

All angles spanning from 82° to 55° can be achieved with the asymmetric dimer design described above only changing the array period, with efficiencies that increase as the angle decreases. Angles ranging from 54° to 36° are achieved using asymmetric trimer nanoantennas. The diameters of the cylinders are $D_1 = 130$ nm, $D_2 = 150$ nm, and $D_3 = 190$ nm, and the gaps are $g_{12} = g_{23} = 50$ nm (see Figure 2.5). Angles between 35° and 31° are achieved with quadrumers adding another disk with D values of 110 nm (for 35° and 34°) and 120 nm (for 33° to

31°) and changing the interparticle gaps to 60 nm (see Figure 2.6). In all cases, the nondiffractive period is kept constant ($P_{nd} = 260$ nm) while the diffractive period P_d is changed to generate the first diffraction order at the corresponding angle. The larger number of particles in nanoantennas designed for smaller bending angles is added to make their scattering diagram more aligned toward the required bending direction and to minimize their scattering in directions of additional diffractive orders opening in the substrate.

Figure 2.5: Numerical and experimental characterization of silicon trimer arrays bending light at 40 degrees. Continued on the following page.

Figure 2.5: Simulated absolute diffraction efficiencies under normally incident light illumination with (A) p-or (B) s-polarization as defined in the text. (E) Simulated relative bending efficiency of the array, representing the amount of power into the desired order relative to the total transmitted one. Measured absolute diffraction efficiencies under normally incident light illumination with (C) p-or (D) s-polarization. The solid lines correspond to back-focal plane measurements, while those with symbols correspond to measurements taken using a free-space microscopy setup. (F) Measured relative bending efficiency of the array. Scanning electron microscopy images of the fabricated sample under (G) top and (H) tilted views. The scale bars correspond to $1\ \mu m$. Reproduced with permission from [135].

Characterization of two examples, corresponding to the arrays bending light at 40° and 35° , can be found in Figures 2.5 and 2.6, respectively. Spectrally resolved back-focal plane imaging measurements were used to characterize the diffraction efficiencies of these samples as described in Methods. The values shown are representative of the whole range of angles, with diffraction efficiencies into the T+1 order exceeding 60% at the operating wavelength, representing more than 85% of the total transmitted power. For angles less than 30° , the usual phase mapping approach can be applied. In case of these small angles, the nanoantenna approach becomes impractical because it requires a complex design optimization to minimize the nanoantenna scattering into all the opened diffraction directions, while the phase mapping approach is simpler from a design point of view and can provide good bending efficiencies.

Figure 2.6: Numerical and experimental characterization of silicon quadrumer arrays bending light at 35 degrees. Continued on the following page.

Figure 2.6: Simulated absolute diffraction efficiencies under normally incident light illumination with (A) p-or (B) s-polarization as defined in the text. (E) Simulated relative bending efficiency of the array, representing the amount of power into the desired order relative to the total transmitted one. Measured absolute diffraction efficiencies under normally incident light illumination with (C) p-or (D) s-polarization. The solid lines correspond to back-focal plane measurements, while those with symbols correspond to measurements taken using a free-space microscopy setup. (F) Measured relative bending efficiency of the array. Scanning electron microscopy images of the fabricated sample under (G) top and (H) tilted views. The scale bars correspond to $1\ \mu\text{m}$. Reproduced with permission from [135].

The lens was fabricated using standard nanofabrication tools, namely, electron beam lithography (EBL) as described in Methods. Scanning electron microscope (SEM) images of the fabricated sample with different magnifications are shown in Figure 2.7 A-C. To characterize its focusing capability, we use the experimental setup schematically depicted in Figure 2.8. The results reveal the generation of a slightly elongated focal spot at a distance of approximately 42 - 47 μm from the lens, as shown in panels D and E of Figure 2.7. This implies an experimental NA of at least ~ 0.94 of metalens - close to NA of the Nikon objective (NA=0.95) it was measured with. The elongated focus profile can be explained by the $\sim 10\ \text{nm}$ bandwidth of the filtered super-continuum laser used for these measurements and associated dispersion effects. The measurements also reveal a highly symmetric intensity distribution in the focal plane ($x - y$ plane), shown in Figure 2.7 F, and a very low level of background. Figure 2.7 G shows a cut across the x -axis passing through the focus, together with the fitting to the Airy pattern, corresponding to a diffraction-limited system. The coefficient of determination of the fit is 0.93, which gives us a measure of the quality of image formation.

Figure 2.7: Experimental characterization of the high-NA flat lens. SEM images of the fabricated sample showing (A) a low-magnification image of the central part, (B) a low-magnification view of the small-angle bending part of the lens, and (C) a high-magnification view of the large-angle bending part of the lens. The scale bars represent $10\ \mu\text{m}$, $5\ \mu\text{m}$, and $500\ \text{nm}$, respectively. (D) Normalized measured intensity map along the x -axis as a function of the distance to the lens, z , around the lens focus. (E) Normalized measured intensity profile along the axis of the lens as a function of the distance, z , around the lens focus. (F) Normalized measured intensity map in the focal plane ($z = 45\ \mu\text{m}$, corresponding to 0.989 NA). (G) Intensity profile along the x -axis, passing through the focus, together with the fitting to the Airy profile. Reproduced with permission from [135].

Figure 2.8: Experimental setup used for the optical characterization of the fabricated flat lens. Reproduced with permission from [135].

The measured full width at half maximum (FWHM) along this cut is 385 nm, corresponding to an NA of around 0.935, which is at the limit of resolution of our measurement system. Note that, because of the limited numerical aperture of the objective (NA_{obj}) used to measure the focal spot (Nikon 100x, $NA = 0.95$), for a diffraction-limited system, the minimum FWHM that can be measured with this setup would ideally be $\frac{\lambda}{2NA_{obj}} \sim 376$ nm. This value, however, is not met in commercial lenses at all wavelengths, as was recently reported [121], in accordance with the slightly larger spot size measured in our experiment.

Using the alternative definition based on the Airy radius obtained from the fitting, the obtained NA is 0.944, in close agreement with the result obtained from the FWHM.

Next, we provide further proof of the limitation of the measured metalens NA imposed by the commercial objective lens used for metalens measurements. We compare the back-focal plane images obtained through this commercial objective lens (NA=0.95) from the focal spot created by the flat lens with that from a strongly scattering medium placed at its focus, which generates a random distribution of wavevectors. This comparison can be found in Figure 2.9, together with the back-focal plane images from focal spots generated by commercial objective lenses with NAs of 0.8 and 0.9.

Figure 2.9: Comparison of the back-focal plane images collected with a commercial objective lens with a $\text{NA}=0.95$ (Nikon 100x) from different sources placed at its focus. (A) Back-focal image from a focal spot generated with a commercial objective lens with $\text{NA}=0.8$ (Zeiss 50x). (B) Back-focal image from a focal spot generated with a commercial objective lens with $\text{NA}=0.9$ (Zeiss 100x). (C) Back-focal image from a strongly scattering medium (a tissue) placed at the focus of the objective. (D) Back-focal image from a focal spot generated with the proposed metalens. In both cases (C) and (D) the generated k-vectors reach the limit of collection of the 0.95 NA commercial lens. The bright ring in (D) corresponds to the signal coming from a large lens area responsible for high-angle bending and squeezed into a narrow ring in the back focal plane image. Reproduced with permission from [135].

By integrating the intensity distribution in the focal plane (we restrict the integration of the intensity to the area shown in Figure 2.7 F), we found that only around 10% of the incident power is focused by the lens. Taking into account the numerically calculated diffraction efficiencies for each individual Fresnel zones in the lens and their area in the lens, we found that the expected total efficiency for the lens, computed as a weighted average taking into account both polarizations, should be around 37%. We attribute the smaller experimental value obtained to the non-optimized packing density of the bending arrays within the circular lens and to observed deviations in the nanoparticle sizes from the designed ones, causing the drop in efficiency as compared to that of simulations. Both of these factors, however, may be circumvented with appropriate design and fabrication corrections, and we feel, nevertheless, that the values obtained are reasonable for a proof-of-concept system.

We also point out that, with further optimization of the nanoantennas corresponding to the different Fresnel zones, designing a metalens with near-unity efficiency is, in principle, possible. Indeed, in recent works, several groups have reported strategies for nanoantenna optimization [149] as well as designs and design principles of so-called metagratings, in which the reported efficiencies reach values close to 100% [143], [150]. We should mention though that these works deal with systems exhibiting strong polarization dependence and there is still a need for further research to arrive at a satisfactory high-efficiency solution for polarization insensitivity required for metalens design. A comparison of the focusing characteristics of the fabricated sample and a commercial objective lens (Zeiss 100x, NA = 0.9) can be found in Figure 2.10, showing a clearly smaller FWHM of the focal spot in the case of the presented flat lens and comparable focus quality characteristics.

Figure 2.10: Comparison of the focus of the fabricated flat lens with that of a commercial objective lens (Zeiss 100x, NA=0.9). The plot shows the normalized intensity profile along the x- axis passing through the focal spot (the measurements were performed with a commercial objective lens Nikon x100 with NA=0.95). Reproduced with permission from [135].

To prove that the NA of the fabricated flat lens indeed reaches its designed value of 0.99, we performed an additional experiment in which the lens is used to collect light from a single subdiffractive scatterer located at its focus. In the experiment, the scatterer is a small dust particle deposited on top of a triangular prism. The particle is excited by the evanescent fields generated in a total internal reflection configuration, as shown in Figure 2.11 A. In this situation, and upon unpolarized excitation, the dust particle generates scattering in all directions and, thus, the full aperture of the lens can be probed while avoiding any other source of illumination of the lens. When the particle is placed at the focus of the metalens and illuminated at the operating wavelength (715 nm), the scattered light collected by the metalens emerges as a collimated beam propagating parallel to its optical axis, as shown schematically in Figure 2.11 A. This is subsequently imaged using a low-NA objective (Nikon 10x, NA = 0.3) to collect, in good approximation, only those rays that are parallel to the optical axis. The collected normalized intensity distribution is shown in 2.11 B. The high-intensity area in the image coincides with the physical size of the lens, therefore demonstrating

that the full area of the lens is functional.

Figure 2.11: Demonstration of the numerical aperture of the metalens by collecting light from a single, subdiffractive particle located at its focus. (A) Schematic of the experimental setup. (B) Normalized intensity of the collimated beam emerging from the metalens. The physical size of the lens coincides with the high-intensity area in the image. Reproduced with permission from [135].

Similar images were collected at different distances from the lens, showing similar results and therefore corroborating that the emergent beam is fully parallel to the optical axis. This, in turn, demonstrates that the lens aperture coincides with its physical size and that the corresponding NA meets indeed the design value of 0.99. As a comparison, we use the metalens to collect light from the focal spot generated by a commercial objective lens with an NA of 0.95. In this situation, only a part of the lens, corresponding to collection angles less than 72° , is functional and/or illuminated, the size of this illuminated area being much smaller than the full size of the lens, in very good agreement with theoretical

estimations. These results, presented in comparison with those from the small scatter, can be found in Figure 2.12.

Figure 2.12: Comparison of the functional area (numerical aperture) of the metalens when it is used to collect light from a small scatterer located at its focus and from the focal spot generated using a commercial objective lens with NA=0.95. (a) Normalized intensity of the beam emerging from the metalens when collecting light from a small sub-diffractive scatterer located at its focus. (b) Normalized intensity of the beam emerging from the metalens when collecting light from the focal spot generated by a commercial objective lens with NA=0.95 (Nikon 100x). The white dashed lines in both plots indicate the edge of the metalens (corresponding to NA=0.99). The cyan, dot-dashed line in panel (b) represents the edge of the functional area corresponding to NA=0.95 calculated using simple geometric optics. Reproduced with permission from [135].

Finally, the designed metalens was used to characterize the photoluminescence (PL) from nitrogen-vacancy (NV) centers in a nanodiamond (ND). This part of work was done in collaboration with the group of L. Krivitsky, further details can be found in [135].

2.5 Broadband visible light high angle beam bending with TiO_2 nanoantennas

Here we further elaborate on this new concept of nanoantenna and diffraction grating combination (Figure 2.2) and show that efficient high angle light bending in the blue and green parts of the visible spectrum is possible using titanium dioxide nanoantenna arrays with engineered scattering patterns. Moreover, we go a step beyond and experimentally demonstrate that nanoantenna characteristics of grating elements indeed can be used to accurately engineer energy distribution between the diffractive orders and achieve highly efficient high-angle light bending through the whole visible spectral range.

To demonstrate the feasibility of this approach for broadband operation, we designed several gratings for beam bending based on asymmetric TiO_2 nanoparticles, supported by a silica substrate. TiO_2 is chosen due to its relatively high refractive index (~ 2.5) and low losses in the visible spectral range. We start by demonstrating moderate angle beam bending ($\sim 53^\circ$) using asymmetric dimers. The structure is optimized to work with green light at $\lambda = 532$ nm. The asymmetric dimer consists of two cylinders with elliptical cross sections (as schematically shown in the inset of Figure 2.13 a) and a height $H = 305$ nm. The designed sizes for the major and minor axes of the larger cylinder are $D_{ly} = 280$ nm and $D_{lx} = 200$ nm, directed along the y- and x-axis, respectively. For the smaller cylinder the designed sizes are $D_{sy} = 200$ nm and $D_{sx} = 150$ nm. The interparticle gap is 50 nm. A subdiffractive period of 350 nm is chosen along the y-direction, while the period defining the beam bending angle, along the x-direction, is chosen as 666 nm, corresponding to a bending angle of 53° at the operating wavelength. We calculated the energy distribution between the supported diffraction orders by means of full numerical finite difference time domain simulations (Lumerical FDTD, the incidence and polarization being indicated in the inset of the Figure 2.13 a). The simulated results of transmission

into the different orders, together with the total transmission, are shown in Figure 2.13 a. The transmission spectrum shows that, at the operating wavelength, most of the energy is channelled into the desired T_{-1} order, reaching an efficiency of $\sim 85\%$ with respect to the incident power and representing more than 90% of the total transmission.

Figure 2.13: (a) FDTD simulations of transmission distribution between the diffractive orders; black dashed line indicates the design operation wavelength, inset shows schematics of the structure designed for 53° bending at 532 nm wavelength. (b) Calculated far-field diagram of a single dimer in the grating at 532 nm (red). Dashed lines denote the diffraction directions. High transmission and bending efficiency are obtained by suppression of radiation into undesired orders not only in transmission but in reflection as well. Simulation results are shown for light polarized along the dimer axis. Reproduced with permission from [134].

Moreover, simulations show a relatively broad spectral range (of around 50 nm, corresponding to $\sim 10\%$ of the central wavelength) in which the total transmission remains above 80% and transmission into the desired order above 60%. Figure 2.13 b shows the calculated far-field pattern of one dimer unit

cell in the grating at the operating wavelength (532 nm). It reveals a strong directivity toward the desired order, with significant suppression of scattering into the remaining diffraction orders, not only in transmission but, additionally, in reflection (indicated by the dashed lines). This explains the low reflectivity of the system and the efficient channelling of energy into the desired order. It is important to remark that, when summing up all the dimers in the grating, the system is only allowed to radiate into the discrete set of diffractive directions (with energy distribution proportional to the far-field scattering intensity of a single dimer unit cell in those directions), while radiation into all other directions is simply compensated by the rest of the elements in the grating. Thus, in order to produce a highly efficient device, suppression of scattering into all undesired diffraction directions, both in transmission and reflection, is required. By slight tuning of the dimer geometry parameters, together with the grating period, it is possible to adjust the bending angle while keeping similar performance.

To experimentally validate the proposed designs, we fabricated the arrays of TiO_2 asymmetric dimers using standard nanofabrication techniques and characterized them using spectrally resolved back focal plane microscopy [135] as described in Methods. This experimental technique provides angular information on the intensity distribution of the light transmitted through the array. Figure 2.14 a shows the measured intensity distribution for the different transmission diffraction orders for the 53° beam bending design. The results are plotted as a function of the incident wavelength and corresponding diffraction angle. The three observed lines correspond to the three diffraction orders supported in transmission, with extracted diffraction efficiencies given in Figure 2.14 b. A scanning electron microscope image of the measured sample is shown in the inset of Figure 2.14 b. As can be seen, almost all the transmitted energy is channelled into the T_{-1} order, reaching a maximum efficiency level above 70%. Experimental spectral minima and maxima are in reasonable agreement with numerical simulations (Figure 2.13 a); exhibiting only a slight spectral

shift and slightly higher transmission for the experimental measurements in the blue spectral region. This can be mainly attributed to slightly different material parameters of TiO_2 in the experiment as compared to those used in simulations. The proposed designs show robustness to small size variations and are suitable for relatively broadband operation. In particular, large efficiencies in the green and blue parts of the visible spectrum are demonstrated with the same nanodimer antenna designs (Figure 2.14 b). This can further be extended for operation in different spectral regions and different bending angles by appropriate design of the nanoantenna geometry and the array period.

Figure 2.14: Diffraction intensity measurements of arrays of asymmetric TiO_2 dimers. (a) Back focal plane intensity measurements and (b) resulting experimental transmission efficiencies for the array of asymmetric dimer structures designed for 53° light bending at 532 nm wavelength. Inset shows SEM images of the fabricated metasurface taken from the top (top image) and at an angle of 30° (bottom image). Majority of the total incident energy is directed into -1 order, whereas the other orders are suppressed, resulting in high diffraction efficiency and transmission levels for a wide range of wavelengths in the green and blue spectral regions. Reproduced with permission from [134].

When trying to apply the same design to increasingly higher angles (close to 90°), several challenges arise. First, the reduction of the unit-cell period necessary to obtain larger angles at a given wavelength leads to an effective reduction of the asymmetry, as the interaction with the closest pillar in the neighboring cell becomes equivalent to the one within the unit cell itself. Second, due to fabrication limitations, the minimum feature size achievable is about 50 nm. This limits the minimum dimer gap achievable, which is the primary parameter defining the scattering directivity/asymmetry, as it controls the strength of the modes/multipoles interaction. A stronger interaction is thus required for a larger tilt of the induced multipole components, which effectively translates into a larger rotation of the directional scattering pattern, bringing the main lobe closer to the array plane ($\sim 90^\circ$ bending).

As a result of these design constraints, the efficiencies achieved using the dimer geometry significantly deteriorate when the diffraction angle increases. To circumvent this issue, we propose a novel geometrical design. The structure is generated by the intersection of a ring and a triangle (reminiscent of a fish shape, as shown schematically in the inset of Figure 2.15 a, for a structure designed for $\sim 82^\circ$ bending angle at 532 nm wavelength). As before, the height of the structures is 305 nm. The outer ring diameter is 290 nm, and its thickness is 70 nm. The triangle base (corresponding to the side parallel to the tangent to the ring) is $H_x = 300$ nm and its altitude is $H_y = 260$ nm. The triangle is positioned in such a way that its closest vertex to the ring is displaced 10 nm inward, with respect to the ring center. The periods are 537 nm (for the diffractive one) and 410 nm (for the non-diffractive one). The ring geometry, as compared to the solid ellipse, suppresses undesired high-order modes excited in the structure and thus simplifies the mode interaction. As discussed above, the distance between the centers of the particles within the asymmetric dimer structure is one of the main parameters defining the directivity and the tilting of the resulting radiation pattern: as a general rule, the smaller the distance, the stronger the interaction

and the larger the tilting angle.

Figure 2.15: (a) FDTD simulations of the transmission distributions into diffractive orders normalized to the incident light energy. The inset is a schematic of the “fish” structure designed for 532 nm wavelength, corresponding to the 82.29° bending angle. (b) Corresponding experimental measurements of the transmission efficiency into different diffractive orders obtained using the free-space optical setup; inset is the SEM images of the fabricated metasurface taken from the top (bottom image) and at an angle of 40° (top image). Reproduced with permission from [134].

In the case of “fish” structure, the effective distance between the centers of the ring and the triangle can be reduced as compared to the case of elliptical dimer, as they can intersect without significantly affecting each other’s modes; that is, they can be considered as two separate but interacting elements. The triangular architecture, however, strengthens the overall asymmetry and pushes up the accessible limits of the efficient high angle beam bending while remaining suitable for the nanofabrication. FDTD simulations predict $\sim 50\%$ energy channelling into the desired diffraction order for the array formed by these structures, as shown in Figure 2.15 a, with less than 10% of the remaining transmitted energy directed into the other suppressed orders. The rest of the

energy is reflected back.

Experimental measurements of this system cannot be carried out using the previous back focal plane microscopy setup, as the metasurface bending angles exceed the collection angle of any available microscope objective (in our case, the largest numerical aperture (NA) available is 0.95, which corresponds to a collection angle of $\sim 72^\circ$). Therefore, experimental measurements were conducted using a free-space optical setup as described in Methods. The SEM images of the fabricated sample are shown in the inset of Figure 2.15 b, and the experimental results of light transmission into the diffracted orders are shown in Figure 2.15 b. One can see a good agreement between the experiment and simulations, with transmission levels into the correct order above 40%. The small wavelength shift, due to fabrication imperfections and slightly different material properties, was observed in the experiment results of optimum efficiency at a slightly reduced operation wavelength of ~ 515 nm leading to a bending angle of 73.5° . This corresponds to the light bending/collection angle of a lens with a numerical aperture $NA = 0.96$.

The fabricated devices based on our concept outperform the conventional structures such as blazed gratings with sawtooth profile or binary blazed gratings. In particular, for medium range bending angles (53°), our design reaches efficiency of $\sim 90\%$, while the sawtooth profile exhibits less than 20% and the binary blazed grating designs are $\sim 80\%$, theoretically [151]. For higher bending angles (82°), the efficiency of the conventional phase mapping method calculated in [141] without considering the discretization issue reaches a theoretical limit $\sim 40\%$ (and is much lower in the case of the sawtooth profile [151]). The design presented here reaches a theoretical efficiency of $\sim 50\%$ with respect to the incident energy with $\sim 80\%$ of the transmitted energy directed into the desired order. In addition to higher bending efficiency, the proposed concept requires shorter structure thickness ($\sim \frac{\lambda}{2}$) and smaller aspect ratios compared to binary blazed gratings [125], [151], which makes it simpler to implement and potentially

more practical.

2.6 Conclusion

The novel concepts presented here, in which nanoantennas with tailored scattering patterns are used to engineer the redistribution of the diffracted energy in periodic arrays, may find important applications in the field of optical gratings, where a precise control of energy distribution is crucial. This method was applied considering the constraints and specifics of the visible regime operation to obtain efficient, high-angle beam bending in transmissive configurations (TiO_2 for green and blue, Si for red parts of the visible spectrum). High efficiency beam bending, exceeding 50% of the incident energy, up to 82° in simulations and 73° in experiments and remarkably broadband operation through the visible spectral range were obtained for TiO_2 structures. For Si device operating at 715 nm wavelength more than 88% of the transmitted energy is concentrated into the correct diffraction order for s-polarized light and more than 82% for p-polarized light. Moreover, the associated flat lens design, with a near-unity numerical aperture, may be useful in a wide range of applications requiring high-resolution focusing, such as photolithography. On the other hand, by reciprocity arguments, the presented lens may find important applications in those situations requiring large angular collection, such as quantum optics experiments involving isotropic emission of a small number of photons. There, this apparently small increase in the NA of the lens translates into a truly significant increase in the collection solid angle, which will boost the efficiency of the mentioned processes. Although for practical applications this would require further optimization of the lens fabrication and design to reach efficiencies comparable to those of commercial microscope objectives, there is no physical limitation to doing so. Overall, we believe that this work represents a clear example of how resonant nanoantennas may help to overcome the limitations of phase mapping approaches to diffractive

optics and how metasurface concepts may lead to truly subwavelength-thick flat optical devices that break the limitations of their traditional counterparts.

2.7 Methods

High angle beam deflection Si metasurface and near-unity NA metalens fabrication

The designed arrays were fabricated using a conventional EBL technique on a quartz substrate (Figure 2.16). Thin films of amorphous silicon (250 nm thickness) were deposited on fused silica substrates via chemical vapor deposition (Oxford PECVD). The samples were then patterned using single-step electron beam lithography (Elionix ELS-7000) using hydrogen silsesquioxane (HSQ, Dow Corning XR-1541-006) resist and a charge-dissipation layer (Espacer 300AX01), and subsequently etched via reactive ion etching in an inductively coupled plasma system (Oxford Plasmalab 100) using chlorine gas.

Figure 2.16: Metasurface fabrication steps.

High angle beam deflection efficiency measurements of Si and TiO₂ metasurfaces

For measuring large-angle bending samples, for which the diffraction angles were larger than the maximum acceptance angle of our collection microscope objectives (NA=0.95), a home-built free-space microscopy setup was used (Figure 2.17, [94]). A supercontinuum source (SuperK Power, NKT Photonics) and band-pass filter (SuperK Varia, NKT Photonics) were used to illuminate the sample. The beam was transmitted through a broadband polarizing beam splitter cube (Thorlabs, PBS252). The linearly polarized light passed then through a quarter wave plate (Thorlabs, WPQ10M-808) to obtain circularly polarized light, which was sent to a rotating linear polarizer (Thorlabs, LPNIRE100-B) to obtain light with the desired linear polarization. Light was then focused onto the sample

using a biconvex lens (Thorlabs) with a 75 mm focusing distance. The diffraction orders were then measured using a pixelated photodetector attached to a digital hand-held laser power/energy meter console (Thorlabs, PM100D) mounted in a rotating arm.

Figure 2.17: Free-space microscopy setup. Reproduced with permission from [94].

Medium and low angle beam deflection efficiency measurements of Si and TiO₂ metasurfaces

Spectrally resolved back-focal plane imaging setup (Figure 2.18, [104]) was used to characterize the diffraction efficiencies of medium and low angle beam deflecting samples. The measurements were performed using an inverted microscopy setup (Nikon Ti-U). In these measurements, white light from a halogen lamp was used to normally illuminate the sample from the substrate side. Polarization was selected using a linear polarizer (Nikon). A spectrometer (Andor SR-303i) with an EMCCD (Andor Newton) detector was placed in the back-focal plane of the collection objective (Nikon 100x, NA = 0.9 for Si and Nikon 100x, NA=0.8 for TiO₂) to measure the transmitted spectrum as well as the k-vector distribution.

Figure 2.18: Back-focal plane microscopy setup. Reproduced with permission from [104].

Fabrication of high angle beam bending TiO_2 metasurfaces

The arrays of TiO_2 asymmetric dimers and “fish” structures were fabricated using standard nanofabrication techniques (EBL). First, a 305 nm thick amorphous TiO_2 film was deposited on top of a glass substrate using ion assisted deposition (Oxford Optofab 3000). Subsequently, a thin chromium (Cr) film (30 nm thick) and a hydrogen silsesquioxane (HSQ) e-beam resist film were deposited on top of the TiO_2 . The Cr film acts as a mask for subsequent etching and prevents charging issues. The nanostructures are then patterned using electron beam lithography (Elionix ELS-7000) and etched using reactive ion etching (ICP-Fluorine, Oxford OIPT Plasmalab System 100). Finally, the sample was cleaned with oxygen plasma and then dipped into Cr etchant to remove the remaining Cr and HSQ mask.

Chapter 3

GaN-based metasurfaces

The material in this chapter was in part presented in [\[152\]](#).

3.1 III-V semiconductors as a platform for metasurfaces

A survey of the dielectric metasurface literature reveals that silicon, more specifically amorphous *Si*, has been extensively used, primarily because of its well- established nanofabrication processes. However, *Si* is not a good material choice at visible wavelengths because of its strong intrinsic absorption. Wide bandgap dielectrics such as *TiO₂* [\[121\]](#), [\[122\]](#), [\[82\]](#) and *Si₃N₄*, [\[153\]](#) which are transparent at visible wavelengths, are currently being investigated as potential low-loss alternatives. These materials, though, are all passive and hence are not suitable for active applications requiring optical gain. Direct bandgap III-V materials are very promising for such active applications because of their strong dipole transition strength and smaller free carrier lifetimes compared to indirect bandgap materials. Typically, the crystal structure of III-V materials does not possess central symmetry, and thus, they exhibit large second order susceptibility (χ^2), which can be exploited to realize optically switchable nonlinear devices. Indeed, recently, *GaAs*-based high aspect ratio nanostructures have been used to

demonstrate optical switching of metasurfaces [91], [154]. Even though *GaAs* is a good material choice for both nonlinear response and emission at near-IR wavelengths, it cannot be applied at visible wavelengths due to its high optical losses. Another potential alternative is *GaP*, which was shown to be an effectively loss-less platform for dielectric metasurfaces above 560 nm [90].

In this chapter and the concomitant paper [152], we experimentally demonstrate that epitaxial *GaN* on sapphire, which is a material of immense technological interest for solid-state lighting technologies, is a viable platform for metasurfaces at visible wavelengths. Thanks to its high transparency through the whole visible spectrum, relatively high refractive index (>2.4 in the visible), and well-developed industrial use as an active material for blue-, cyan-, and green-emitting LEDs and lasers for general lighting, backlighting, and other applications, this platform may pave the way for applications of dielectric metasurfaces to nonlinear and quantum optics. Indeed, III-Nitride materials have already been used to demonstrate electrically driven [155], room-temperature [156] single photon emission. Here, we experimentally show a high-efficiency beam deflecting metasurface and a polarization-splitting metasurface as examples of the viability of *GaN* as a platform for nanophotonics. The metasurfaces were realized on top of epitaxial *GaN* on sapphire wafer by etching the nanostructures directly into the epitaxial *GaN* layer. Very recently, first demonstrations of *GaN* based focusing lenses with a transmissivity of $\sim 86\%$ for blue wavelength operation have been published [157]. In these examples, *GaN* nanostructures were fabricated directly on top of a sapphire substrate. Here we consider *GaN* nanoantennas located on the *GaN* epitaxy with the same refractive index, which opens up a wider range of applications but provides additional design constraints, as explained next.

3.2 GaN-based beam deflection metasurfaces

The primary building block of our metasurface, optimized for operation at a wavelength of 460 nm, a typical emission peak for digital lighting and backlighting [3], is a nanopillar of height 460 nm, as schematically illustrated in Figure 3.1 a. Each pillar can be considered as a waveguide that allows certain modes to propagate with an effective mode index defined by the pillar diameter. The phase shift and transmission through the unit cell, which are dependent on the diameter and the height of the nanopillar, were calculated by numerical modeling using the finite difference time domain (FDTD) technique in commercially available Lumerical FDTD software. The relative phase accumulated along the nanopillar, with the unit cell size fixed at 330 nm in both lateral dimensions and 460 nm in height, can be seen in Figure 3.1 b. The period in which nanopillars are repeated was chosen such that the resulting nanopillar array is sub-diffractive (in air) and small enough to achieve sufficient phase sampling while being also large enough to neglect interactions between nanopillars. Using the nanopillars with diameters tuned from 80 to 210 nm, we are able to achieve a full phase coverage of 2π , enabling complete wavefront control, while simultaneously maintaining high optical transmission ($>70\%$).

Figure 3.1: (a) Schematic illustration of the proposed metasurface capable of deflecting the incident beam from the substrate into the T_{+1} direction. The substrate dimensions, height, and sizes of designed nanopillars are as shown. The diameter D was varied from 80 to 210 nm to realize a linear phase gradient between 0 and 2π . (b) Numerical calculations of the relative phase shift introduced by the nanopillars and transmission for a plane wave of 460 nm wavelength, incident from the substrate side. (c) SEM image of the fabricated *GaN* sample. Reproduced with permission from [152].

To realize a metasurface capable of beam deflection, we introduce a supercell in the x -direction by choosing nanopillars of diameters 124, 143, 167, and 207 nm, which introduce respective phase shifts of approximately $\frac{\pi}{2}$, π , $\frac{3\pi}{2}$ and 2π (marked by green solid circles in Figure 3.1 b). This supercell introduces a linear phase gradient in the x -direction with a periodicity of $1.32 \mu m$. In the y -direction, the metasurface is sub-diffractive with a unit cell period of 330 nm. The designed phase gradient will cause the metasurface to deflect a plane wave incident from

the substrate into the T_{+1} diffractive order. In principle, if the phase sampling is continuous and the transmission is constant, it is possible to achieve 100% deflection efficiency [138], [139], meaning that there is negligible power in T_0 and T_{-1} orders at the operating wavelength.

However, the 4-level discretization, which we chose to use here, limits the theoretical absolute efficiency of beam deflection into the first order to $\sim 81\%$ [158]. We should also note that in our present system, since the metasurface is of the same material as the underlying epitaxial substrate, the resulting diffraction into the substrate cannot be avoided. This can be expected to result in a further reduction in the diffraction efficiency.

The sample, as described above, was fabricated using standard e-beam lithography and inductively coupled reactive ion etching processes as described in Methods. The *GaN* epi-wafers in this work were grown on double-sided polished c-plane sapphire substrates by metal-organic chemical-vapor deposition (MOCVD) system [159].

A representative scanning electron microscope (SEM) image of the metasurface studied in this work is shown in Figure 3.1 c. The sample was characterized by illuminating it using a halogen lamp under normal incidence through the substrate and collecting the back-focal plane image (with an input slit) using a charged coupled device (CCD) camera [135] as described in previous chapter. The images captured on the CCD show spectral and k -dependence of the energy distribution in various diffractive orders [134], [135]. The results for the p-polarization (electric field along the long period of the supercell) are shown in Figure 3.2 a. The white dashed lines represent the expected diffraction orders (in air) for our design. Clearly, most of the incident light is deflected into the T_{+1} order with the deflection angle being dependent on the operating wavelength, as expected from a diffractive design. Figure 3.2 b shows the measured diffraction intensity normalized to the transmitted intensity through the substrate. These curves are obtained by averaging five image pixels on either side of the diffraction

orders depicted as white dashed lines in Figure 3.2 a (the number of pixels is selected to fully integrate the energy going into each individual diffraction order at the image).

Figure 3.2: Measured energy distribution into different diffraction orders as a function of the wavelength for a beam deflecting metasurface illuminated by the p-polarized (a), (b), and (c) and the s-polarized (d), (e), and (f) light through the substrate. The white dashed lines in (a) and (d) represent the diffraction orders into air calculated for the supercell period of $1.32 \mu m$. The color bar in (a) and (d) represents the transmitted intensity normalized to incident light at each wavelength. The experimental data (b) and (e) are obtained by averaging five pixels on either side of the diffracted orders (the white dashed lines) normalized to the substrate transmission (the number of pixels is selected to fully integrate the energy going into each individual diffraction order at the image). The black dashed curves are the simulated results. Relative efficiency (c) and (f), defined as the transmitted intensity into the desired diffraction order normalized to the total transmitted intensity. Reproduced with permission from [152].

The corresponding FDTD simulations are shown as dashed curves. Figure 3.2 c shows the relative efficiency, which is defined as the ratio of intensity in the desired diffraction order to the total transmitted intensity, reaching about 90% at the design wavelength of 460 nm where the deflection angle is 20° . The corresponding measured and simulated data for the s-polarization (electric field perpendicular to the long period of the supercell) are shown in Figure 3.2 panels d-f. The experimental measurements correspond closely to the simulations and show a peak transmission efficiency of $\sim 70\%$ for both the s- and p-polarizations. The transmission into the T_0 and T_{-1} orders is quite small and is limited to about 6% and 1%, respectively. The polarization insensitive behavior of our device is not surprising given the circular cross-section of the nanopillar design. The main features predicted by the numerical simulations are well reproduced in the experiment. Small discrepancies related to the absence in experiment of sharp spectral features predicted by simulations around 440 nm can be attributed to unavoidable nanofabrication imperfections in sidewall profiles and corner rounding, which are different for nanopillars of varying dimensions.

3.3 GaN-based polarization beam splitting metasurfaces

To show the versatility of the proposed *GaN* platform, we now demonstrate a metasurface with the polarization beam splitting functionality. A polarization selective metasurface can be realized by replacing the circular nanopillar by an elliptical nanopillar, wherein the phase velocity of the mode is dependent on the orientation of the input polarization with respect to the major axis of the ellipse. Here, we design and experimentally show a polarization beam splitting metasurface that deflects the p-polarized incoming light into the T_{+1} diffractive order and the s-polarization into the T_{-1} diffractive order. The design principles are similar to the phase gradient concepts discussed previously with

one major difference — the ellipses in the supercell are arranged such that the phase gradients point in opposite directions for the p- and s-polarizations, as schematically shown in Figure 3.3 a.

Figure 3.3: (a) Schematic illustration of the phase gradients employed to demonstrate polarization beam splitting metasurface. The phase introduced by each nanopillar is dependent on the radii and the orientation relative to the polarization direction. (b) A representative SEM image of the fabricated GaN device. (c) and (f) Spectrally resolved back focal plane images showing the intensity of light transmitted in various diffraction orders for the p- and s-polarizations, respectively. For the p-polarized illumination, the transmitted light deflects predominantly into the T_{-1} diffraction order, while for the s-polarized illumination, the light is directed into the T_{+1} order. Continued on the next page.

Figure 3.3: (d) and (g) Spectral dependence of intensity in the T_{-1} , T_0 , and T_{+1} diffraction orders for the p- and s-polarizations, respectively. (e) and (h) Relative efficiencies of light channeling into the T_{+1} and T_{-1} orders, for the p- and s-polarizations, respectively. The measured peak relative efficiencies of beam deflection are 73% for the p-polarization and 67% for the s-polarization at 430 nm illumination. The solid colored curves represent the measured values, while the black dashed ones correspond to the numerical simulations. Reproduced with permission from [152].

The amplitude transmission coefficient and phase maps obtained by varying the radii of elliptical nanopillars and the specific design parameters used are given in Figure 3.4 b and c, correspondingly. The design height was kept fixed at 460 nm similar to the beam deflecting metasurface above.

Figure 3.4: Schematic illustration of the FDTD simulation parameters (a) used to calculate the transmission coefficient (b) and the corresponding accumulated phase (c) as a function of semi-axis of *GaN* elliptical nanopillars on *GaN* substrate. Pillar height was fixed at 460 nm. Radii span from 25 to 140 nm provides sufficient 2π phase coverage, while maintaining a sufficiently high transmission level. Reproduced with permission from [152].

A representative SEM image of the fabricated *GaN* metasurface sample is shown in Figure 3.3 b. The back focal plane measurements shown in Figure

3.3 panels c and f demonstrate the input light deflecting into the T_{-1} and T_{+1} orders for the p- and s-polarizations, respectively. The spectral dependence of the measured diffraction orders, along with the corresponding numerical simulations, is shown in Figure 3.3 panels d and g. Experimentally, we measure $\sim 50\%$ of the transmitted light channeled into the T_{-1} order for the p-polarized light and $\sim 40\%$ into the T_{+1} order for the s-polarized light. The relative diffraction efficiencies achieved in our experiments are $\sim 74\%$ for the p-polarized light and $\sim 66\%$ for the s-polarized light (Figure 3.3 panels e and h). The experimentally realized extinction ratios are 8.6/1 and 6.2/1 for the p- and s-polarizations, respectively.

3.4 Conclusion

In conclusion, we experimentally demonstrated *GaN* as a suitable material platform for realizing a wide range of high-efficiency metasurface-based devices with enhanced functionalities operating across the whole visible spectrum including the blue spectral region around 450 nm. As a proof-of-concept demonstration, we have experimentally showed an epitaxially grown *GaN* based polarization insensitive metasurface that diffracts incoming light at 460 nm wavelength to an angle of 20° with $\sim 70\%$ absolute transmission efficiency and $\sim 90\%$ relative transmission efficiency. These reasonably high efficiencies are achieved despite the fact that the refractive index of the metasurface is the same as the underlying substrate, which is widely believed to lower the efficiency. Additionally, we have also demonstrated a polarization beam splitter working at 430 nm wavelength and capable of separating the p- and s-polarizations with the relative efficiencies of 73% and 67%, respectively. We expect that further development of metasurfaces based on *GaN* and its alloys with *InN* and *AlN* will pave the way for active, nonlinear, and quantum nanophotonics.

3.5 Methods

GaN metasurfaces fabrication

The *GaN* metasurfaces, as described above, were fabricated using standard EBL. To pattern metasurfaces on the *GaN* epitaxy, a HSQ resist layer (XR1541-06 Dow Chemicals) was spin-coated on the substrate, followed by electron beam lithography (EBL) (Elionix ELS 7000 100kV). The patterns were developed using a high contrast salty developer (1 wt% NaOH + 4 wt%NaCl in water) [160]. The HSQ pattern was transferred into the GaN layers by inductively coupled plasma reactive ion etching (ICP-RIE) using Cl_2 chemistry (Oxford Instruments, Plasmalab 100).

Chapter 4

LED and metasurface integration

The material in this chapter was in part presented in [161].

4.1 Direct approach: a metasurface patterned on top of an LED

After designing a highly efficient metasurface on *GaN* platform described in detail in the previous chapter, the next logical step would be to pursue direct integration on top of the *GaN* - based LED. The phase mapping approach used to design the metasurface assumed a plane wave excitation as the light source. However, in the case of the LED, the excitation profile becomes a broad Lambertian, instead of planar, as shown in Figure 4.1 a. This and the other radiation pattern results shown in the chapter were obtained numerically based on the reciprocity principle [162] as described in Methods. The polar plot shown in Figure 4.1 a corresponds to a cut of the full 3D pattern (identical for any azimuthal angle due to rotational symmetry). In numerical simulations the thickness of the MQW region was 100 nm, the depth under the LED surface (or p-doped layer thickness) was 2 μm . Here, the p-doped layer thickness does not influence the shape of the radiation diagram.

Figure 4.1: Emission diagrams of LED devices obtained from the reciprocity principle calculations: (a) the standard flat semi-infinite *GaN* LED (b) the gradient metasurface consisting of four elements integrated directly into the p-doped layer of the semi-infinite LED structure. The polar plot shown corresponds to a cut of the full 3D pattern along the long axis of the metasurface supercell. The simulation wavelength is 460 nm. The blue, solid line represents the unpolarized emission while the red and green dashed lines denote the p-polarized and s-polarized light components, respectively. Insets illustrate the schematics of the structures.

As mentioned before, the excitation profile for the proposed direct *GaN* metasurface integration is the broad Lambertian. Figure 4.1 b shows the numerical simulations of the radiation pattern of the metasurface device directly placed onto the LED slab (*GaN* metasurface on *GaN* substrate). The metasurface consists of four cylinders with circular cross-section periodically repeated along the surface, which provides a polarization insensitive output. The cylinder diameters are 124, 143, 167, and 207 nm; the height is 460 nm (same as in the beam deflecting metasurface demonstrated in the previous chapter). The simulation wavelength corresponds to the typical emission of the blue *GaN* LED - 460 nm [122].

The polar plot shown corresponds to a cut of the full 3D pattern along the long axis of the metasurface supercell. The resulting emission diagram with the

metasurface has a negligible difference with respect to the original Lambertian without the metasurface, hence the beam deflecting metasurface completely loses its functionality. The thickness variation of the p-doped layer (1 nm - 2 μm) and MQW thickness (1 nm - 100 nm) slightly reshapes the emission pattern. However, the differences are almost imperceptible and do not qualitatively influence the result. As a conclusion, the metasurface designed with a plane wave excitation generally loses its functionality due to the Lambertian profile of excitation, hence, the direct patterning of the metasurface onto the LED slab does not seem a viable approach.

4.2 Collimating cavity principle

In this section a new method for direct integration of metasurface devices on top of an LED is proposed. The method relies on a collimating element, in particular, a resonant cavity. Both the metasurface and the cavity LED can be designed separately and, thus, the method allows integration of any functional metasurface designed to work under normally incident plane wave illumination, provided that it works in the transmission regime. Moreover, due to its resonant character, the presence of the cavity enhances the emission of the device.

The proposed method is schematically depicted in Figure 4.2. First, spatially incoherent emission of the LED is modified by restricting the initial broad Lambertian angular distribution to a narrow range of angles. This is achieved by introducing a resonant Fabry-Pérot (FP) cavity into the LED architecture. Note that the spatial coherence increased by resonant cavities [17] (which have been used in the LED community for more than two decades to enhance LED efficiencies [12], [14], [15]) could also be realized using other approaches, such as zero index materials [110], plasmonic [163] and photonic crystals [19]. In this work, we use the resonant FP cavity as it provides sufficient collimation, emission enhancement and does not require complex fabrication steps. Finally,

the metasurface performing the desired functionality is integrated at the output plane of the cavity to realize the compact device.

Figure 4.2: Working principle of the LED and metasurface integration. The wide angle Lambertian distribution of the plain LED slab is transformed into highly directional emission using the resonant cavity. Next, metasurface is fabricated on top providing the desired functionality (beam deflection as an example). Reproduced with permission from [161].

The initial choices for practical implementation of the collimating cavity concept were *GaN*-based LED source and *TiO₂* as the material for metasurface.

Using the MOCVD [159], the typical growth process for the *GaN*-based LED is performed on a sapphire substrate (which can be pre-patterned). Initial layer of undoped *GaN* grown on sapphire will be of low quality due to the lattice mismatch. Hence, in order to fabricate a high-quality LED this layer has to be several microns thick, followed by several microns of the n-doped *GaN*, quantum wells and p-doped *GaN* layer.

Hence, ready for our fabrication process, the resulting *GaN* stack is 7 μm thick. The FP cavity has to operate on the lower order resonances, otherwise radiation pattern exhibits many side lobes and the whole collimation process is inefficient. As the operation wavelength for our *GaN*-based LED is 460 nm, the first order FP resonances correspond to a slab thickness of several hundred nanometers. Thus, the initial thick stack grown on sapphire had to be thinned down. The typical process for the flip-chip LED configuration was used. The initial LED stack is attached to a metallic mirror (which acts as a part of the FP cavity) on the *GaN* side. Then the sapphire substrate is detached via laser lift-off (LLO) process. However, when the remaining stack is being dry-etched in the ICP-RIE setup (thinned down to FP resonance thickness) it exhibits catastrophic micromasking and non-uniform etch. Micromasking is a result of the non-uniform etch due to defects or contaminants, where the area surrounding the defect is etched at one rate and the area under the defect is “masked” and does not etch.

Micromasking after ICP-RIE etch process may have originated from several sources: carrier wafer etching and re-deposition of the particles on the sample, contamination of the gas chamber itself, defects and nucleation of the initial layers of the *GaN* grown on sapphire due to the lattice mismatch. We were able to exclude some of the problematic sources: the carrier problem was solved with switching to carbon carrier wafer (from *Si* and *SiO₂*) and contamination of the sample from chamber was decreased using different gas chemistry (*BCl* and *Cl*) with intermittent ultrasound cleaning. However, nucleation is a part of the *GaN*

standard growth procedure and it can not be solved with available methods and resources.

The quality and flatness of the resulting *GaN* slab without solving the nucleation problem does not meet the requirements set for the metasurface fabrication. Therefore, to circumvent the lattice mismatch issue, the *GaP* (gallium phosphide) LED platform has been chosen as an alternative. *GaP* is another industrially well-developed platform for LEDs, mainly focusing on the red region of the visible spectra. Compared to *GaN*, *GaP* can be grown on *GaAs* substrate where the lattice mismatch is minimal and quality of the material is high. Thus, thinner LED stacks can be fabricated, then detached (by means of sacrificial layer) and transferred to any desired substrate. *Si* can be used as a metasurface material due to its low losses in the red part of the visible spectrum, where *GaP* LEDs typically emit. The refractive index of *Si* is higher than that of *TiO₂*, translating into smaller and lower aspect ratio particles composing the metasurface.

4.3 Numerical simulations of the collimating cavity and metasurface device

For the theoretical analysis of the integrated device, we performed full numerical simulations based on the reciprocity principle [162], as described in the previous section. This approach allows precise computation of the radiation pattern, the relative contribution of each polarization and the enhancement/suppression of the total emission relative to a plain LED. As mentioned above, we choose to work with a *GaP* LED structure with an active layer consisting of MQW emitting at around 620 nm wavelength. Following the characteristics of the samples used in the subsequent experiment, we assume that the active layer thickness is 100 nm with its first interface located 200 nm below the top *GaP* surface.

The radiation patterns for various LED configurations are given in Figures

4.3 a-c. The respective insets show the schematic side views of the considered structures. First, the Lambertian emission from a semi-infinite LED stack is calculated and plotted in Figure 4.3 a. The red and green lines represent, respectively, the emission patterns of the p-polarized and s-polarized light.

Figure 4.3: Numerical simulations of emission diagrams of the LEDs in various configurations. (a) Emission pattern of a semi-infinite GaP LED stack. Results show the characteristic Lambertian shape. (b) Emission pattern of a hybrid Bragg-gold RCLED. Results show directional emission towards the surface normal. (c) Emission pattern of the hybrid RCLED with the integrated, beam deflecting metasurface, as described in the text (the polar plot shown corresponds to a cut of the full 3D pattern along the long axis of the metasurface supercell). Results show an emission lobe at 30° with respect to the normal, corresponding to the deflection angle of the metasurface when operating under plane wave excitation. Continued on the next page.

Figure 4.3: All diagrams are normalized to the maximum directional emission of the semi-infinite LED. The blue, solid line represents the unpolarized emission while the red and green dashed lines denote the p-polarized and s-polarized light components, respectively. The insets show the side view schematics of the different systems under consideration. Reproduced with permission from [161].

In order to transform the Lambertian LED emission into the one having a narrower angular distribution, which can be manipulated by the metasurface, we use a hybrid Bragg-gold resonant FP cavity as a collimating element (see inset in Figure 4.3 b). Such an asymmetric choice of the FP cavity mirror is dictated, on one hand, by the developed GaP wafer transfer to gold (which acts as the bottom mirror) and, on the other one, by the low losses of the Bragg mirror (acting as the top reflector). The gold layer is chosen to be several hundred nanometers thick to ensure high reflection and the Bragg reflector consists of alternating TiO_2 and SiO_2 layers, chosen due to their high refractive index contrast and negligible losses in the visible spectral range. The thicknesses of the layers satisfy the quarter wavelength condition - 63 nm for TiO_2 and 107 nm for SiO_2 . An analysis of the FP cavity performance as a function of the number of layers in the Bragg stack reveals that 2 pairs of layers are enough to ensure sufficient emission collimation. Numerical simulations of the emission pattern and cavity bandwidth analysis depending on the number of Bragg stack pairs are shown in Figure 4.4 a,b. Higher number of Bragg stack pairs results in a better-quality cavity and, therefore, collimation. However, as a tradeoff, the operation bandwidth is narrowed as well and the total thickness is increased. The thickness of the GaP is then optimized to be 415 nm to support a Fabry-Perot resonance at the central wavelength of the LED emission (around 620 nm). The MQW layer thickness and location remain the same as before. The resulting emission diagram of this system, generically known as the resonant cavity light-emitting diode (RCLED) [17], [12], [14], [15] is shown in Figure 4.3 b. As expected, in this situation the light is concentrated in a narrow angular range close to the surface normal,

corresponding to those angles for which the cavity resonance condition is met.

Figure 4.4: Numerical simulations of the hybrid Bragg gold RCLED cavity bandwidth and emission diagram as a function of the Bragg stack pairs number. a) RCLED cavity reflection. Red, blue and green curves denote 1, 2 and 3 Bragg pairs correspondingly. PL of the LED (brown curve) is taken as a Lorentzian with a FWHM value corresponding to the experimentally measured GaP LED. b) Emission diagrams of RCLEDs with 1, 2 and 3 Bragg pairs (red, blue and green curves) in the top reflector at 620 nm wavelength, corresponding to cavity reflection minimum. Uncovered LED emission (brown curve) is given for reference. Each of the emission diagrams is normalized to its own maximum. Reproduced with permission from [161].

As a last step, the beam deflecting silicon metasurface described above is now placed on top of the RCLED (inset of Figure 4.3 c). A thin SiO_2 spacer (210 nm) is added between them to decouple the metasurface from the near-fields of the cavity and to serve as a low index substrate for the metasurface. The metasurface consists of a rectangular lattice of periodically repeated supercells with 300 nm (sub-diffractive) and 1200 nm (diffractive) periods. The elements constituting the supercell are amorphous silicon cylinders with a height of 350 nm and a circular cross-section to guarantee polarization-independent operation. The diameter of the cylinders increases gradually within the supercell (114 nm, 124 nm, 133 nm and 170 nm), leading to an increasing phase accumulation under coherent plane

wave illumination and mapping a tilted wavefront of a deflected beam. When illuminated by a normally incident plane wave at a wavelength of 620 nm, this metasurface is able to deflect it at an angle of 30° with a 94% relative efficiency and around 50% overall transmission for two orthogonal polarizations (see Figure 4.5).

Figure 4.5: Numerical simulations of the metasurface performance: transmission, reflection and absorption (TRA) spectra of the Si metasurface excited with a normally incident plane wave; the metasurface is designed to deflect the incoming light at around 30° angle (into the -1 diffraction order) at 620 nm incident wavelength. Reproduced with permission from [161].

The emission pattern from the metasurface combined with RCLED device, depicted in Figure 4.3 c, shows a single, angularly narrow emission lobe with a clear maximum at around 30° with respect to the normal, thus confirming the validity of the proposed concept. Since we used cylinders with a circular cross section as building blocks of the metasurface, the beam deflection effect is almost polarization insensitive, as seen by comparing the red and green curves representing, respectively, the emission patterns of p-polarized and s-polarized light. An important aspect to note is that independent design of the cavity and the metasurface can only be applied to highly transmissive metasurfaces (or at

least to those with low reflection). If that is not the case, the reflected light from the metasurface can affect the cavity performance. In that situation, a more precise engineering considering both the metasurface and the cavity as a whole is required.

For the sake of completeness, we show in Figures 4.6 a,b the performance of a device with a simpler cavity, consisting of two gold mirrors, which could be advantageous for electrically-driven devices. As can be seen, the performance of this device is very similar to the hybrid cavity case, but with a lower level of emission power. We finalize the theoretical characterization of the presented devices by calculating the total emitted power relative to the bare LED stack. This is done by integrating the directional emission over the top hemisphere. These results are presented in Figure 4.6 c.

Figure 4.6: Numerical simulations of the emission diagrams of the RCLED combined with the metasurface and the total emission efficiency comparison. (a) The emission diagram of the pure gold RCLED; the directionality is well pronounced. (b) Directional emission of the gold cavity RCLED integrated with the Si metasurface, the deflection angle of 30° corresponds to the theoretically designed angle for the plane wave excitation. The insets demonstrate the side view schematics of the structures, red and green lines in the emission patterns denote the p-polarized and s-polarized light components, respectively, while the blue line denotes their sum. (c) Total emission efficiency comparison for all proposed LED designs, calculated as an integral emitted power over the top hemisphere, normalized to that in the case of the plain semi-infinite LED stack. Reproduced with permission from [161].

The RCLED including the Bragg reflector leads to an enhancement of the

total emission of almost three times compared to the bare LED, which is about one order of magnitude higher compared to the RCLED based on gold mirrors. Including the metasurface on top of the RCLED slightly reduces, in both cases, the total emitted power. We attribute this observation to the non-negligible absorption of amorphous *Si* at the operating wavelength as well as to the small reflection from the metasurface (which was designed independently of the cavity). Nevertheless, the results still show that the total emitted power of the hybrid RCLED with the metasurface is almost twice higher than that of the bare LED and one order of magnitude higher than that of the configuration based on the gold FP cavity. Overall, both configurations - with gold or Bragg top reflector in the FP cavity - have their own advantages in terms of efficiency, design, fabrication simplicity and suitability for electrically-driven devices and, therefore, we believe that both architectures can find applications depending on the device requirements.

4.4 Experimental realization of the collimating cavity and metasurface device

For the experimental demonstration of the proposed concept, we focus on the hybrid Bragg-gold cavity configuration. To realize the device, we start with a commercially available *GaP* p-i-n junction with multiple quantum wells on a gold-coated silicon substrate. *GaP* LEDs on gold substrates were purchased from Yangzhou Changelight Co. The stack of multiple quantum wells of around 100 nm in thickness is sandwiched in the *GaP* p-n junction. The whole initial LED stack thickness is around 430 nm. The thickness of the gold substrate is around 1 μm . The quantum wells emission is centered at 628 nm with a FWHM of around 20 nm.

Although the thickness of the *GaP* layers can be precisely controlled during the growth, in the case of a very thin LED layer the lattice mismatch with the

substrate material deteriorates the quality of the film and affects the efficiency. This issue was solved by growing thicker structures and subsequently thinning them down by inductively coupled plasma-reactive ion etching (ICP-RIE, Oxford Plasmalab 100). We thus obtained, with a high precision, the desired slab thickness corresponding to the low order Fabry-Perot resonance at the emission wavelength. We combined the dry etching based on *Ar* and *Cl* chemistry with reflection spectrum measurements to adjust the thickness to approximately 416 nm. In those cases, in which the *GaP* layer was slightly over-etched, a thin layer of amorphous *Si* was deposited to precisely meet the Fabry-Perot condition. For that we used plasma-enhanced chemical vapour deposition (PECVD) (Oxford Plasmalab 100). Next, two pairs of *TiO₂/SiO₂* layers (63 nm /107 nm) were deposited on top using ion-assisted deposition (IAD Oxford Optofab 3000), followed by a *SiO₂* spacer layer (210 nm).

The optical characterization of the fabricated structures was performed using a spectrally-resolved back focal plane imaging setup [152], [134], as described in the previous chapters, incorporating a Nikon 100x microscope objective with a numerical aperture $NA = 0.95$. Figure 4.7 presents a comparison of the angularly-resolved reflection and photoluminescence spectra for the initial, uncovered LED slab on gold and the cavity LED incorporating the top Bragg mirror. The angularly-resolved reflection spectra were obtained using the white light excitation. For the uncovered sample (Figure 4.7 a) the measurements reveal two bands of slightly reduced reflectivity, corresponding to weak Fabry-Perot resonances that arise from the reflections between the gold substrate and the *GaP*-air interface. With the Bragg mirror on top, the resonant cavity LED exhibits much more pronounced Fabry-Perot resonances with almost vanishing reflectivity, one being centered at the emission wavelength of the *GaP* quantum wells of 628 nm (Figure 4.7 b).

Figure 4.7: Experimental optical characterization of the uncovered LED and hybrid Bragg-gold RCLED. (a)-(b) Angularly-resolved reflection spectra of the (a) uncovered and (b) resonant cavity LED (RCLED). (c)-(d) Angularly-resolved photoluminescence (PL) spectra of the uncovered LED (c) and the hybrid Bragg-gold RCLED. Continued on the next page.

Figure 4.7: (e)-(f) Back focal plane images of PL emission (including all wavelengths and polarizations) of the uncovered LED (e) and the hybrid Bragg-gold RCLED (f). PL was excited by a laser at the wavelength of 488 nm and collected using a microscope objective with NA=0.95. Reproduced with permission from [161].

After characterizing the reflectivity of the device to corroborate the presence of the Fabry-Perot resonance at the desired working wavelength, we measured the photoluminescence (PL) signal by optically pumping the system with a continuous wave (CW) laser at 488 nm wavelength. We pumped from the *GaP* side for the uncovered sample and from the Bragg reflector side for the RCLED. Figures 4.7 c-d show the measured angularly-resolved PL spectra for the uncovered LED and the hybrid Bragg-gold RCLED. The angle represents the polar angle measured with respect to the normal direction to the sample surface. For the uncovered LED case, the PL emission is spread into all directions, including the large angles (Figure 4.7 c), resembling the Lambertian pattern. A slight deviation is observed, however, as it is affected by the weak Fabry-Perot resonance observed in the reflection spectra, producing a slight narrowing of the angular emission distribution. On the other hand, the RCLED demonstrates clearly enhanced directivity and angular narrowing (Figure 4.7 d) as a result of the precise overlap of the PL emission maximum with the strong Fabry-Perot resonance. Figures 4.7 e-f show the full angular distribution of unpolarized PL including all emission wavelengths. Note that panels 4.7 c-d correspond to spectrally-resolved cuts along the y-axis in panels 4.7 e-f. Figure 4.7 e indeed corroborates the quasi-Lambertian emission of the uncovered sample, while the hybrid Bragg-gold RCLED (Figure 4.7 f) presents clear directionality towards the normal to the sample surface.

Subsequently, the *Si* gradient metasurface is fabricated on top of the device as described in Methods. The metasurface supercell consists of four cylindrical elements and was designed to deflect the incoming wavefront at an angle of 30°

with respect to the normal. The SEM image of the metasurface fabricated on top of the RCLED is given in Figure 4.8 a. Back focal plane measurements of the hybrid Bragg-gold RCLED with a metasurface on top are presented in Figure 4.8 b-c. As designed, metasurface shifts the PL emission maximum towards the designed 30° angle when compared to the RCLED case without the metasurface (Figure 4.7 d-f). Note that, while in the first two cases (uncovered and RCLED without metasurface) the emission is azimuthally symmetric, this is not the case when the metasurface is incorporated on top. The full PL shown in Figure 4.8 c. Figure 4.8 b is a wavelength resolved cut along the y-axis of Figure 4.8 c, corresponding to the long axis of the metasurface supercell. It should be noted that, while numerical simulations of directivity were performed for a single wavelength, the experimental emission pattern contains a certain bandwidth. However, as it is a relatively narrow band (~ 20 nm), the device preserves the collimation and beam deflection functionality for the whole PL signal, including all wavelengths in the LED emission spectrum (Figure 4.8 c).

Figure 4.8: (a) SEM image (taken at 30°) of the amorphous silicon-based, beam deflecting metasurface, fabricated on top of hybrid Bragg-gold RCLED. A supercell of the metasurface is highlighted in false red color. (b) Angularly-resolved photoluminescence (PL) spectra of the hybrid Bragg-gold RCLED with metasurface. Continued on the next page

Figure 4.8: (c) Back focal plane images of PL emission (including all wavelengths and polarizations) of the hybrid Bragg-gold RCLED with metasurface. PL was excited by a laser at the wavelength of 488 nm and collected using a microscope objective with NA=0.95. Reproduced with permission from [161].

Nevertheless, a fairer comparison between the theory and experiment can be obtained by comparing the directionality cross sections at a specific wavelength (for which we chose, in particular, 634 nm wavelength providing the highest collimation). This comparison is shown in Figure 4.9. As can be seen from the figure, the experimental results have a very good correspondence with the theory, the main deviation being between the uncovered case and the pure Lambertian emission from a semi-infinite LED stemming from the weak Fabry-Perot resonance observed in this system, as discussed above.

Figure 4.9: Comparison between the numerical simulations and experimentally measured emission directivity at a single wavelength. Simulated (a) and measured (b) emission patterns corresponding to the uncovered LED (red curve; simulated results correspond to the semi-infinite LED) and the RCLED without (green curve) and with (blue curve) the beam deflecting metasurface on the top. The experimental results correspond to the cross section of the angularly-resolved PL spectrum at 634 nm wavelength. Reproduced with permission from [161].

We finalize the characterization of the fabricated devices by estimating their

relative emission efficiencies. This can be done by integrating the emission power in Figures 4.7 e-f and Figure 4.8 over the full measured solid angle. The directly obtained efficiencies are 39% for the RCLED alone and 3% for the RCLED with the integrated metasurface, both relative to the plain *GaP* LED on gold. One should note that the emitted power is affected by different pumping conditions for each device. While all of them are pumped from the same side, the pump beam experiences different reflections depending on whether it is the uncovered case, the case containing the Bragg stack, or that having both the stack and the metasurface. Thus, these directly obtained efficiencies should be normalized to the actual pumping power reaching the quantum wells in each case. Numerical simulations estimate that the real excitation power, relative to the uncovered case, is around 58% for the RCLED and 38% for RCLED with the integrated metasurface.

In addition, simulations estimate an excitation enhancement of around 2.5 times for the uncovered case of the LED layer on gold compared to the bare semi-infinite LED. Taking into account these numbers we arrive at an estimated total emission efficiency of around 168% for the hybrid RCLED relative to the bare LED (without the gold substrate), and of around 20% when the metasurface is integrated on top. These values are still lower than those predicted by the simulations (shown in Figure 4.6 c). This can be attributed to the wavelength dependence of the efficiency within the bandwidth of the Fabry-Perot resonance (~ 12 nm) which, moreover, is also narrower than the PL bandwidth (~ 20 nm), an effect not taken into account in the simulations. Underestimation of material losses in *Si*, *Au*, and the remaining EBL resist on top of the structures, as well as fabrication imperfections, might also affect the efficiency of the fabricated devices.

As a last step, we illustrate how the proposed concept allows to realize full wavefront control, and thus possible functionalities that are not limited to the beam deflection. To do so, we fabricated and integrated on top of the same hybrid

gold-Bragg RCLED another metasurface designed to generate a beam with an orbital angular momentum (OAM) and a topological charge of +1. Note that this functionality cannot be achieved unless full control of the wavefront is realized. The design of this metasurface follows the same approach of phase mapping, gradually varying the radius of the cylinders to achieve $0-2\pi$ phase variation along a full round of the azimuthal angle, thus imprinting the desired topological charge. The SEM image of this vortex beam generating metasurface fabricated on top of the hybrid Bragg-gold RCLED is shown in Figure 4.10 a. After pumping the device, we measured the intensity profile of the emitted light. The result is plotted in Figure 4.10 b. As can be seen, the measured photoluminescence exhibits the characteristic “doughnut-shape” profile corresponding to a vortex beam with an orbital angular momentum and the expected topological charge, thus confirming the predicted functionality of the device.

Figure 4.10: Experimental characterization of the GaP cavity LED with the vortex metasurface on top. (a) SEM image of the central area of the vortex metasurface, taken at 30° tilt. (b) Intensity profile of the PL from the vortex beam metasurface LED, excited at 590 nm wavelength. Reproduced with permission from [161].

4.5 Conclusion

In conclusion, we proposed a solution for integration of metasurfaces on top of LEDs to endow these devices with on-demand functionalities. The proposed solution is based on a collimating element, which can be realized, e.g., using a resonant cavity. This resonant cavity transforms the Lambertian-like emission from the bare LED into a directional one, thus having a large spatial coherence that can then be manipulated using any transmissive metasurface designed for plane wave excitation. Both the cavity and the metasurface can, thus, be designed separately, as long as the reflection of the metasurface is low. Otherwise, the concept can still be used, provided that the system is optimized as a whole. Due to the resonant character of the cavity, the angular narrowing of the emission pattern does not necessarily come at the expense of a weaker emission, which can even be enhanced. We experimentally demonstrated the proposed concept using a *GaP*-based LED combined with a hybrid gold-Bragg cavity and a silicon metasurface. We show two different functionalities, namely, beam deflection and vortex beam generation.

The suggested method is general and can be applied to realize other, more complex functionalities such as LED beam focusing or hologram generation. Although demonstrated here using optical pumping, the concept can be readily adapted to obtain electrically-driven devices, either employing a fully metallic RCLED architecture, such as the one proposed based on gold layers, which corresponds to the flip-chip LED configuration, or substituting the bottom dielectric layer of the Bragg stack by some transparent conducting layer (e.g. *ITO*). Finally, we note that this concept can be applied to other random emitting media such as fluorescent molecules and quantum dots. The proposed solution represents a step forward towards multipurpose, efficient, integrated optical devices, opening venues for compact integration of metasurfaces, which enable unprecedented control of light, with random sources. This can enable integration of LED sources instead of lasers into optoelectronic devices finding broad

applications in optical communications, Li-Fi, displays, solid-state lighting, sensing and many more.

4.6 Methods

Numerical simulations of the emission patterns

LED radiation pattern results were obtained numerically based on the reciprocity principle [162] using the commercially available Lumerical FDTD and COMSOL Multiphysics software. To obtain them the proposed LED designs were inversely excited with plane waves at different angles covering the full, top hemisphere. For each angle these plane waves form a spatial power density distribution within the LED. The integral of this power density over the active region of the LED (multiple quantum wells, MQW) is proportional to the direct power emission of the LED quantum wells at that specific angle. This simulation, thus, represents the time reversed process of light emission.

Fabrication of the beam deflecting Si metasurface

For the metasurface fabrication we started with a deposition of an amorphous silicon film on silica substrate using the PECVD (Oxford Plasmalab 100) process. We subsequently spin-coated hydrogen silsesquioxane (HSQ, Dow Corning XR-1541) as a resist for the following electron beam lithography (EBL) process as well as a layer of Espacer 300AX01 (Showa Denko) as a charge dissipation layer. We then proceeded to expose the sample using an EBL (Elionix ELS-7000, 100kV) to define the metasurface structure. Next, the sample was developed using a 25% solution of tetramethylammonium hydroxide (TMAH). Finally, the sample was etched using the same ICP-RIE tool with Cl_2 gas. The remaining HSQ resist on top of the etched structures was not removed, as the hydrofluoric acid (HF) solution commonly used for its removal might damage the SiO_2 substrate and the FP cavity stack.

Chapter 5

External collimating cavity

5.1 Principle and numerical simulations

The approach for LED and metasurface integration exploiting the resonant cavity LED, which was introduced in the previous chapter, demonstrated the required full control of the wavefront and the potential efficiency enhancement of the LED platform. That approach, however, requires the intermediate step of enclosing the emission layer in a resonant cavity. This procedure imposes a number of additional fabrication processes that might be unnecessarily costly, time-consuming or simply incompatible with certain specific types of random emitters used as a light source, as well as with their standard production methods. OLEDs, quantum dots, perovskites, 2D active materials and other types of random emitters may exhibit strong chemical sensitivity to the environment as well as to their temperature regime. Several fabrication processes (dry and wet etch, lift-off techniques, thermal evaporation, high temperature PECVD and others) intended for the transfer, deposition and tuning of the resonant cavity thickness might damage the emitters and, therefore, drastically reduce their light emission efficiency (IQE). Finally, the surface roughness and defect quality of the fabricated device have to satisfy the necessary requirements for metasurface fabrication. In particular, for a typical lateral GaN LED grown on sapphire,

transfer on the reflector material is required, with subsequent thinning down from several microns to several hundreds of nanometers of thickness. Problems of micromasking and nucleation that occur in those processes, discussed in the previous chapter, resulted in low quality, rough surface with multiple defects - thus being completely unsuitable for further metasurface patterning.

In this chapter, we introduce another novel approach for LED and metasurface integration based on an external resonant cavity. This approach is similar to the resonant cavity approach discussed in the previous chapter, but mitigates all the important concerns and fabrication constraints that we described above. The external cavity approach can be applied to a wider range of light emitters and minimizes the processing of ready-made light source platforms.

Figure 5.1 illustrates the key concept of the external collimating cavity approach. The external cavity transforms the broad Lambertian profile into a collimated, narrow angle spread emission i.e. enhances the spatial coherence of the emission. “External” in this case is related to its position with regards to the active layer of emitters: the cavity does not enclose the emitters and is located outside, as compared to the previously introduced “internal” cavity case where the emission layer is positioned inside the resonant cavity. Next, the desired functionality is obtained by placing or patterning a metasurface on top of the external resonant cavity device.

The important advantages of the external cavity approach are the flexibility and complete separation of the light source from the cavity and metasurface, while the compact size of the device is conserved. With this approach, a standard industrially-fabricated light source with all the electrical contacts and mandatory protective encapsulation is suitable for further metasurface integration as long as surface quality required for deposition and patterning are satisfactory. Moreover, the bonding of the cavity to the light source device can be performed after the fabrication of the metasurface on top of the cavity, making this approach simple to implement and, therefore, attractive for a broad range of applications.

Figure 5.1: Working principle of the LED and metasurface integration using the external collimating cavity. The wide angle Lambertian distribution of the plain LED slab is transformed (filtered) into highly directional emission by the resonant cavity deposited on top of the device. Next, a metasurface pattern with a desired functionality is added (beam deflection as an example).

The distance between the metasurface and the light source will define the photon flux incident on the metasurface, and hence, implicitly impact the light extraction efficiency. Nevertheless, the concept will hold for both long and short distances (relative to the lateral size of the device).

In this chapter we will demonstrate the the full wavefront control of the

GaN-based LED device by means of external Bragg cavity and the dielectric (*TiO₂*) metasurfaces. The numerically simulated system is shown in Figure 5.2 a. This consists of the *GaN* based p-n junction with a MQW region emitting at 450 nm; the p-doped *GaN* layer is 200 nm thick and the MQW region is 100 nm thick [3], [164]. The external cavity consists of two Bragg reflectors with a 224 nm *SiO₂* layer (defining the Fabry-Perot resonance of the cavity) in between them. The number of layers in the Bragg reflector influences the bandwidth and quality of the cavity: the higher number is, the better cavity is with enhanced collimation and the narrower bandwidth is. In our structure each Bragg reflector contains two pairs of *TiO₂/SiO₂*. It is an optimal number both for the bandwidth (which then overlaps with the LED emission bandwidth and does not reflect/filter out most of the emission) and the collimation quality (which is sufficient for metasurface excitation). The Bragg stack was designed in a way that *TiO₂* and *SiO₂* film thicknesses correspond to quarter wavelength of the LED emission (450 nm) in the corresponding material – 42 nm and 77 nm, respectively. When combined, the pair of *TiO₂* and *SiO₂* films introduces a π phase shift relative to the incident wave (completely out of phase); this condition maximizes the reflection for the incident plane wave. The Bragg cavity is covered with a spacer *SiO₂* layer of 200 nm thickness to be used subsequently as a substrate for the *TiO₂* metasurface.

Figure 5.2 b,c demonstrate the numerical simulations of the external cavity *GaN*-based LED reflection, illuminated from the top, as a function of the excitation wavelength and incident angle for the p- and s-polarized light. The cavity resonance is tuned to overlap with the maximum of the electroluminescence (EL) emission of the *GaN* LED (450 nm), hence providing the desired collimation (angular filtering) of the EL signal. Reflection maps were obtained using the transfer matrix method implemented in commercially available Lumerical software package.

Figure 5.2: Device schematics and numerical simulations of the external cavity GaN LED. (a), Scheme of the external cavity GaN LED with two Bragg reflectors forming a cavity and a silica layer as a substrate for the subsequent metasurface deposition. Number of Bragg pairs (TiO_2/SiO_2) inside each reflector is set to 2 (not shown in the schematics); this number is optimized both for the emission bandwidth and the collimation quality. Continued on the next page.

Figure 5.2: (b)-(c), Numerical simulations (transfer matrix method) of reflection maps as a function of the wavelength and the incident angle for p- and s-polarizations correspondingly. The cavity is tuned to overlap with the maximum of the electroluminescence signal of the GaN LED - 450 nm. (d), Emission pattern of the external cavity LED device, obtained with reciprocity principle calculations. Emission wavelength is 450 nm.

Numerical simulations of the external cavity GaN LED emission pattern at 450 nm wavelength obtained using the reciprocity principle are shown in Figure 5.2 d. Emission pattern is transformed from a broad angle Lambertian into a narrow, spatially more coherent emission, which is presumed to be sufficient for further metasurface excitation. The total extracted power (including all directions) is 39% relative to the case of uncovered semi-infinite GaN LED. The extracted power is decreased as the high angle components of Lambertian are filtered. The Purcell factor due to the external nature of the cavity is essentially close to unity as well.

The metasurfaces for functional LED implementation are designed using the common phase-mapping approach, described in detail in previous chapters, where the required output wavefront profile of the optical component (lens, beam deflector, vortex beam generator, etc.) is discretized and mapped using the metasurface elements. We chose the cylinder geometry to ensure the polarization-insensitive response, TiO_2 as a metasurface material possessing negligible losses in the whole visible range. The calculated phase induced by each element as a function of the cylinder diameter and optical transmission of the periodic structure consisting of these elements is demonstrated in Figure 5.3 a. The inset of this figure displays the unit cell geometry of the simulated structure: semi-infinite SiO_2 substrate along the vertical direction with a cylinder of TiO_2 on top; periodic Bloch boundary conditions are applied along the horizontal axes. The system is excited from the glass substrate with a plane wave excitation, the wavelength is 450 nm. The cylinder height is fixed at 460 nm, which maintains

a high transmission level, whereas the diameter is varied, tuning the retardation to cover the whole $0 - 2\pi$ range. The unit cell period is 300 nm and is symmetric along orthogonal in-plane directions. Refractive index of glass is 1.46 and TiO_2 is 2.67 at 450 nm wavelength, taken from experimental ellipsometry measurements.

As a first principle demonstration we use the beam deflecting metasurface consisting of four elements. These elements discretize the $0 - 2\pi$ phase gradient into four steps with $\frac{\pi}{2}$, π , $\frac{3\pi}{2}$ and 2π corresponding to 111 nm, 127 nm, 145 nm and 178 nm in diameter, respectively, taken from Figure 5.3 a. The metasurface efficiency is calculated using 4-element supercell on a glass substrate (inset of Figure 5.3 b) with periodic boundary conditions. The power distribution efficiencies are given in Figure 5.3 b: blue solid line denotes the power into the +1 diffraction order normalized to the incident wave (from the glass substrate), green dashed line is the total transmitted power and red dash-dotted line is the total reflection. The absolute beam deflection efficiency is above 85%, while the one relative to the whole transmitted power is above 95% and the reflection is kept below 12% for the wavelength of interest around 450 nm. Absorption losses in TiO_2 at 450 nm wavelength are low and, therefore, neglected in these calculations. The whole process of metasurface design used simple plane wave excitation and was fully separated from the LED design and simulations. Low level of reflection is one of the important aspects to be considered or, alternatively, a highly reflective metasurface might otherwise destructively interfere with Bragg stacks of the collimating cavity and reduce the overall efficiency of the device.

Figure 5.3: Numerical simulations of metasurface phase and efficiency. (a), Blue solid line - output phase of the metasurface element as a function of the cylinder diameter, the inset shows the metasurface element consisting of TiO_2 cylinder on semi-infinite glass substrate with periodic boundary conditions. Height of the cylinder is 460 nm, wavelength of interest is 450 nm. Dashed green line shows the transmission of the system as a function of the diameter. (b), Beam deflecting metasurface efficiency calculations: blue solid line is the power directed into the +1 diffraction order, green dashed line is the total transmission, red dash-dotted line is the reflection. All the power is normalized to the power of the incident plane wave. Inset illustrates the simulated system, consisting of four TiO_2 cylinders on glass substrate with periodic boundary conditions.

5.2 Experimental results of electrically-driven GaN LED device

The proposed system was experimentally fabricated using the *GaN* LED stack grown with metal-organic chemical vapor deposition (MOCVD) on the patterned sapphire substrate (PSS) [164]. The thickness of the whole LED stack was 7 μm , consisting of a 200 nm thick p-doped layer, a 100 nm thick multiple quantum

well (MQW) region, a 3 μm thick n-doped layer and the rest being undoped layer of *GaN*. Emission is centered at 449 nm wavelength with FWHM of 16 nm. The LED wafer was manually diced and partially covered with an adhesive tape, that defines the areas for electric contacts.

The Bragg cavity was deposited using the ion assisted deposition (IAD) (Oxford Optofab 3000), $\text{TiO}_2/\text{SiO}_2$ pair thicknesses correspond to the numerically simulated 42 nm / 77 nm, the intermediate SiO_2 layer (defining the FP resonance) between the reflectors is 231 nm, and, finally, the SiO_2 spacer layer on top, acting as a base for the metasurface, is 200 nm. The TiO_2 material of 460 nm in thickness was also deposited using the IAD. Next, the adhesive tape protecting the areas for electric contacts was removed. Finally, the thin layer of chromium (*Cr*, ~ 30 nm) was deposited using the electron beam evaporation to increase the conductivity of the system and as a hard mask for the subsequent electron beam lithography (EBL) process.

The metasurface was patterned using the EBL process as described in Methods. Surfaces for n-contacts were then etched using inductively coupled plasma (ICP) - reactive ion etching (RIE) with Cl_2 chemistry, the metasurfaces and areas for p-contacts were protected with simple glass slides of several hundred microns of thickness. The electric contacts were then applied by mechanical pressing of indium (*In*) bids towards the corresponding p- or n-doped *GaN* surface and connection to the direct current (DC) power source. The schematics and real image of the resulting metasurface integrated functional, electrically-driven LED-based device are given in Figure 5.4 a and b correspondingly. Panel b shows the device in operation with the applied voltage of 3 V. The SEM image of the TiO_2 beam deflecting metasurface fabricated on top of the external cavity LED is shown in Figure 5.4 c. Slight tapering of the cylindrical structures was observed due to long etching of the metasurface layer (460 nm) and designed relatively high aspect ratio.

Figure 5.4: Electrically-driven external cavity GaN LED device with metasurface (a), Scheme of the electrically-driven functional LED device. (b), Real image of the metasurface integrated functional LED device with an applied voltage of 3 V. (c), SEM image of the beam deflection TiO_2 metasurface fabricated on top of the external cavity LED device, image taken at an angle of 30° .

The optical characterization of the sample was performed using the back focal plane microscopy setup [152], [104] with a 0.8 NA objective (100x, Nikon, 4 mm working distance), voltage applied to the LED device is 3V. Back focal plane images for different device cases (uncovered LED, with external cavity and with metasurface) are given in Figure 5.5.

Figure 5.5: Back focal plane measurements. (a), (c), (e) Full back focal plane images. (b), (d), (f) Spectrally resolved back focal plane images corresponding to the cut along y-axis in the full back focal plane images. (a), (b) correspond to the uncovered GaN. (c), (d) correspond to external cavity on top of GaN LED. (e), (f) correspond to beam deflecting metasurface integrated with external cavity GaN LED.

Panels a, c and e of Figure 5.5 show the back focal plane images including all wavelengths of the LED radiation bandwidth, all polarizations and all directions. Panels b, d and f of Figure 5.5 are spectrally resolved back focal plane images corresponding to the y-axis cut of the full back focal plane images from panels a, c and e correspondingly. The LED emission is transformed from the broad Lambertian-like shape of the uncovered LED (Figure 5.5 a, b) into collimated, more spatially coherent radiation by means of the external cavity (Figure 5.5 c, d). Finally, the beam deflecting functionality is added shifting the main emission lobe of the collimated beam to 22° ($\text{NA} \sim 0.38$), exactly as designed separately for the metasurface with plane wave excitation without LED structure. Slight non-uniformity in back focal plane images can be explained by the spatial gradient in the excitation of the *GaN* MQW layer. The conductivity of the p-doped layer is much lower as compared to the n-doped layer. Maximum emission intensity is concentrated close to the p-contact and has some slight nonuniform gradient from p- to n- contact. Nonetheless, the metasurface deflection direction is orthogonal to this gradient and, therefore, the LED emission gradient has minimal influence on the beam deflection results.

Extraction efficiencies can be obtained from full back focal plane images by integrating over the top hemisphere (Figure 5.5 a, c, e). However, due to the LED current and, therefore, emission intensity gradient the direct qualitative comparison of the three cases becomes imprecise. Each emission pattern becomes strongly dependent on the location and the distance to the p-electrode. The uncovered LED measurements can only be performed on the areas much closer to the p-electrode, thus, the LED excitation and emission is several times higher. Hence, for the correct measurements of the extraction efficiency we need to minimize the LED emission gradient effect.

5.3 Conclusion

In conclusion, we have proposed a novel concept for compact LED and metasurface integration exploiting the external resonant cavity. The resonant cavity collimates (filters) the Lambertian emission of the plain LED, making it suitable for further metasurface integration. The external character of the cavity minimizes the fabrication processing of the light source, generating more options for the metasurface integration with the ready-made industrial devices. Moreover, external cavity enables a wider range of sources including QDs, OLEDs, fluorescent dyes and other sources demanding a delicate temperature and chemical processing for functional integration with the metasurface. Experimentally, we have demonstrated the efficient beam deflection functionality on the electrically-driven *GaN* LED with an external Bragg cavity.

5.4 Methods

Fabrication of the beam deflecting TiO_2 metasurface

The metasurface was patterned using the EBL process. 30 nm thick chromium (Cr) layer was deposited using the electron beam evaporation (Angstrom EvoVac), followed by spin coating of hydrogen silsesquioxane (HSQ) as a negative resist mask. After EBL (Elionix ELS-7000) exposure the *Cr* mask was further etched with ICP - RIE (Oxford Plasmalab System 100) with Cl_2 gas, followed by TiO_2 etch with CHF_3 . The remaining *Cr* layer was then removed with Cr etchant (Sigma-Aldrich).

Chapter 6

Conclusions and Recommendations

6.1 Conclusions

In this dissertation the idea of metasurface and LEDs integration was proposed and practically implemented in order to combine and enhance the competitive advantages of both in a single device: the highly-efficient, lightweight, bright, long-lifetime and robust light sources with the functional wealth, low-loss and small footprint of dielectric metasurfaces.

First, highly efficient metasurfaces based on dielectric and semiconductor materials (*Si*, *TiO₂*, *GaN*) were designed and fabricated. Functionalities realized include a beam deflector, a polarization beam splitter (PBS), a nonzero orbital angular momentum (OAM) beam generator and a near-unity numerical aperture lens. The advanced metasurface designs with beam deflecting and focusing functionality, presented using *Si* and *TiO₂*, outperform the existing conventional bulky and previously reported flat optical components. The novel concept behind ultra-high angle beam deflecting metasurfaces was explained and demonstrated using *Si* for the red and *TiO₂* for the green and blue parts of the visible spectrum. By engineering the radiation pattern of the individual nanoantenna in the periodic metasurface grating one can control the energy distribution between the grating orders and hence suppress and/or enhance the desired directions. The relative

efficiencies reach 99% theoretically in some cases and $\sim 80 - 90\%$ demonstrated experimentally. Based on these results the near-unity numerical aperture ($\text{NA} \sim 0.99$) lens was designed and fabricated out of *Si*, demonstrating one of the promising applications of the asymmetric nanoantenna metasurface.

Second, *GaN* metasurfaces were etched directly into the regular *GaN* slab for potential LED integration. Despite the low index contrast between the metasurface and the substrate (both *GaN*), the metasurfaces exhibited high efficiencies when excited with plane waves within the designed part of the visible range. The demonstrated beam deflecting and polarization beam splitting devices exhibit high relative efficiencies reaching $\sim 90\%$ in some cases.

Next, the challenge of direct integration of the LEDs with the proposed metasurfaces was revealed. Due to the Lambertian profile of the LED emission, most of the conventionally designed (with a plane wave excitation) metasurfaces exhibit extremely low efficiency and have minimal impact in the plain LED output.

Finally, the solution for full wavefront control of the LED radiation was proposed by means of combining the collimating cavity and metasurfaces. This implies integration with metasurfaces via an intermediate step of spatial coherence enhancement by means of the resonant collimating cavity. Importantly, this concept allows to combine the individual strengths of LEDs and metasurfaces, while keeping the design procedures of the light source and metasurface parts of the device separate, without paying any price in terms of efficiency drops or additional design constraints. In particular, all the functional wealth of the previously reported metasurfaces, as designed to operate under simple planewave excitation, is available for integration with LEDs using the proposed method. We demonstrated the feasibility of the proposed concept using a commercially available *GaP* LED platform, which we endowed with advanced functionalities such as precise beam steering and vortex beam generation with a nonzero orbital angular momentum, by directly integrating a hybrid cavity and a

dielectric (*Si* based) metasurface. The electrically-driven functional LED device was realized on the *GaN* platform by means of the external cavity and *TiO₂* beam deflection metasurface. The external collimating cavity extends the applicability of the approach to ready-made industrial light sources (LEDs, OLEDs, QDs, etc.) by minimizing processing steps required for metasurface integration. The external type of the cavity allows for separate fabrication of the constituent device parts (metasurface on cavity and light source) with further bonding.

In summary, the results presented in this dissertation constitute an important step forward, towards compact, advanced, efficient and integrated optical devices, by leveraging on the attractive platform of LEDs and the emerging field of metasurfaces, which enable unprecedented control of light. We foresee that our method, which may be applied to any other incoherent light source beyond LEDs, will find broad applications in optical communications, Li-Fi, displays, smart lighting and many more.

6.2 Recommendations for future work

Research results in this dissertation have a potential for further development and real-life practical and industrial applications. Several directions of development in order to enhance the device efficiency, to increase the commercial attractiveness and to broaden the concept applicability are suggested:

1. Ultra-high NA lens and ultra-high beam deflecting metasurfaces operation might be expanded to a broadband regime by means of freeform geometries [142], [165], [166] and precise dispersion engineering [124], [129]. Moreover, some machine learning and neural networks techniques such as generative adversarial networks (GAN) may provide a non-trivial optimized solution [167] and assist in broadband dispersion engineering. In that case, metasurfaces might fully substitute the conventional bulky analogues in a broader range of applications.

2. A *GaN* LED platform can be potentially customized for internal resonant cavity integration with metasurfaces. Electrochemical wet etch allows to form nanoporous Bragg stack with high reflection within the *GaN* LED itself [168]. This method involves a high level of doping of several *GaN* layers during MOCVD growth with further selective electric-current assisted wet etch. Hence, this method allows to fabricate a RCLED without dry etching of the sample and avoid all the difficulties encountered.
3. External cavity *GaN* LED results presented in this dissertation can be improved by additional current spreading layer for the p-contact in the LED architecture. Transparent oxides, such as ITO, can be used for this purpose. This step will, potentially, decrease the emission intensity gradient in the LED and increase the overall efficiency of the device platform.
4. Various light sources such as QDs and OLEDs can be used to demonstrate the full potential and flexibility of the internal and external resonant cavity concepts. The list of metasurface functionalities integrated with such random light sources may be extended with a lens, a hologram [131] and a vortex beam generator in the case of electrically-driven devices.

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