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Dielectric Metasurfaces for Controlling Classical and Quantum Light

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Rebecca Aschwanden

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Abstract

This thesis investigates dielectric metasurfaces as versatile nanophotonic platforms for controlling light in linear, nonlinear, and quantum optical regimes. Metasurfaces have emerged as an active area of research as they allow the encoding of a wide range of functionalities within a single layer.

In the first project, a metasurface designed as a multiport beam splitter is presented. The propagation phase is employed to encode tailored diffraction orders and efficiencies into the metasurface. The cascade of two metasurfaces enables the realization of a high-dimensional interferometer and phase-controlled manipulation of the transmitted light. The investigation is further extended to the quantum optical regime using single photons. The metasurface enables the measurement of second- and third-order correlations within a single optical element. Phase-dependent interference in the cascaded system is also demonstrated at the single-photon level. A theoretical matrix-based model reproduces the classical experimental observations.

The second project investigates direction-dependent high-harmonic generation in circular hybrid silicon-silicon nitride nanopillars. In contrast to extended metasurfaces, the nonlinear frequency conversion is achieved within an individual nanoresonator. The structural asymmetry in the nanopillar induces bianisotropic mode coupling, resulting in illumination-direction-dependent field confinement and, consequently, asymmetric nonlinear enhancement. Asymmetric generation of the third, fifth, and seventh harmonic frequencies is demonstrated.

The results presented highlight how tailored nanophotonics can address practical limitations and provide compact, multifunctional solutions.

Kurzfassung

Diese Arbeit untersucht dielektrische Metaoberflächen als vielseitige Plattformen für die Kontrolle von Licht im linearen, nichtlinearen und quantenoptischen Regime. Metaoberflächen haben sich als aktives Forschungsgebiet etabliert, denn sie bieten die Möglichkeit, ein breites Spektrum an optischen Funktionalitäten innerhalb einer ultradünnen Schicht zu realisieren.

Das erste Projekt untersucht eine Metaoberfläche, die als Strahlteiler mit mehr als zwei Ausgängen fungiert. Die Propagationsphase wird verwendet, um ein bestimmtes räumliches Phasenprofil in die Metaoberfläche zu kodieren. Die Kaskade zweier Metaoberflächen erlaubt die Realisierung eines hochdimensionalen Interferometers, um die phasenabhängige Transmission zu untersuchen. Diese Untersuchungen werden in das quantenoptische Regime ausgeweitet, indem einzelne Photonen verwendet werden. Die Metaoberfläche erlaubt die gleichzeitige Messung von Intensitätskorrelationen zweiter und dritter Ordnung mittels eines einzigen ultradünnen Bauteils. Phasenabhängige Interferenz im kaskadierten System wird ebenfalls auf Einzelphotonenebene gezeigt. Eine matrixbasierte Beschreibung ermöglicht die theoretische Modellierung der experimentellen Ergebnisse im klassischen Regime.

Das zweite Projekt untersucht die richtungsabhängige Erzeugung höherer harmonischer Frequenzen in kreisförmigen hybriden Silizium-Siliziumnitrid-Nanoresonatoren. Im Gegensatz zu ausgedehnten Metaoberflächen, die aus vielen einzelnen Nanostrukturen zusammengesetzt sind, erfolgt hierbei die Frequenzkonversion an einer einzelnen dieser Strukturen. Aufgrund der strukturellen Asymmetrie entsteht eine bianisotrope Modenkopplung zwischen Resonanzen, die wiederum zur elektrischen Feldverstärkung führt, die von der Beleuchtungsrichtung abhängig ist. Diese asymmetrische Feldverstärkung führt zur richtungsabhängigen Erzeugung der dritten, fünften und siebten harmonischen Frequenzen.

Zusammenfassend verdeutlichen die Ergebnisse dieser Arbeit, wie mittels maßgeschneiderter nanophotonischer Strukturen kompakte und multifunktionale Bauteile realisiert werden können.

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Chapter 1

Introduction

The discovery of quantum mechanics marked a turning point in physics and, importantly, in our understanding of light. For centuries, the nature of light has been subject to intense debate with competing theories describing light as either a stream of particles or as a wave. On the one hand, the double-slit experiment conducted by Thomas Young demonstrated the wave-like behaviour of light through interference [1]. But on the other hand, at the beginning of the twentieth century, Albert Einstein explained the photoelectric effect by viewing light as quantized, discrete energy packets called photons [2]. These contradictory observations led to the concept of wave-particle duality, which is one of the fundamental principles of quantum mechanics.

A sub-discipline of quantum mechanics, quantum optics, concerned with the interaction of light and matter at the quantum level, emerged in the second half of the twentieth century. A key milestone was the work of Roy J. Glauber, who in 1963 developed the theory of optical coherence and photon statistics, and provided the framework to distinguish between classical and non-classical light states [3]. Around the same time, the field of nonlinear optics was established through the experimental observation of second harmonic generation (SHG) by Franken et al. demonstrating that light-matter interaction can give rise to light with a different frequency when strong electromagnetic fields are used [4]. The observation of SHG became only possible because of the invention of the laser a few years earlier by Maiman in 1960 [5]. The laser, as well as transistors or the atomic clock, are examples of technologies that emerged during what is now termed the first quantum revolution. During this period, fundamental insights into quantum mechanics enabled these transformative technologies. The invention of the laser provided a coherent and controllable light source, which became essential for applications ranging from telecommunication to precision measurements. The transistor revolutionized

information technology by enabling miniaturization of electronic circuits and the integration of billions of these elements on a single chip governed by Moore's Law [6]. These developments laid the technological foundations for today's information society; however, despite their quantum origin, these devices do not fully exploit the unique properties of a single quantum state.

Today, we are witnessing what is often called the second quantum revolution. Now, we are studying the ability to actively control, manipulate, and engineer quantum states [7]. Quantum phenomena such as superposition, entanglement, and non-classical light states are no longer only observed but are harnessed as resources.

Quantum technology is mainly used in three areas, namely quantum computation, communication, and metrology [8–10]. A central challenge in realizing practical quantum applications is scalability, as many optical modes need to be generated and manipulated to allow useful functionality. Currently, the generation as well as the manipulation of a large number of optical modes is still an open research question. Similar to the evolution of classical electronics governed by Moore's law, the development of complex quantum systems requires miniaturization and integration of all functional components onto a compact platform. This includes the generation, manipulation, and detection of quantum states within a unified architecture.

Large-scale photonic networks are not only relevant for quantum technologies but also for all-optical computing. Here, photons serve as the information carrier and their advantages such as high propagation speed are employed to enhance the speed and data transmission rate compared to conventional computing based on microelectronics. Realizing optical computing requires integrated optical switching and logic operations with fast switching speeds and low power consumption [11].

Achieving integrated photonic architectures therefore requires either the miniaturization of conventional optical components or the development of new devices operating on sub-wavelength scales. Currently, conventional optical components such as lenses, mirrors, or beam splitters serve as the fundamental building blocks of both classical and non-classical optical systems. However, the functionality of bulk optical components like lenses relies on gradual phase accumulation during light propagation, which inherently requires macroscopic dimensions limiting miniaturization capabilities. Beam splitters, which commonly consist of two prisms, are used to create interference, superposition and entanglement [12]. They are limited to map two

inputs to two output modes. If more than two modes need to interfere, optical networks consisting of many bulky beam splitters need to be built. Classical cube beam splitters are difficult to miniaturize and integrate into on-chip platforms. Furthermore, their design flexibility is limited, restricting the range of achievable optical functionalities. The limitations of conventional bulky optical components motivate the development of alternative approaches based on artificially structured materials. Metamaterials, composed of sub-wavelength building blocks, enable the engineering of electromagnetic responses that are not found in natural materials. Early theoretical work by Victor Veselago in 1968 predicted the existence of materials with a negative refractive index [13], which later was discussed in the more specific framework of metamaterials [14]. Early experimental observations include optical cloaking [15] and negative refractive index materials [16–19] predominantly in the microwave regime where fabrication constraints are less severe. However, the realization of three-dimensional metamaterials remains technologically challenging due to the required nanometer-scale structuring for optical frequencies leading to a transition towards two-dimensional metasurfaces [20, 21]. They are planar arrays of subwavelength nanostructures that locally control the amplitude, phase, and polarization of incident light. By engineering the geometry and arrangement of these meta-atoms, it is possible to tailor the optical response enabling a wide range of functionalities within an ultrathin device [22].

Metasurfaces have demonstrated remarkable versatility across a wide range of applications. In linear optics, they enable ultrathin implementations of meta-lenses [23–26], meta-holograms [27–29], beam shaping elements [30, 31], imaging [32], sensing [33], and polarization control elements [34–36]. Beyond the linear response, metasurfaces have also been extended to the nonlinear regime, where they are used to generate and enhance harmonic generation [37–40], enable asymmetric generation of harmonics [41], and are used for nonlinear holography [42], nonlinear imaging [43, 44], or as optical switches [45]. More recently, metasurfaces have been explored in the context of quantum optics, where they provide new opportunities for the generation and manipulation of quantum states of light [46, 47].

This thesis studies metasurfaces fabricated from amorphous silicon (Si) and silicon nitride (SiN) for linear, nonlinear, and quantum optical applications. The goal is not only to replace bulky optical elements but to encode further functionalities.

The first scientific goal of this thesis is to investigate a metasurface acting as a multiport beam splitter (i.e., a beam splitter with more than two output modes) and building a high-dimensional interferometer from two cascaded metasurfaces. The metasurfaces consist of rectangular Si meta-atoms that are arranged to form a diffraction grating with customized diffraction orders and efficiencies. The experiment is performed both with classical light and non-classical light generated by semiconductor quantum dots. The results demonstrate that metasurfaces are a suitable platform to achieve multiport beam splitting functionality because many modes can be manipulated at the same time, both for classical and non-classical states of light. Additionally, it is demonstrated that optical networks with useful functionality can be constructed from the cascade of many metasurfaces.

A further scientific goal is to investigate a single meta-atom for nonlinear applications, which enables the direction-dependent generation of high harmonics up to the seventh order. Illuminating the sample from the front side gives rise to a different harmonic signal than illuminating the sample from the back side. This effect is induced by bianisotropic mode coupling. The direction-dependent response is explored across resonant and off-resonant excitation conditions.

The presented work is structured in the following manner: In the **second chapter**, I discuss how metasurfaces control light and present different methods for encoding phases into metasurfaces.

In the **third chapter**, the fabrication of the metasurface is explained, and examples of fabricated metasurfaces are given. Additionally, the optical characterisation of silicon and silicon nitride thin films via spectroscopic ellipsometry is presented, as the process-specific refractive index of the material is essential to know for good theoretical prediction of the metasurface behaviour predicted with numerical calculations.

In the **fourth chapter**, the metasurface acting as a multiport beam splitter is investigated. The chapter begins with a short literature review and an introduction to the fundamentals of quantum optics such as single-photon generation and correlation functions. The design and functionality of the beam splitting metasurface is explained. Next, the single metasurface itself and the cascade of two metasurfaces are characterized with classical light and non-classical light. Finally, the results are summarized and an outlook is given. The **fifth chapter** switches gears and presents the study of direction-dependent high harmonics generation. The concept of bianisotropic mode

coupling is introduced and the design of the nanopillars is presented. Next, the direction-dependent generation of high harmonics is discussed. This chapter also ends with a conclusion and outlook.

Chapter 2

Optical Metasurfaces for Wavefront Engineering

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Light has fascinated humanity for centuries, from naturally occurring phenomena such as rainbows, colourful skies during sunsets or auroras to the colourful illumination of stained glass windows. Depending on the physical regime of interest, light can be described both as a classical electromagnetic wave and as a quantum object consisting of discrete energy quanta, known as photons. The interaction of light with matter can occur in the linear regime, where the material response is proportional to the incident electromagnetic field, or in the nonlinear regime, where the response depends nonlinearly on the electromagnetic field. The presented work spans aspects of linear, nonlinear, and quantum optics. However, the classical electromagnetic description provides the foundation for understanding light-matter interactions and, in particular, the interaction of light with metasurfaces. A key concept in this context is the phase of the electromagnetic field. Controlling the spatial distribution of the optical phase enables manipulation of optical wavefronts, thereby controlling the direction, shape, and classical properties of propagating light. Conventional optical elements, such as lenses, achieve this control through gradual phase accumulation during propagation, whereas metasurfaces only need sub-wavelength length scales to achieve this. At their

interfaces, metasurfaces impose abrupt phase changes and are engineered to provide spatially varying modulations of phase, amplitude, and polarization. This enables compact and highly versatile control of electromagnetic waves on microscopic length scales.

The aim of this chapter is to introduce the physical concepts relevant to metasurfaces. It begins with a discussion of light-matter interactions and optical material properties, including the refractive index and absorption. These concepts are also relevant to spectroscopic ellipsometry, which is discussed in the third chapter, to measure the refractive index and extinction coefficient in silicon and silicon nitride. Building on these fundamentals, the chapter then focuses on scattering processes in metasurfaces and the mechanisms that enable wavefront manipulation. Particular emphasis is placed on how metasurfaces achieve optical phase control.

2.1 Classical Electromagnetic Description of Light

The interaction of electromagnetic radiation with matter is based on Maxwell's equations that govern the dynamics of electric and magnetic fields in space and time. In a vacuum, electromagnetic waves travel at the speed of light $c = 1/\sqrt{\mu_0\epsilon_0}$, where μ_0 is the physical constant of the vacuum permeability and ϵ_0 describes the vacuum permittivity. Introducing a homogeneous dielectric medium into a region of free space modifies Maxwell's equations and the propagation speed to

$$v = \frac{1}{\sqrt{\mu\epsilon}} = \frac{1}{\sqrt{\mu_0\mu_r\epsilon_0\epsilon_r}}, \quad (2.1)$$

where μ_r is the material-dependent relative magnetic permeability and ϵ_r is the material-dependent relative electric permittivity. The constitutive relations describing the displacement field D and magnetic flux density B need to be considered instead of the electric field E and magnetic field H . These constitutive relations connect the electromagnetic field to the material's response through parameters such as the electric permittivity ϵ , magnetic permeability μ , and electrical susceptibility χ , thereby linking the macroscopic field description to the microscopic properties of the medium. Together they describe how electromagnetic waves propagate, are modified, or are attenuated in a material and thus form the basis of describing optical phenomena such as refraction, reflection, dispersion, and absorption. For most optical materials,

the magnetic response is negligible, and the optical behaviour is governed solely by the electric response. This leads to the introduction of the complex permittivity $\epsilon(\omega) = \epsilon'(\omega) + i\epsilon''(\omega)$. The complex permittivity can also be expressed as the complex refractive index $\tilde{n} = n + i\kappa$. The phase speed of the wave in a medium is now calculated as $v = 1/\sqrt{\epsilon\mu} = c/n$, indicating that the propagation of light inside a medium depends on its refractive index n . The extinction coefficient κ accounts for absorption and is linked to the absorption coefficient via $\alpha = 4\pi\kappa/\lambda$. Physically, absorption arises from the transfer of energy from the electromagnetic field to the material, for example, through excitations of electronic transitions or lattice vibrations [48].

The complex refractive index, as well as the complex permittivity, depends on the wavelength, giving rise to dispersion. A microscopic explanation of this behaviour was provided by Lorentz in his theory of matter developed in the 1890s [49]. He considered electrically charged particles bound within matter that experience a restoring force when displaced from their equilibrium position by an external electric field. The modern picture associates these particles with electrons bound to atomic nuclei. It should be noted that the atomic nucleus had not yet been discovered. The nuclear model of the atom was developed after Rutherford's theory of scattering and Geiger and Marsden experiments in the 1910s [50, 51]. The frequently used analogy of an electron attached to a spring as in [48] is also a modern interpretation of Lorentz's restoring force rather than a description used in his original work. When a dielectric material is exposed to an electric field, the internal charges are deformed, meaning that electric dipole moments are generated, which in turn contribute to the total internal field. The resulting dipole moment is called electric polarization

$$P = (\epsilon - \epsilon_0)E = \epsilon_0\chi E. \quad (2.2)$$

The bound electrons are treated as driven harmonic oscillators. Their displacement x depends on the frequency ω of the driving field and the polarization can be written as

$$P = q_e x N. \quad (2.3)$$

Neglecting damping, the displacement is calculated by

$$x = \frac{q_e E}{m_e(\omega_0^2 - \omega^2)}, \quad (2.4)$$

which yields

$$P = \frac{Nq_e^2}{m_e(\omega_0^2 - \omega^2)}E \quad (2.5)$$

for the polarization. If the term for P is inserted into Equation 2.2, the frequency/wavelength-dependence of the refractive index n is derived to be [48]

$$n^2(\omega) = 1 + \frac{Nq_e^2}{\epsilon_0 m_e} \left(\frac{1}{\omega_0^2 - \omega^2} \right) \quad (2.6)$$

Lorentz considered the force of the spring and the driving force of the electric field. Additionally, a phenomenological damping term was added later that leads to the Lorentz oscillator model used to describe optical dispersion and absorption. There are also other dispersion models, e.g. the Drude model, which considers that electrons move freely through the material and are not bound to atoms as is the case for conduction electrons in metals [48]. A more detailed discussion about the dispersion models is presented in Chapter 3 about ellipsometry.

Light cannot only be absorbed inside a material but also transmitted and reflected at planar interfaces. The Law of Reflection states that the angle of incidence equals the angle of reflection [48]. The Law of Refraction relates the angle of incidence θ_{in} and the angle of the transmitted beam θ_t through $n_{in} \sin \theta_{in} = n_t \sin \theta_t$. A more rigorous description of reflection and transmission at interfaces is provided by the Fresnel equations, which follow from applying the boundary conditions of Maxwell's equations to electromagnetic fields at the interface. In general, the reflection and transmission depend on both the polarization of the incident light and the refractive indices n_1 and n_2 of the media. The polarization dependence of light-matter interaction is of interest for this thesis as it is used to achieve a polarization-sensitive response of the metasurface to control phase and amplitude. Furthermore, spectroscopic ellipsometry exploits the different reflection properties of p- and s-polarized light to determine the properties of thin films. For these reasons, the concept of ellipsometry is briefly introduced here. The polarization of a light wave describes the geometrical direction of the oscillation. The most general state is elliptically polarized light, in which the electric field rotates around the beam's direction of travel. Linearly polarization is a special case in which the electric and magnetic fields oscillate perpendicular to each other in a single direction. If a light beam illuminates a sample under oblique incidence, a new reference frame is defined, namely the plane of incidence.

It is the plane defined by the wave vector pointing in the direction of travel and the surface normal. Linearly polarized light can be classified as either p-polarized or s-polarized meaning it is either parallel or perpendicular to the plane of incidence. For s-polarized light, the electric field is perpendicular to the incidence plane. For the reflection and transmission

$$r_s = \frac{n_1 \cos \theta_i - n_2 \cos \theta_t}{n_1 \cos \theta_i + n_2 \cos \theta_t} \quad t_s = \frac{2n_1 \cos \theta_i}{n_1 \cos \theta_i + n_2 \cos \theta_t} \quad (2.7)$$

are valid. When the electric field is parallel to the plane of incidence, the light is referred to as p-polarized and the reflection and transmission are calculated with:

$$r_p = \frac{n_2 \cos \theta_i - n_1 \cos \theta_t}{n_2 \cos \theta_i + n_1 \cos \theta_t} \quad t_p = \frac{2n_1 \cos \theta_i}{n_2 \cos \theta_i + n_1 \cos \theta_t} \quad (2.8)$$

These equations reveal that transmission and reflection not only depend on the refractive index but also on the angle of incidence and polarization. A consequence of this behaviour is the existence of the Brewster angle, which is the angle of incidence where the reflection of p-polarized light vanishes [48].

2.2 Nonlinear Description of Light

Although the primary functionality of metasurfaces discussed in this thesis is based on linear scattering, the last chapter investigates nonlinear frequency conversion in dielectric meta-atoms. For this reason, the basic framework of nonlinear optics is introduced here.

The definition of the electric polarization given in the previous chapter does not fully describe the response of a material system under strong electromagnetic excitation. It is interesting to perform a power series expansion of this quantity with respect to the electric field to reveal other interesting properties, leading to

$$P(\omega) = P_0 + \epsilon_0 \chi^{(1)} E + \epsilon_0 \chi^{(2)} E^2 + \epsilon_0 \chi^{(3)} E^3 + \dots \quad (2.9)$$

The first susceptibility term refers to the linear susceptibility that was also described in the previous chapter, while higher-order terms $\chi^{(2)}, \chi^{(3)}, \dots$ describe the nonlinear susceptibilities [52].

The second-order nonlinear susceptibility $\chi^{(2)}$ describes second-order nonlinear effects such as second harmonic generation (SHG) [4], sum-frequency generation (SFG) [53], and optical parametric oscillation [54]. In SHG, two photons with the same frequency ω interact with the nonlinear medium, and

a photon at 2ω (or half the wavelength) is generated. These processes can only occur in noncentrosymmetric crystals such as GaAs or LiNbO₃; hence, it is not observed in many material systems. Bulk silicon and silicon nitride are centrosymmetric materials and therefore bulk second-order nonlinearities are absent. Consequently, their nonlinear response is typically dominated by odd-order processes, which are observed in this thesis.

The third-order nonlinear susceptibility $\chi^{(3)}$ describes third-order nonlinear effects such as third harmonic generation (THG) [55], electric field-induced second harmonic generation (EFISHG) [56], or the intensity-dependent refractive index [57] that gives rise to self-focusing [58]. During the process of THG, three photons of the same frequency ω interact with the nonlinear medium to generate radiation at 3ω (or a third of the wavelength). It occurs in both centrosymmetric and non-centrosymmetric materials.

Higher-order nonlinear susceptibilities give rise to increasingly higher harmonic orders. In the perturbative regime, the intensity of the n -th harmonic approximately scales as $I_{n\omega} \propto I_{\omega}^n$, resulting in rapidly decreasing conversion efficiency for increasing harmonic order. At sufficiently high optical intensities, nonlinear interactions are no longer limited to second- or third-order frequency conversion, and harmonics of much higher orders can be generated.

Dielectric metasurfaces can enhance the efficiency of the generated harmonics by supporting optical resonances with strong electric field confinement. One example includes the use of Mie resonances, which enable a strong field enhancement for resonant wavelengths [59].

2.3 Scattering and Mie Theory

The functionality of dielectric metasurfaces is based on scattering of electromagnetic waves by subwavelength resonators. By engineering the scattering response of individual resonators, the amplitude, phase and polarization of light can be tailored [59]. Understanding this scattering process is therefore central to the design of metasurfaces.

Scattering is the process by which a nanoparticle removes energy from an incident electromagnetic wave and re-radiates it. The scattering behaviour depends on the ratio between the wavelength and the size of the scatterer. For particles that are much smaller than the wavelength λ , Rayleigh scattering occurs. In this regime, the scattering intensity heavily depends on the wavelength because it scales as $I \propto \frac{1}{\lambda^4}$ [48].

Because meta-atoms have dimensions in the range of the optical wavelength,

their response is governed by Mie scattering rather than the Rayleigh regime. The interaction can no longer be approximated by a simple dipole model. However, a rigorous analytical solution to Maxwell's equations for the scattering of electromagnetic waves can only be obtained for spherical particles and was presented by Gustav Mie in 1908 [60]. While exact solutions are limited to spheres, the underlying physics can be generalized to arbitrary shapes using numerical methods.

In the Mie framework, the scattered field E is expressed as a sum of multipole contributions $E = E_{\text{ed}} + E_{\text{mp}} + E_{\text{EQ}} + E_{\text{MQ}} + \dots$, where E_{ed} , E_{mp} , E_{EQ} and E_{MQ} are the electric fields generated by the electric dipole moment, the magnetic dipole moment, the electric quadrupole, and the magnetic quadrupole. The total scattering cross section and extinction cross section of the particle are expressed as:

$$\sigma_{\text{scat}} = \frac{2\pi}{k^2} \sum_{n=0} (2n+1)(|a_n|^2 + |b_n|^2) \quad (2.10)$$

$$\sigma_{\text{ext}} = \frac{2\pi}{k^2} \sum_{n=0} (2n+1)\text{Re}(a_n + b_n) \quad (2.11)$$

where $k = 2\pi/\lambda$ is the wavenumber [61]. The coefficients a_n and b_n are called Mie coefficients and describe the strength of the respective electric and magnetic multipoles. The index n describes the order of the multipole, with $n = 1$ indicating the dipole, $n = 2$ indicating the quadrupole and so on. The multipole expansion not only provides a mathematical description of the scattered field but also reflects the underlying electromagnetic current distributions excited inside the resonator. The incident electromagnetic field induces resonant displacement currents whose spatial symmetry determines the dominant multipole contribution. For example, oscillating charge separation gives rise to an electric dipole moment, whereas circulating displacement currents generate a magnetic dipole moment [61]. More complex current distributions correspond to higher-order multipoles such as electric and magnetic quadrupoles. An example of such a mode is a non-radiating anapole mode which arise from the destructive interference of electric dipole and toroidal dipole moments. These modes are of particular interest for enhancing nonlinear optical processes due to strong internal field enhancement [62]. Consequently, the optical response of a dielectric nanoresonator can be understood in terms of the multipolar resonances supported by its geometry.

In conventional dielectric materials, the optical response is primarily determined by electric polarization arising from microscopic electric dipoles

induced in the constituent atoms. Since the atomic dimensions are much smaller than the optical wavelength, magnetic contributions are generally negligible. In contrast, dielectric meta-atoms have dimensions comparable to the wavelength of light [63]. The incident field therefore excites resonant electromagnetic current distributions throughout the entire resonator, giving rise not only to electric but also to magnetic multipole resonances. By carefully tailoring the geometrical properties, it is possible to engineer an overlap between magnetic and electric dipole resonances. Since both electric and magnetic dipoles contribute to the scattered field, their interference determines the angular scattering pattern formalized in the Kerker conditions. The first Kerker condition is achieved when electric and magnetic dipole moments are equal in amplitude and phase. Then destructive interference suppresses backward scattering. This effect is used in so-called Huygens metasurfaces, which enable full phase control and simultaneously near-unity transmission [64]. Additionally, if electric and magnetic dipole moments are equal in amplitude but have opposite phases, forward scattering can be suppressed. This is known as the second Kerker condition [64].

In high-index dielectric particles, the magnetic resonance appears at wavelengths comparable to the particle size and can be as strong as the electric resonance. Importantly, the spectral position and strength of these resonances are determined by the particle geometry and material properties, as well as by the refractive index. The scattering behaviour is determined by the ratio of particle radius r and wavelength λ . When $x = 2\pi r/\lambda \ll 1$, the scattering response is dominated by the electric dipole contribution. As the particle size increases, the electromagnetic field can excite increasingly complex current distributions within the resonator, giving rise to higher-order multipole resonances [61].

For many nanophotonic systems, the optical response is dominated by the lowest-order terms, i.e. the electric and magnetic dipole contributions. In this dipolar regime, higher-order multipoles can often be neglected [65], and the scattering can be described in a simplified way in terms of induced dipole moments [66]. Within this approximation, the multipole expansion reduces to the electric and magnetic dipole contributions, allowing the scattering response to be equivalently described in terms of induced dipole moments. In the most general case, the induced electric and magnetic dipole moments are related to the local electric and magnetic fields E_{loc} , H_{loc} through the electric,

magnetic, and magnetoelectric polarizability tensors α [66]:

$$\vec{p} = \alpha_{ee}\vec{E}_{\text{loc}} + \alpha_{em}\vec{H}_{\text{loc}} \text{ and } \vec{m} = \alpha_{me}\vec{E}_{\text{loc}} + \alpha_{mm}\vec{H}_{\text{loc}}. \quad (2.12)$$

The equations for the dipole moments introduce so-called cross-terms that describe electromagnetic and magneto-electric coupling. The cross-polarizability terms α_{em} and α_{me} describe magnetoelectric coupling, whereby an incident magnetic field induces an electric dipole moment and, conversely, an incident electric field induces a magnetic dipole moment. Meta-atoms exhibiting nonzero magnetoelectric coupling are referred to as bianisotropic. Such a response can arise in dielectric nanoresonators lacking inversion symmetry [66]. Bianisotropy provides an additional degree of freedom for tailoring the scattering response of dielectric resonators. Unlike conventional dielectric resonators, whose optical response is governed solely by electric and magnetic polarizabilities, bianisotropic resonators exhibit coupled electric and magnetic responses. This coupling enables asymmetric scattering, polarization conversion, and different optical responses for illumination from opposite directions. Direction-dependent scattering arising from Ω -type bianisotropy is exploited in Chapter 5 to realize asymmetric high-harmonic generation. Therefore, the basic concepts of bianisotropy are briefly introduced here. Depending on the symmetry of the induced response, bianisotropy can be classified into two principal types. In chiral bianisotropy, the induced electric and magnetic dipole moments are collinear with the driving fields. Such structures lack mirror symmetry and exhibit optical activity and polarization-dependent scattering. In contrast, Ω -type bianisotropy is characterized by orthogonal electric–magnetic coupling arising from asymmetric current distributions. This response is commonly realized in geometrically asymmetric dielectric resonators and is particularly important for engineering directional scattering and asymmetric wave propagation in metasurfaces [66].

While the above discussion focused on the response of individual resonators, most optical devices rely on the collective behaviour of many such elements. In particular, arranging sub-wavelength scatterers into two-dimensional arrays enables engineering of spatially varying optical responses. This concept forms the basis of metasurfaces, which are discussed in the following section.

2.4 Wavefront Control with Optical Metasurfaces

Metasurfaces can be defined as artificial two-dimensional materials composed of (periodic) arrays of subwavelength nanostructures (=meta-atoms) designed to manipulate light. They are physical systems whose properties and functionalities do not solely depend on the materials they are made of, but on the arrangement of their sub-elements. Metasurfaces typically consist of a single layer of meta-atoms whose thickness is smaller than the wavelength of the light. They can be considered as optically thin interfaces that impose tailored boundary conditions on the electromagnetic field [67]. The functionality of metasurfaces comes from the ability to locally vary the optical response. It is possible to engineer a specific spatial distribution of phase, amplitude and polarization by varying the size or orientation of the individual meta-atoms. This allows metasurfaces to replicate of traditional optical components such as lenses and wave plates [26, 36]. It is even possible to extend the functionality of these components and to engineer functionalities not possible with bulk optics like the beam splitting metasurface that is presented in Chapter 4. Depending on the design, metasurfaces can either operate in transmission [68] or in reflection [31], and can be tailored for narrowband or broadband operation. In all cases, the underlying principle is wavefront engineering through scattering at the interface.

Meta-atoms are the building blocks of metasurfaces. They can have various shapes, such as nanopillars, nanoposts or more complex geometries including ellipses and are typically fabricated from plasmonic metals or high-index dielectric materials such as silicon [69], silicon nitride [41], or gallium arsenide [70]. The optical response of every meta-atom is determined by its geometry (shape, height, diameter, orientation), material properties and the surrounding environment. By tuning these parameters, the spectral position of the optical resonance as well as the amplitude and phase of the scattered light can be controlled. Due to high ohmic losses in plasmonic metasurfaces [71], the focus in research has shifted towards dielectric metasurfaces. They can support different types of resonances, which arise from the confinement of the electromagnetic field within the meta-atom. Thus, meta-atoms are not only used to engineer the spatial phase profile of light, but can also enhance linear light-matter interactions and nonlinear frequency conversion.

A defining feature of metasurfaces is the subwavelength spacing between adjacent meta-atoms. To mitigate unwanted diffraction effects, the lattice

period is chosen such that $p < \lambda/n$, where λ is the wavelength of light and n is the refractive index of the material. Under this condition, higher diffraction orders are suppressed, and only the 0-th order can propagate into the far field [72].

The functionality of the metasurface arises from scattering, more specifically from the collective interference of the scattered fields from individual meta-atoms. Each meta-atom acts as a secondary source re-radiating light with a specific amplitude and phase, and the superposition of all fields determines the overall response. A central requirement for the design of metasurfaces is the ability to cover the full 0 to 2π phase range while having high transmission or reflection efficiency. This can be achieved through different mechanisms such as resonant phase, geometric phase, or propagation phase [71]. Each of these approaches has advantages in terms of bandwidth, efficiency, and polarization dependence.

The resonant phase is linked to the use of Mie resonances and it relies on the wavelength-dependent response of a meta-atom. Each meta-atom can be viewed as a small harmonic oscillator, which is driven by the incident electromagnetic field. The phase delay of the light field interaction with the meta-atom changes from 0 to π as the wavelength sweeps over the width of the resonance. Different phase shifts at a fixed wavelength can be obtained by using meta-atoms of different sizes and thus different resonance frequencies. Resonant phase control is inherently dispersive, which makes it only suitable for narrowband applications [72].

A second approach is the geometric phase, also called Pancharatnam-Berry (PB) phase [73, 74]. It relies on different orientations of anisotropic meta-atoms and not on the spectral response. When such a structure, e.g., an elliptical pillar or nanorod, is rotated by an angle θ , the light acquires twice the phase shift $\phi = \pm 2\theta$. The PB phase is broadband because it only relies on the orientation of the nanoantenna, but not on its exact size. On the other hand, it relies on the circular polarization state and the polarization state is also inverted upon interaction with the metasurface [71].

The third approach is the propagation phase. It is implemented by using waveguide-like meta-atoms such as nanopillars or nanoposts that support guided modes along their height. Light propagating through such a structure accumulates the phase $\phi_{\text{prop}} = k_0 n_{\text{eff}} h$, where n_{eff} is the effective refractive index of the guided mode, and h is the height of the structure. The accumulated phase can be varied by changing the lateral dimensions of the nanopillar,

which changes the effective refractive index [75]. Propagation phase metasurfaces operate away from resonances, thus having reduced absorption losses and are therefore suitable for high-efficiency transmission applications. This approach is also generally less wavelength-sensitive compared to resonant phase control, enabling broader bandwidth operation. Additionally, propagation phase control is polarization-independent if isotropic meta-atoms are chosen. A drawback from a fabrication perspective is the need for rather large thicknesses but small lateral dimensions, which can result in increased fabrication complexity due to high aspect ratio structures.

The ability of metasurfaces to impose a spatially varying phase profile alters the classical behaviour of light at an interface. The generalized Law of Refraction was formulated by Yu et al. in 2011 based on Fermat's principle [76]. If an interface has an abrupt spatially varying phase shift $\phi(\vec{r}_s)$, the Law of Refraction needs to be extended to the Law of generalized Refraction as the conventional law assumes a continuous phase of the electromagnetic field at the interface, meaning that the phase evolves smoothly at the interface. The presence of the phase gradient $d\phi/dx$ along the interface influences the transmission and reflection of the beam. Hence, a new generalized Law of Refraction needs to be formulated, which implies that the refracted beam can have an arbitrary direction:

$$n_t \sin \theta_t - n_{in} \sin \theta_{in} = \frac{\lambda}{2\pi} \frac{d\phi}{dx} \quad (2.13)$$

With metasurfaces, arbitrarily phase shifts $\phi(x)$ can be introduced, e.g. a linear phase gradient leads to the deflection of the beam or a quadratic phase distribution $\phi(x, y) = \frac{k_0}{2f}(x^2 + y^2)$ produces a focusing effect at focal distance f analogous to a conventional lens. The different phase engineering mechanisms serve as the tool to create the desired phase gradient.

Chapter 3

Nanofabrication and Optical Characterization of Dielectric Metasurfaces

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Modern nanofabrication has its roots in the development of microelectronics. A major milestone was the invention of the integrated circuit by Jack Kilby in 1958 [77] and Robert Noyce in 1959 [78]. Both demonstrated for the first time that multiple electronic components can be integrated on a single semiconductor substrate [79]. This concept of monolithic integration enabled the miniaturization of electronic systems and laid the foundation for semiconductor technology. Driven by Moore's Law [80], the continuous reduction of feature sizes in integrated circuits stimulated the development of increasingly advanced nanofabrication techniques that allow the fabrication of feature sizes down to 8 nm [81]. These technological advances are not only used in microelectronics but also have become essential for nanophotonics. In particular, the fabrication of metasurfaces requires precisely controlled nanostructures with typically sub-wavelength dimensions of a few tens to a few hundred nanometers. Many techniques used to fabricate such structures are directly derived from processes originally developed for the semiconductor industry. Technologies that once enabled the integration of electronic circuits now provide the foundation for the realization of complex photonic

nanostructures such as metasurfaces.

In this work, metasurfaces based on amorphous silicon and silicon nitride are fabricated using top-down processes. This chapter first discusses the motivation for using silicon and silicon nitride as material platforms. Subsequently, the fabrication processes employed for the realization of the different metasurfaces are presented. Lastly, this chapter introduces spectroscopic ellipsometry, which represents the key characterization technique for determining the optical constants of both materials.

3.1 Silicon and Silicon Nitride as Materials for Metasurfaces

Silicon and silicon nitride are among the most important material platforms for nanophotonic devices due to their favourable optical properties, CMOS compatibility, and the availability of mature fabrication processes. In particular, their high refractive indices, strong nonlinear responses, and compatibility with established semiconductor fabrication techniques make them attractive for metasurface applications. The relevant properties of both materials are discussed in the following sections.

Crystalline silicon (c-Si), commonly used in silicon-on-insulator (SOI) wafers, was originally developed as a platform for microelectronics and is widely used in devices such as transistors. Consequently, highly optimized fabrication processes, including electron-beam and optical lithography, established resist systems, dry and wet etching techniques, and high-quality wafer growth, are readily available [82]. Besides crystalline silicon, amorphous silicon (a-Si) is also widely used in nanophotonics [69, 83]. Although both materials consist of silicon, their structural differences lead to distinct optical and electronic properties. Unlike crystalline silicon, amorphous silicon lacks long-range atomic order. Variations in bond lengths and bond angles broaden the electronic density of states, resulting in localized band-tail states and defect states caused by dangling bonds. These defects strongly influence carrier recombination and the electronic properties of the material [84]. Nevertheless, a-Si retains the short-range covalent bonding structure of crystalline silicon and therefore exhibits semiconductor-like properties. The band gap of crystalline silicon is approximately 1.1 eV, corresponding to a wavelength of around 1100 nm, whereas amorphous silicon exhibits a larger band gap of approximately 1.7 eV

(730 nm) [84]. Reported values vary depending on hydrogen concentration and deposition conditions, with band gaps ranging from 1.3 eV to 1.9 eV reported in literature [85]. The increased band gap of a-Si reduces absorption at shorter wavelengths, making it advantageous for nanophotonic applications in the visible and near-infrared spectral range [83].

A further advantage of amorphous silicon is its compatibility with thin-film deposition processes, allowing uniform deposition over large areas on different substrates with controlled thicknesses [86]. During deposition, hydrogen incorporation can reduce the density of dangling bonds and improve the material properties, resulting in hydrogenated amorphous silicon (a-Si:H), which was first investigated in the 1960s [84]. Plasma-enhanced chemical vapour deposition (PECVD), which is also employed in this work, is a commonly used deposition technique, although other approaches such as sputtering are also possible [85]. The optical and electrical properties of a-Si depend strongly on the deposition conditions, particularly the hydrogen concentration [87, 88].

For photonic applications, the optical properties are of primary importance. Amorphous silicon provides low absorption down to approximately 730 nm due to its larger optical band gap. Additionally, amorphous silicon exhibits a high refractive index ($n \approx 3.7$ at 800 nm), enabling strong optical confinement, high field enhancement, and compact photonic structures. For example, SOI waveguides typically require widths of approximately 400 nm for single-mode operation [89]. Furthermore, silicon exhibits a strong third-order nonlinear response, making it attractive for applications such as all-optical switching, frequency conversion, and Kerr nonlinear optics. Since silicon is centrosymmetric, bulk second-order nonlinear processes are forbidden within the electric dipole approximation. Therefore, nonlinear interactions in silicon are dominated by odd-order processes, including third-harmonic generation [59].

Silicon nitride (SiN) represents a complementary material platform for integrated nanophotonics due to its low optical losses and broad transparency window. In contrast to silicon, silicon nitride remains transparent at wavelengths down to approximately 400 nm, enabling operation in the visible and near-infrared spectral regions [90]. Furthermore, SiN exhibits negligible two-photon absorption at telecommunication wavelengths, which is beneficial for nonlinear optical applications requiring high optical intensities [91]. Compared to silicon, silicon nitride has a lower refractive index ($n \approx 2.0$ at 800 nm), resulting in weaker optical confinement. Similar to silicon, its properties can be tuned through different deposition techniques, including

low-pressure chemical vapour deposition and PECVD, by controlling the SiN_x stoichiometry and hydrogen incorporation [90, 92]. Due to their compatible fabrication processes, silicon and silicon nitride can also be combined in multilayer architectures, enabling hybrid metasurfaces. An example of such a hybrid silicon–silicon nitride structure is presented in Chapter 5, where it is used to demonstrate direction-dependent high-harmonic generation.

3.2 Fabrication of Dielectric Metasurfaces

In this work, nanofabrication using techniques such as electron beam lithography (EBL) combined with dry etching is employed. A schematic overview of the fabrication is shown in Figure 3.1. The workflow consists of sample cleaning, dielectric thin-film deposition, electron beam lithography, metal mask deposition, and reactive ion etching. Each step is described in detail in the following sections.

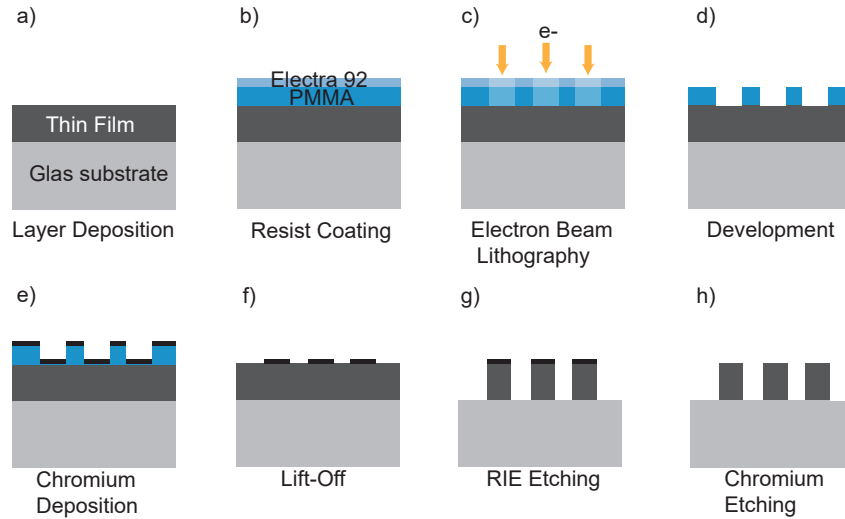


FIGURE 3.1: Fabrication flow of dielectric metasurface: (a) A silicon or silicon nitride layer is deposited on a glass substrate. (b) The dielectric layer is coated with a positive-tone electron-beam-sensitive resist, which is patterned by electron beam lithography. (c) Exposure of the resist leads to local bond breaking, enabling selective development of the exposed regions. (d) The patterned resist is developed, leaving the desired nanostructure mask. (e) A thin chromium layer is deposited onto the sample. (f) A lift-off process is performed, removing the remaining resist and leaving only the chromium on the surface. (g) The pattern is transferred into the dielectric layer via reactive-ion etching, where chromium acts as an etch mask and protects the covered regions, resulting in the formation of nanopillars. (h)

In the final step, the chromium mask is removed by wet etching.

The fabrication process begins with cleaning of the fused silica substrates using a standard ultrasonic bath to remove the protective coating that was applied to the wafer for cutting. Samples are first immersed in acetone for

10 min to remove organic contaminants, followed by 5 min in isopropanol and 5 min in deionized water to eliminate non-polar residues left by the acetone. The samples are dried with a nitrogen gun. Finally, the samples are treated in an oxygen plasma for 5 min. The plasma removes residual contaminants still left on the fused silica substrate.

3.2.1 Plasma-Enhanced Chemical Vapour Deposition

In this work, both amorphous silicon and silicon nitride dielectric layers are deposited by plasma-enhanced chemical vapour deposition (PECVD). In PECVD, plasma is used to provide the energy required for gas-phase dissociation and surface reactions, enabling deposition at relatively low temperatures compared to other CVD processes. Since the reactions are not thermodynamically controlled, film composition and optical properties depend on deposition parameters such as pressure, temperature, and gas flow [82]. Because of this, sub-stoichiometric films can be deposited, and careful optical characterization of the grown films is necessary.

The system (Oxford Plasmalab 80) used consists of a heated table serving as the lower electrode, a top electrode, a gas inlet, and a vacuum pumping system. The plasma is induced between the grounded lower and the top electrode, which is driven by a radio frequency (RF) of 13.56 MHz. For amorphous silicon deposition, a gas mixture of 2 % silane (SiH_4) diluted in argon (Ar) with a flow rate of 400 sccm is injected through the gas inlet into the chamber. The substrate temperature is maintained at 300 °C, and the chamber pressure is held at 1 Torr. Under these conditions, a growth rate of approximately 20 nm min⁻¹ is obtained depending on the number of samples placed in the chamber. As the main reaction in the plasma, SiH_4 molecules dissociate into silyl radicals SiH_3 through electron impact dissociation, as well as smaller fragments (SiH_2 and SiH) and ions. These species are adsorbed onto the surface of the substrate and form the hydrogenated amorphous silicon layer (a-Si:H) [93]. Increasing the substrate temperature reduces the hydrogen content in a-Si:H films due to enhanced hydrogen desorption and reduced hydrogen incorporation, resulting in improved optical film quality [94]. To prevent sample cracking, the substrate table was cooled to 150 °C before the samples were taken out of the PECVD chamber.

Silicon nitride films are deposited using the same silane-argon mixture with an additional nitrogen (N_2) flow of 20 sccm to provide nitrogen atoms. The pressure and table temperature remain unchanged. Under these conditions, a growth rate of 12 nm min^{-1} is achieved. For the fabrication of Si-SiN double-layer structures used in high-harmonic generation experiments, a 1200 nm thick silicon layer was first deposited, followed by a chamber cleaning process. Subsequently, a 900 nm thick silicon nitride layer was deposited.

3.2.2 Electron Beam Lithography

As the next fabrication step, lithography is performed to laterally pattern the sample. There are many different ways to achieve lateral structuring of a sample. Examples include optical lithography, electron beam lithography, focused ion beam lithography or laser lithography [95]. In optical lithography, a reticle is imaged onto the sample, and the mask pattern is transferred to the resist [82]. This technique allows high throughput; however, the resolution depends on the wavelengths of the light. When moving to smaller and smaller wavelengths in the UV region, technical difficulties such as absorption in lenses occur. Solving these difficulties by using mirrors instead of lenses is a possibility; however, it is extremely costly and thus not suitable for R&D purposes [96]. If high resolution is the ultimate requirement but high-throughput is not important, EBL is often the method of choice [97]. In EBL, electrons are directly used to write onto the resist, thus requiring no illuminated reticle [98]. EBL allows a higher resolution due to the smaller electron wavelength, leading to a smaller spot size and smaller feature sizes. However, the factor limiting the resolution is not only the spatial dimension of the electron beam but rather aberrations, scattering, the resist resolution [98], and the resolution of subsequent fabrication steps. Conditions to achieve a high resolution include a high electron energy, a small aperture, a low beam current, and a low-sensitivity resist with a thin thickness [99] as they reduce aberrations and scattering.

The sample needs to be prepared before it can be coated with an electron beam-sensitive resist. It is cleaned again with acetone, IPA, deionized water, and the plasma treatment previously described to remove any dust particles from the sample surface. The spin coating process consists of three different steps. First, a thin layer of an adhesion promoter (Allresist, AR 300-80) is spin-coated onto the sample with a speed of 4000 rpm for 30 s and then annealed

on a hotplate for 120 s at 165 °C. After the baking, the sample is rinsed in isopropanol. The adhesion promoter is used to ensure good adhesion of the subsequently applied resist to the sample surface.

As the second step, the electron beam-sensitive polymethacrylate (PMMA) resist is used. PMMA is a standard electron-beam resist that enables very small feature sizes (6 nm [100]) while maintaining low sensitivity and ease of use. Low sensitivity means that a higher dose is necessary to clear the resist. This resist is a positive-tone resist, meaning that exposed areas are developed away. In contrast, in a negative-tone resist, the exposed areas remain after development. For the fabrication of the different metasurfaces, two types of resists have been used. The thinner resist (Allresist, AR 679.03), resulting in a thickness of 160 nm, was used as the default choice, but a 270 nm thick resist (Allresist, AR 679.04) was necessary for the thicker Si-SiN nanopillars to ensure good lift-off quality. This thicker resist was also used for the fabrication of the nanoholes in the SiN film, as in this case the PMMA was used as the etch mask. The thinner resist was spin-coated onto the sample with a speed of 4000 rpm for 30 s and then annealed on a hotplate for 2 min at 165 °C to remove the ethyl lactate solvent. For the thicker resist, the duration was increased to 60 s while all other processing parameters remained unchanged.

As a final preparation step, the sample was coated with Electra 92 resist (Allresist, AR-PC 5090), a conductive polymer resist based on polyaniline dissolved in water and isopropanol. The conductive layer suppresses charging during electron beam lithography on insulating substrates. Without such a layer, electrons accumulate in the substrate because they cannot dissipate to the grounded sample holder, generating electric fields that deflect the incident electron beam and distort the written pattern. Electra 92 was spin-coated in two steps. First, for 3 s at 1000 rpm, followed by 30 s at 4000 rpm. The coated sample was then baked on a hotplate at 90 °C for 2 min.

Next, the resist is exposed by the electron beam. The lithography system used is a Raith Voyager with an acceleration voltage of 50 kV. The aperture can be set to 10 μm , 30 μm , or 60 μm . A smaller aperture increases the resolution, but it also decreases the beam current, thereby leading to larger exposure times. In this work, aperture sizes of 30 μm and 60 μm are used as they allowed for sufficient resolution. The electron beam directly draws the designed pattern into the resist, leading to breaking of the PMMA polymer chain [98]. These fragments are removed during development. However, every resist requires a certain electron dose (number of electrons per area) to achieve scission of the

chemical bonds. The required nominal dose also depends on the shape of the pattern (e.g. circle vs rectangle), the pattern density, the underlying material thickness, the resist thickness, etc. Therefore, it is difficult to give a universal value. Here, a dose of $300 \mu\text{C cm}^2$ was enough to clear the 160 nm PMMA resist, but generally, each sample contained several copies of the metasurface written with different doses. Overexposing the resist leads to an increase in the size of the individual meta-atoms, while underexposing reduces the size, although below a certain dose, the resist will not be fully cleared during development.

An important effect that occurs during the exposure is the proximity effect, which describes how energy from incident electrons can extend to adjacent areas due to scattering, which leads to overexposure of dense areas [98]. In the material system, different interactions can occur. Electrons can elastically scatter from atoms (either of the resist or deeper materials) due to the large mass difference. This process is governed by Rutherford's elastic scattering cross section [101]. The electron does not lose its energy, but changes its direction. A second type of interaction is inelastic electron-electron scattering, described by Bethe's continuous slowing down approximation model [101], in which the electron passes some of its energy to the second electron. In addition, the inelastic interaction can lead to the formation of secondary electrons. If the transferred energy exceeds the binding energy, these electrons are emitted as secondary electrons. They typically only have low energy and travel a few nanometers in the resist [99].

These processes result in a lateral spreading of deposited energy. Forward scattering broadens the primary beam within the resist, while backscattered electrons originating from the substrate contribute to long-range exposure. In addition, the generation of secondary electrons further distributes the deposited energy on the nanometer scale [102]. The electron scattering is influenced by the electron beam energy, the density of the substrate, and the resist thickness. The higher the electron beam energy, the smaller the scattering angle and the narrower the distribution. A substrate with a higher density or higher atomic number leads to more backscattering, and a thinner resist allows a better resolution as discussed previously [99]. The collision of electrons in the material, the deflection direction and thus the trajectory of the scattered electrons are random processes which can be simulated with a Monte Carlo simulation. If thousands of electron trajectories are simulated, a pear-shaped energy distribution profile displayed in Figure 3.2c emerges. This

distribution can be mathematically described by the point spread function (PSF). It describes the deposited energy distribution in resists from a single point of incidence. By convolving the PSF with a Gaussian beam, the real electron beam in the system is described.

As the proximity effect is unwanted, several approaches have been developed to mitigate the reduced resolution. These methods include dose correction, pattern size compensation and background exposure compensation [99]. The first two correction pathways are used in this work and are shown in Figure 3.2. Panel a shows the adaptation of the shape. The goal is to pattern rectangular nanoposts; however, because of the proximity effect, sharp corners often are rounded. To counteract this effect, additional smaller rectangles are placed on every corner to achieve overexposure of the corner, which in the ideal case leads to a sharper feature. This idea might work for larger feature sizes, but for my samples, the nanoposts are still too round, as depicted in the SEM micrograph. Only a rectangle size of 10 nm was tested for my feature sizes, ranging from 100 nm to 155 nm, which was not sufficient to achieve sharp corners. Therefore, the second pathway of proximity effect correction by dose adaptation was followed. The commercial software "Beamer" (GenISys) was used to calculate the required dose values to achieve uniform exposure and sharp features. As a first step, the PSF of the material system was calculated and used for the dose correction. The pattern is divided into regular sub-shapes (rectangles), and the dose of each smaller geometry is adjusted to achieve equal energy distribution. This method increases the number of vertices and thus the size of the GDS file that is written, which results in significantly increased writing times (>10 h compared to ≈ 3 h).

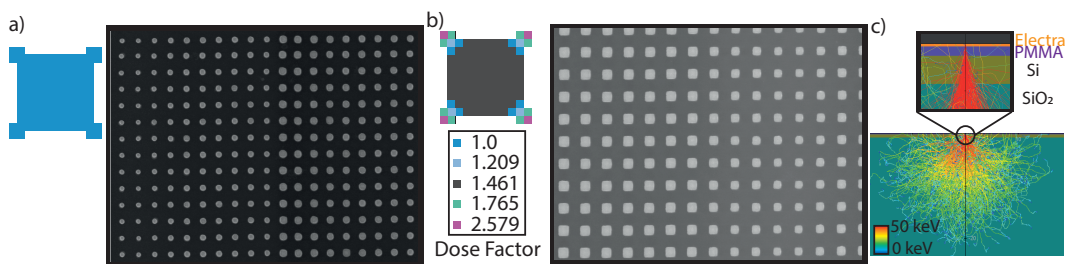


FIGURE 3.2: Comparison of proximity effect correction strategies in electron beam lithography: (a) Pattern size compensation, where additional smaller exposure shapes are placed at the corners of the main pattern to increase the local dose and compensate for proximity effects. However, the resulting scanning electron micrograph shows that this approach does not yield sufficiently sharp features. (b) Improved proximity effect correction using optimized doses, resulting in sharper and better-defined structures. (c) Simulation of the electron point-spread function in the resist and underlying dielectric layer, illustrating electron scattering and the resulting lateral broadening.

As a final lithography step, the resist needs to be developed. First, the conductive layer is removed by placing the sample in a water bath for 30 s. Next, the PMMA resist is developed in methyl isobutyl ketone (MIBK) based developer (Allresist, AR 600-56). The 160 nm thin layer is developed for 85 s, while the 270 nm thick resist is developed for 110 s. Finally, the samples are placed in an isopropanol bath for 30 s to stop the development.

3.2.3 Etchmask Deposition

The lateral pattern needs to be transferred into the layer, so that nanoposts or nanopillars are formed. This is achieved by the combination of etch-mask deposition, lift-off and etching. PMMA exhibits poor etch resistance during silicon etching. Therefore, a thin metal layer is deposited to serve as a hard mask. The deposition process must satisfy two key requirements. The process temperature must be below the glass transition temperature (T_g) of the polymer resist, and the deposition must be highly directional. If the process temperature exceeds T_g , the resist softens, becomes viscous, and loses its shape. For PMMA, the glass transition temperature is approximately 105 °C [100]. High directionality is equally important to prevent deposition on the resist sidewalls, ensuring a clean lift-off after metal deposition. Electron beam evaporation fulfils both requirements and was therefore chosen for hard-mask deposition.

In electron beam evaporation, a material is heated by an electron beam and evaporated. It can be used for a wide range of materials such as chromium, silver, and gold. The deposition rate depends on the distance and relative position between the substrate and the evaporation source. It is mathematically described by Knudsen's cosine law. This law states that the deposition rate is a function of evaporation source to sample distance, the angle of the sample with respect to the evaporation source, the temperature-dependent vapour pressure of the material and the pressure in the evaporation chamber [99]. One deduction of this law is that a sample placed at a certain distance from the evaporation source will always have a non-uniform thickness distribution. The non-uniformity can be decreased by increasing the distance between the sample and the source, which also leads to a less efficient use of source material [99].

In our case, chromium is selected as a mask as it withstands the silicon etching and has good adhesion to silicon. The sample is placed on the sample holder,

and the chamber is evacuated to a pressure of approximately 10^{-6} mbar. The electron beam heats the chromium inside the crucible. The film is evaporated at a rate of 0.1 nm s^{-1} while the film thickness is measured with a quartz crystal monitor. The thickness of the chromium needs to be thick enough so that the mask withstands the full etching process, but also thin enough that the resist can be lifted off. A guideline is that the resist needs to be at least three times as thick as the mask. For all samples except the Si-SiN pillars, a 20 nm thick layer was used. As the thickness of the Si-SiN nanopillars was $2.1 \text{ }\mu\text{m}$, a 45 nm thick Cr layer was deposited.

After the mask deposition, the so-called lift-off is performed. Here, the remaining resist with excess Cr is removed, and only the Cr mask remains so that the sample can be etched. For the lift-off process, the sample was first immersed in an acetone bath at 65°C to dissolve the PMMA resist. Subsequently, the sample was ultrasonicated at 50°C while remaining in the acetone bath to facilitate the removal of the resist. Afterwards, the sample was transferred to a resist remover (Allresist, AR 300-76) containing dimethyl succinate (DMS), which is an ester-based organic solvent derived from succinic acid. Due to its polar ester groups and high boiling point, it can effectively penetrate, swell, and dissolve PMMA while remaining in contact with the polymer for an extended time. The remover was heated to 85°C and ultrasonicated for up to 2 min. The immersion time in acetone and DMS was adjusted between 15 min and 60 min, depending on the pattern density of the metasurface. Higher pattern densities required longer lift-off times to ensure complete removal of the residual resist and unwanted Cr, as denser patterns reduce the open area that allows solvents to penetrate into the resist. Additionally, the initial ultrasonication step was found to significantly improve the lift-off quality as it helps to remove the resist mechanically. After completion of the lift-off process, only the EBL-defined Cr pattern remained on the silicon substrate.

3.2.4 Reactive-Ion Etching

As the next step, inductively coupled plasma reactive ion etching (ICP-RIE) is used to transfer the metasurface pattern into the dielectric material. An inductively coupled radio frequency (RF) source generates a high-density plasma, while a separate RF bias controls ion energy, enabling high etch rates and anisotropic profiles. This allows control of the ion energy and ion

flux separately, giving greater control of the etching quality. Energetic ions accelerated towards the substrate can eject atoms from the surface. This purely physical sputtering contributes, on the one hand, to material removal and also helps remove native oxides and contamination layers from the surface. In addition, ions can directly participate in chemical reactions with the surface atoms. Ion bombardment can dissociate adsorbed gas molecules on the surface, generating reactive radicals which further contribute to the chemical etching process. These radicals can diffuse across the substrate surface and react with the material atoms to form volatile compounds, which are removed from the chamber [98].

To achieve highly vertical sidewalls and high aspect ratios, an exceptionally well-controlled anisotropic etching process is necessary. The quality of the etching depends on many externally controlled parameters such as gas flow rates, RF power, chamber pressure, substrate temperature and bias voltage [99]. Due to the number of parameters and sensitivity to even small changes, the etching recipe needs to be thoroughly optimised. Another aspect that needs to be considered during optimization is the etch mask selectivity. Selectivity describes how much the mask material is etched compared to the material. As a rough estimate, one can etch 1 μm of Si with a 20 nm thick Cr layer.

For the optimization only the gas flow rates and RF power (DC bias) are varied, but the rest of the parameters are kept constant at previously optimized values that had proven to be successful for this specific ICP-RIE apparatus. The power of the ICP source and the pressure were kept constant at 900 W and 10.5 mTorr, respectively. During the course of the different projects, the recipe needed to be optimized several times because the RIE machine used was also used to etch other materials, which led to growing amounts of residue at the chamber walls, altering the chamber conditions, and thus reducing the etch quality. Since these residues cannot be completely removed by the standard chamber cleaning procedure, a manual cleaning of the chamber was required approximately once per year. After such maintenance procedures, the etching parameters needed to be optimized again. Figure 3.3a illustrates several typical non-ideal etching profiles that occur when non-optimized process parameters are used. Panel b shows the optimized etching results for Si nanoposts, Si nanopillars, and Si-SiN double-layer pillars. During the optimization process, several issues were observed, including overly isotropic etching, mask undercut, sidewall tapering and excessive polymer passivation. Isotropic etching and the mask undercut result from excessive chemical etching by too many F-radicals, so reducing the SF_6 flow reduces this issue.

Also, a higher bias voltage, higher C_4F_8 flow and lower pressure help to increase the anisotropy. Sidewall tapering describes tilted sidewalls resulting from too little passivation during the etching. Even small variations in the etching parameters can significantly influence the resulting structure geometry. Second, changing one gas flow also changes other parameters such as the DC bias. Optimization of the etching process is a highly empirical process.

In addition to these effects, the etching behaviour can also be influenced by

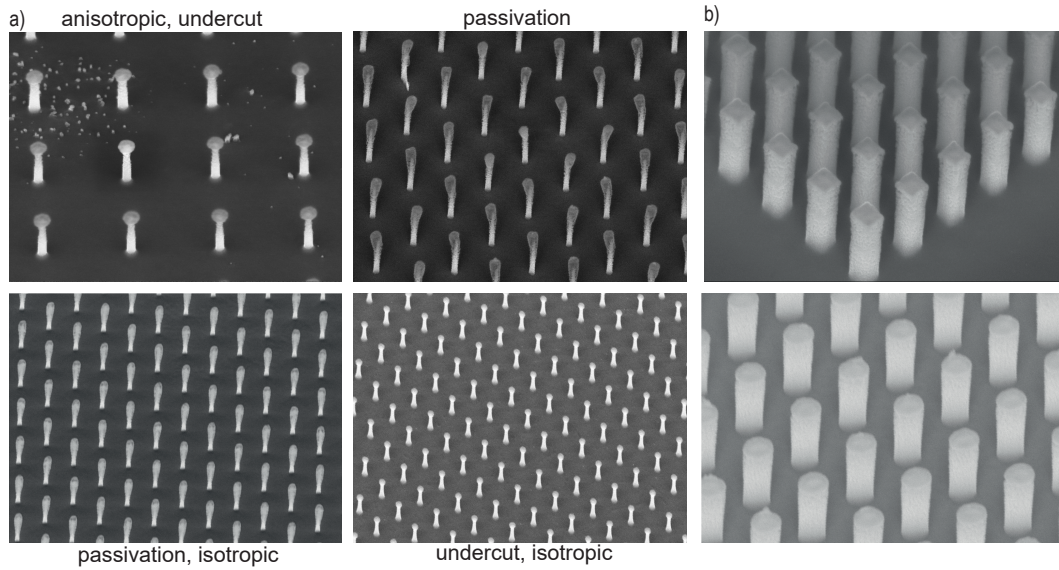


FIGURE 3.3: Scanning electron micrographs of etched silicon metasurfaces fabricated by reactive-ion etching: (a) Etching results obtained with non-optimized process parameters. Mask undercut occurs when lateral etching beneath the chromium mask leads to reduced pillar width. Isotropic etching refers to a non-directional etch profile, resulting in uniform material removal in both vertical and lateral directions. An excessively high passivation gas flow can additionally lead to unwanted sidewall deposition, degrading the verticality of the structures. (b) Metasurface structures fabricated using optimized etching parameters show well-defined and vertical sidewalls.

pattern-dependent phenomena. For example, in areas with a high density of structures, the local consumption of reactive species can reduce the etch rate compared to isolated features. This effect is commonly referred to as microloading [103]. Another frequently observed phenomenon is microtrenching, where enhanced etching occurs near the bottom corners of the structures due to ions that are reflected from the sidewalls and subsequently impact the substrate. Furthermore, charge accumulation at insulating interfaces can lead to notching, where increased lateral etching occurs near the material interface [104].

The substrate temperature can also influence the etching behaviour. Higher temperatures generally increase the rates of surface reactions and the mobility

of adsorbed species, which often enhances the chemical contribution to the etch process and can lead to increased lateral etching [99]. One solution to reduce the substrate temperature during etching is helium backside cooling. For this, helium is injected between the wafer on which the sample sits and the wafer table to allow for effective heat transfer between the wafer and the cooled wafer table. The advantage of helium is its high thermal conductivity and chemical inertness, so that it does not react with the process gases.

After discussing the factors influencing the etching, the optimization will be described in the following section. For silicon etching, a mixture of sulphur hexafluoride SF_6 and octafluorocyclobutane C_4F_8 is used. The silicon is etched by SF_6 , which is dissociated into fluorine radicals in the plasma and chemically reacts with silicon to form SiF_4 , which is pumped out of the chamber. On the other hand, C_4F_8 dissociates into CF_x , forming a fluorocarbon polymer, which leads to sidewall deposition and protects the surface from F radicals. During the process, there is a competition between the fluorine-based chemical etching and the fluorocarbon polymer passivation that is removed by ion bombardment.

This process concept is closely related to the Bosch process [105], which is widely used for deep silicon etching. In the classical Bosch process, etching and passivation steps are performed sequentially. During this process, silicon is etched isotropically by fluorine radicals, while the subsequent passivation steps deposit the fluorocarbon polymer layer on the sidewalls. Repeating these cycles results in highly anisotropic etching but produces a periodic sidewall roughness known as scalloping. In our process, both gases are present in the chamber simultaneously, balancing passivation and etching, which reduces scalloping effects but requires careful tuning of the gas ratio and ion energy. During the process optimization over time, different suitable gas mixtures were found and presented in Table 3.1. The etching time varies depending on the specific recipe, but a silicon layer of 600 nm thickness is etched in around 80 s, resulting in an etch rate of 7.5 nm s^{-1} .

SF_6 (sccm)	C_4F_8 (sccm)	RF power (W)	DC bias (V)
18	45	38-41	127
16	38	27	127
14	38	27	127
13	39	31	130

TABLE 3.1: Different optimized etching parameters for anisotropic etching of amorphous silicon.

For the etching of silicon nitride a different gas mixture is used, namely oxygen

O₂ and trifluoromethane CHF₃. In the plasma, CHF₃ dissociates to generate fluorine radicals and CF_x species while the oxygen gas produces reactive oxygen radicals. The primary chemical etching pathway involves the reaction of fluorine with silicon atoms, as it is also the case for amorphous silicon. The resulting SiF₄ and N₂ molecules are removed by vacuum pumping from the chamber. Simultaneously, the surface is also bombarded with ions, which ensures directional etching. The CHF₃ also contributes to the formation of a fluorocarbon polymer that acts as a passivation layer on the sidewalls of the meta-atoms. The additional oxygen gas is necessary to regulate the polymer thickness by oxidizing excess fluorocarbon species into volatile by-products, thus preventing excessive surface passivation. The additional oxygen gas is not necessary during the etching of silicon because SF₆ and CF₄ form fewer polymer-forming CF_x species compared to the hydrogen-containing CHF₃ gas. Therefore, it is important to properly tune the gas ratio and etching parameters to allow for highly selective and smooth, well-defined structures. For the etching, the following optimized parameters were used, resulting in highly anisotropic pillars with smooth sidewalls: A flow of 1 sccm for O₂ and 16 sccm of CHF₃ was used. The pressure was kept constant at 7.5 mTorr, the ICP power was set to 2 W and the RF power was set to 9 W, resulting in a DC bias of 127 V. The etch rate of this recipe is 7.5 nm min⁻¹. Compared to the etch rate of the silicon layer, this etching recipe is significantly slower due to the low oxygen flow and ICP power. These conditions were chosen intentionally because metasurfaces using PMMA as the etch mask have been fabricated (Fig. 3.4d/e) in addition to metasurfaces that use Cr as the hard mask. However, this is only possible if a gentle and slow etching recipe is used.

After the etching, the Cr mask needs to be removed. The sample is immersed in a bath of Cr etch solution containing ceric ammonium nitrate and perchloric acid (Cr etch solution 1, Technic), which is diluted with water at a ratio of 1:3. The sample is etched for 60 s and then placed in a water bath at 90 °C for 5 min to remove the etching solution.

Figure 3.4 presents a selection of metasurfaces fabricated during the course of this thesis. Together, they illustrate the range of material platforms and design concepts explored throughout this work. While only the devices shown in panels a and b are investigated in detail in the following chapters, the remaining structures highlight additional research directions and collaborative projects

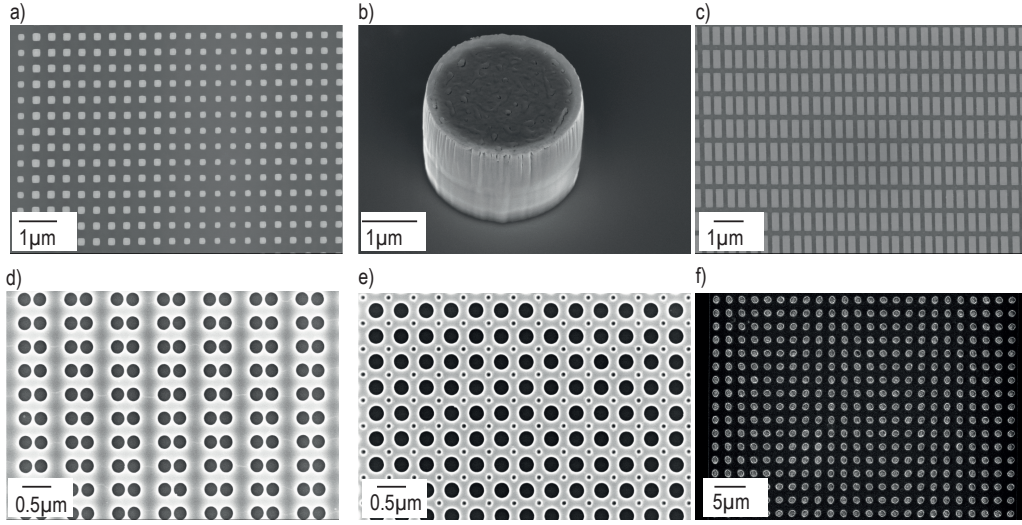


FIGURE 3.4: Examples of fabricated metasurfaces: The Metasurfaces shown in (a) and (b) are further discussed in this thesis. (a) Metasurface composed of nanoposts arranged in a rectangular lattice to enable multiport beam splitting. (b) A single silicon-silicon nitride nanoresonator is used for high-harmonic generation. (c) The silicon metasurface is fabricated on an ITO-coated substrate to enable intensity- and direction-dependent reflectivity. (d, e) Holes in a square and hexagonal lattice are etched in a silicon nitride film to enhance photoluminescence from emitters that are deposited into the holes. (f) holes etched in the SiN layer to enable high-harmonic generation and simultaneous phase-control.

that emerged during the thesis. The scanning electron micrograph in Fig. 3.4a shows a metasurface designed as a transmissive diffraction grating for beam splitting. The device consists of amorphous silicon nanoposts fabricated on a fused silica substrate, where rectangular meta-atoms with varying lateral dimensions are arranged into supercells of 22×1 elements. This spatial variation enables control of the local phase response and thus the diffraction behaviour. The experimental results obtained from this system are presented in Chapter 4.

The second image in Fig. 3.4b shows a hybrid silicon-silicon nitride pillar designed for direction-dependent high-harmonic generation. Compared to other metasurfaces, only a single nanoresonator is used to generate a nonlinear signal. Owing to the longer excitation wavelengths of $3 \mu\text{m}$ to $4 \mu\text{m}$, the resonator diameter is approximately $2 \mu\text{m}$ to support Mie modes. The results of the experiments are discussed in Chapter 5.

The metasurface shown in Fig. 3.4c is also based on amorphous silicon and is fabricated on an ITO-coated fused silica substrate. Its unit cell consists of two rectangular nanoposts and is designed to support a resonant mode with substantial field overlap in the underlying ITO layer. Near the epsilon-near-zero wavelength, the nonlinear refractive index of ITO leads to an

intensity-dependent shift in the refractive index, resulting in a corresponding modulation of the metasurface reflectivity. Preliminary measurements revealed pronounced intensity- and direction-dependent behaviour of the reflectivity. However, the laser appears to damage the sample permanently, as repeated measurements do not reproduce the same results. As a theoretical description is still under development, the experimental results are not discussed within this thesis.

Panels d-f demonstrate additional metasurfaces based on silicon nitride with holes etched inside the material. The samples shown in panels d and e were fabricated as part of a collaborative effort and were provided to Sergey Kruk at the University of Technology Sydney for experimental investigations of luminescence enhancement in emitters embedded within the nanoholes (manuscript in preparation).

Finally, panel f presents a metasurface that combines high-harmonic generation with wavefront control. The design employs the Pancharatnam-Berry phase, encoded through stepwise rotation of the unit cell, to impart spatial phase control to the generated harmonics. At the time of completion of this thesis, experimental characterization of this device has not yet been completed.

3.3 Spectroscopic Ellipsometry

This chapter describes the measurement of the optical properties of amorphous silicon and silicon nitride with ellipsometry. Although Si and SiN are often used materials in various industries and among various applications, depending on the deposition methods, the optical properties are different. Hence, it is not sufficient to assume values given in literature are also valid for our PECVD process parameters. Therefore, it is necessary to characterize the optical properties of both materials.

First, a short introduction to the concept of ellipsometry is given. Then the performed measurements and analysis of the data are described. Finally, the obtained results are presented and discussed.

Ellipsometry is a non-destructive, highly sensitive optical characterization technique that measures the interaction between light and the material. More specifically, the change in polarization upon interaction with the sample is measured. This change depends on the film thickness and the optical properties of the material. The polarization change upon reflection is described by the amplitude ratio Ψ and phase difference Δ . Although ellipsometry is mostly used to determine the refractive index n and the extinction coefficient k , it is also useful to characterize surface and interface roughness, composition, crystallinity, anisotropy, thickness and uniformity [106]. Ellipsometry measures the relative change of the p- and s-polarized light components caused by reflection or transmission at the sample surface. A known polarization is incident on the sample, and the output polarization is measured. The change in polarization is written as: $\rho = \frac{r_p}{r_s} = \tan(\Psi)e^{i\Delta}$. The amplitude ratio Ψ describes how strong p-polarized light is reflected compared to s-polarized light, while Δ describes the relation of phase change between s- and p-polarized light [106].

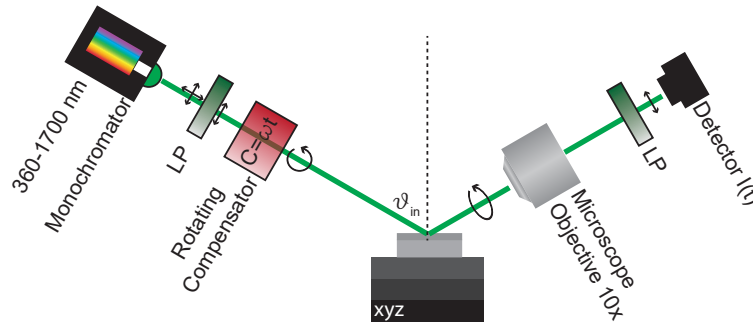


FIGURE 3.5: Schematic representation of Imaging Ellipsometry.

Figure 3.5 shows a schematic of the ellipsometer (Accurion EP-4), which is built as a rotating compensator ellipsometer (RCE). Other possible options are rotating analyzer (RAE) and rotating polarizer ellipsometry (RPE). The advantage of RCE in comparison to RAE and RPE is that it allows for determining all four Stokes parameters [106]. The ellipsometer is designed in a PC_rSA geometry, meaning the order of optical elements is polarizer (P), rotating compensator (C_r), sample (S) and analyzer (A). In principle, the positions of the sample and the compensator can also be interchanged [107]. The white light source generates unpolarized light in a wavelength range of 360 nm to 1700 nm, which is spectrally filtered with a monochromator. A specific linear polarization state is set with a polarizer. Typically, an orientation angle of 45° is used to ensure equal contributions of p- and s-polarized light. The light passes through an optical retarder, which is also named a rotating compensator and is a quarter-wave plate. It imprints a controlled phase shift δ between the two polarization components. As the compensator continuously rotates at a constant angular velocity ω , the phase retardation becomes time-dependent, resulting in a periodic time modulation of the polarization state of light which evolves from linear to elliptical to circular and back. Upon interaction with the sample surface, the polarization components are modulated and experience different amplitude attenuations and phase shifts that are described by the Fresnel equations. This interaction encodes the material-specific information into the ellipsometric angles Ψ and Δ . Finally, the analyzer (a second polarizer) projects the elliptical polarization state onto a fixed reference frame, thereby transforming the polarization modulation into a measurable intensity signal. Mathematically, the transformations are described by a Müller matrix formalism, which can also account for partially polarized and depolarizing light, in contrast to the Jones formalism. The polarization state is presented by the Stokes vector

$$\vec{S} = \begin{bmatrix} S_0 \\ S_1 \\ S_2 \\ S_3 \end{bmatrix}, \quad (3.1)$$

where S_0 denotes the total intensity and S_1 , S_2 , and S_3 describe the linear and circular polarization components. For the PC_rSA configuration, the detected intensity is obtained from the multiplication of the Müller matrices of every optical element $I(t) = AM_S M_C(\delta, \omega(t))PS_{in}$, where P and A describe the polarizer and analyzer matrices which correspond to the ones of a linear polarizer, M_S the sample matrix and M_C the rotating compensator matrix.

The sample matrix has the form [108]

$$\begin{aligned}
 M_S &= \begin{bmatrix} 1 & -N & 0 & 0 \\ -N & 1 & 0 & 0 \\ 0 & 0 & C & S \\ 0 & 0 & -S & C \end{bmatrix} \\
 &= \begin{bmatrix} 1 & -\cos(2\Psi) & 0 & 0 \\ -\cos(2\Psi) & 1 & 0 & 0 \\ 0 & 0 & \sin(2\Psi) \cos(\Delta) & \sin(2\Psi) \sin(\Delta) \\ 0 & 0 & -\sin(2\Psi) \sin(\Delta) & \sin(2\Psi) \cos(\Delta) \end{bmatrix}, \quad (3.2)
 \end{aligned}$$

with

$$N = \frac{|r_p|^2 - |r_s|^2}{|r_p|^2 + |r_s|^2}, C = \frac{2|r_p||r_s|\cos(\phi_p - \phi_s)}{|r_p|^2 + |r_s|^2}, S = \frac{2|r_p||r_s|\sin(\phi_p - \phi_s)}{|r_p|^2 + |r_s|^2}, \quad (3.3)$$

which is derived from the corresponding Jones matrix. Inserting the ellipsometric identities $\tan \Psi = r_p/r_s$ and $\Delta = \phi_p - \phi_s$ this can be rewritten as $N = \cos(2\Psi)$, $C = \sin(2\Psi) \cos(\Delta)$, and $S = \sin(2\Psi) \sin(\Delta)$. Carrying out the matrix multiplication of all optical elements gives the time-harmonic measured detector signal. According to Fourier theory, every periodic function can be expanded into a Fourier series; thus, the time-harmonic intensity can be expressed as [109]:

$$\begin{aligned}
 I(t) &= I_0 + C_2 \cos(2\omega t) + S_2 \sin(2\omega t) + C_4 \cos(4\omega t) + S_4 \sin(4\omega t) \quad (3.4) \\
 &= I_0 \left[1 + \frac{C_2}{I_0} \cos(2\omega t) + S \sin(2\omega t) + N \cos(4\omega t) + C \sin(4\omega t) \right],
 \end{aligned}$$

where C_i and S_i are the experimentally determined Fourier coefficients, which are also called ellipsometric transfer quantities (ETQ). These coefficients are directly related to the Müller matrix elements of the sample N, C and S and contain polarization information of the reflected light. For isotropic non-depolarizing samples, the Fourier coefficients are directly related to the ellipsometric angles Ψ and Δ via [109]

$$C_4 = \cos(2\Psi), S_4 = \sin(2\Psi) \cos(\Delta), S_2 = \sin(2\Psi) \sin(\Delta). \quad (3.5)$$

Ideally, the coefficient C_2 vanishes for an ideal quarter-wave plate, an isotropic non-depolarizing sample and ideal alignment of analyzer and polarizer. The decomposition of the intensity signal into the Fourier series provides therefore

$\Psi(\lambda)$ and $\Delta(\lambda)$ from which the ratio of r_p and r_s can be calculated. Now a model for the material system is built that depends on the values of the refractive index n , extinction coefficient k and layer thickness d for every material of the layer stack and the expected theoretical values Δ_{calc} and Ψ_{calc} are calculated and compared to the measured values. Technically, the dielectric function is used for modeling, from which the refractive index and extinction coefficient can be calculated. This is done because the dielectric function can be deduced from physical properties of the material, such as free carriers in metals.

The quality of the extracted optical properties can be enhanced if several considerations are taken into account during the measurement. First, it is beneficial to cover a larger spectral range during the measurement, as this tends to make it easier to evaluate the quality of the fit. Second, the difference in reflection of s- and p-polarized light is largest at the Brewster angle $\tan(\theta_B) = n_2/n_1$. Hence, it is beneficial to use an angle of incidence (AOI) close to the Brewster angle for the specific material combinations. For a-Si, the Brewster angle is $\theta_B \approx 74^\circ$ for $n_2 = 3.7$ and $n_1 = 1$. SiN, on the other hand, has a smaller refractive index $n_2 \approx 2$, resulting in a smaller Brewster angle of $\theta_B \approx 63^\circ$. Lastly, measurements at more than one AOI also help to improve the fit quality. After the general method of ellipsometry is explained, the performed measurements of Si and SiN are described.

A Si layer was deposited on a fused silica substrate with a target thickness of 600 nm. The film thickness was determined using a profilometer, and the measured area is marked on the sample surface. Spectroscopic ellipsometry measurements were performed in a spectral range of 500 nm to 1700 nm in 5 nm steps for different angles of incidence (AOIs) of 47° , 52° , 57° , and 62° . The ellipsometer is equipped with a microscope objective allowing direct imaging of the sample surface. The sample was positioned so that the previously marked area could be measured. Six regions of interest (ROIs) were selected, each a few square micrometres in size, where the ellipsometric angles are measured separately to independently verify the optical properties at different locations. A second option is to map the entire area that is illuminated, but then the average of the optical properties over the whole region is analyzed.

Silicon nitride and fused silica are both transparent throughout the visible and near-infrared spectral range. Consequently, ellipsometric modelling of SiN on fused silica is complicated by multiple internal reflections and backside reflections from the transparent substrate. To mitigate these effects,

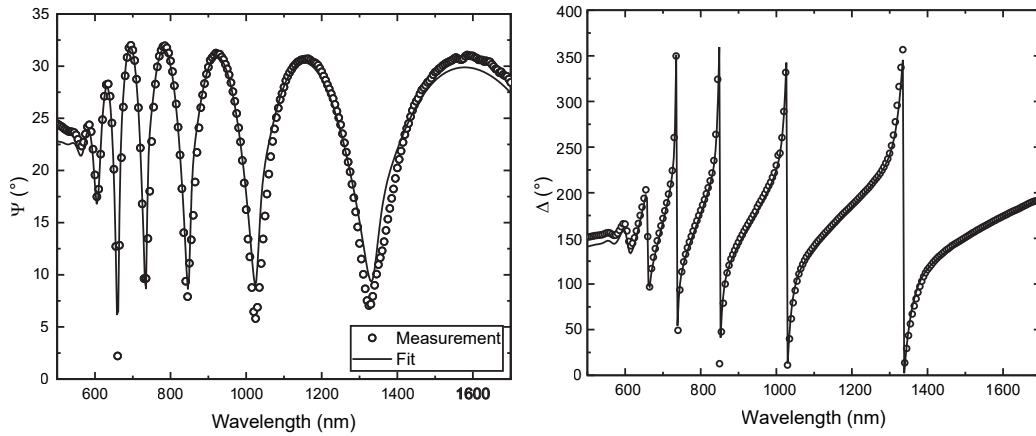


FIGURE 3.6: Wavelength-dependent ellipsometric angles Ψ and Δ for a 600 nm thick Si layer on a glass substrate. The dots indicate the measured values and the solid line shows the calculated data points when the silicon layer is modelled with a Cody-Lorentz model.

approximately 1000 nm of SiN is deposited onto a crystalline Si wafer, whose absorbing substrate suppresses backside reflections and provides a more robust optical model. The ellipsometric measurement is performed in a spectral range of 500 nm to 1700 nm in 10 nm steps for different AOIs of 63° , 65° , and 67° .

Figure 3.6 presents the measured ellipsometric quantities Ψ and Δ of the a-Si film on fused silica. The measured values are shown as discrete points (dots), while the solid black line represents the fitted model, which is discussed in detail later. The raw features of the spectroscopic ellipsometry spectra contain valuable information about the film structure and optical properties. Understanding these features aids in constructing an appropriate optical model. The measured spectra indicate two regions: From 500 nm to 600 nm, both Ψ and Δ have a constant decrease/increase, while above 600 nm, both curves show oscillations indicating a transparent film. These oscillations arise from optical interference within a thin film, and their number and spectral position depend on the refractive index and film thickness. Thicker films or materials with a higher refractive index produce a greater number of interference features. The amplitude of the Ψ oscillations is influenced by the optical contrast between the film and the substrate. A larger refractive index difference leads to stronger peak-to-valley variations in Ψ .

As the wavelength decreases (< 700 nm), the a-Si film begins to absorb light, indicated by damping in the oscillations until they completely vanish below 600 nm, because only the front surface reflection contributes to the detected signal. The transition to absorption for smaller wavelengths occurs when

the photon energy exceeds the material's bandgap. In a-Si, this transition is gradual due to weak absorption near the bandgap. In contrast, direct bandgap materials exhibit a much sharper transition between absorbing and transparent regions.

To extract meaningful physical properties from the measured spectra, the optical constants of each layer in the model must be accurately described. Since the refractive index n and extinction coefficient k vary with wavelength, the optical model must account for their spectral dependence across the entire measured range. The most direct approach is to introduce optical constants as tabulated values at each wavelength, obtained from the literature. While this method is effective for stable and well-characterized materials such as crystalline Si wafers, it becomes impractical for thin films whose optical properties depend strongly on microstructure, composition, and deposition conditions. Therefore, suitable optical dispersion models are required to have physically meaningful descriptions of the wavelength-dependent optical constants. In such cases, the tabulated values can serve as initial estimates and must be refined through fitting. A more efficient and physically meaningful approach is to describe optical constants using mathematical functions known as dispersion relations, which are based on physical theory to describe the wavelength-dependent behaviour of optical constants. However, carefully selecting the appropriate model is critical as even the wrong model can produce good fit results indicated by a low mean-squared error (MSE) but physically meaningless parameters.

The following section introduces common dispersion models and discusses their suitability for different material systems. Over the years, a variety of models has been developed. The most simple model is the Cauchy equation $n(\lambda) = A + B/\lambda^2 + C/\lambda^4 + \dots$ that was defined in 1830 by Augustin-Louis Cauchy [110]. A , B and C are the fit parameters that control the shape of the curve. B and C control the curvature of n towards smaller wavelengths, and A gives the limit of the refractive index at large wavelengths. It is an empirical model which can be used for transparent materials ($k(\lambda) = 0$) exclusively. In absorbing materials, it can be used in the spectral region where $k(\lambda) = 0$ holds true to determine the thickness of the layer.

For the total description of absorbing materials, oscillator models are used. One example is the Lorentz oscillator that is based on classical dispersion theory, where bound electrons are treated as damped harmonic oscillators responding to electric fields. Each oscillator resonance corresponds to a

specific electronic or vibrational transition. The dielectric function is given by

$$\tilde{\epsilon} = \epsilon_{1,\text{offset}} + \frac{AE_c}{E_c^2 - E^2 - i\Gamma E}, \quad (3.6)$$

where A describes the amplitude of the oscillator, Γ the broadening, E_c the central energy, and $\epsilon_{1,\text{offset}}$ the high-frequency dielectric constant [111]. The Lorentz model describes wide bandgap semiconductors (e.g. GaN) or transition metal oxides (e.g. TiO₂) very well, but is less effective for amorphous semiconductors [112].

For amorphous semiconductors like a-Si, the absorption coefficient exhibits a characteristic energy dependence that follows the Tauc relation $\omega^2\epsilon_2 \propto (\hbar\omega - E_g)^2$ [113]. By incorporating this behaviour into $\epsilon_2(E)$ and combining it with a Lorentz-type oscillator, the Tauc-Lorentz model describes both the smooth absorption onset and the resonant interband transitions of such materials. It is mathematically described by

$$\epsilon_2(E) = \begin{cases} 0, & E \leq E_g \\ \frac{A(E - E_g)^2}{E[(E^2 - E_0^2)^2 + \Gamma^2 E^2]}, & E > E_g \end{cases} \quad (3.7)$$

Compared to the Lorentz-oscillator, the bandgap E_g of the material is an additional fit parameter. In this dispersion model, the imaginary part of the dielectric function $\epsilon_2(E)$ is parametrized while the real part $\epsilon_1(E)$ is obtained via Kramers-Kronig integration. The Cody-Lorentz model was developed by Ferlauto et al. and modifies the Tauc-Lorentz approach by changing the functional form of the absorption onset. It is describes by [112]:

$$\epsilon_2(E) = \begin{cases} \frac{E_t G(E_t) L(E_t)}{E} \exp\left(-\frac{E - E_t}{E_u}\right) & 0 \leq E \leq E_t \\ G(E) L(E) = \frac{(E - E_g)^2}{(E - E_g)^2 + E_p^2} \frac{AE_0 \Gamma E}{(E^2 - E_0^2)^2 + \Gamma^2 E^2}, & E > E_t \end{cases} \quad (3.8)$$

For a photon energy greater than E_t , the dielectric function is given as the product of the variable band edge function $G(E)$ and the Lorentz oscillator $L(E)$. For a photon energy below E_t , the dielectric function is described by an exponential decay called the Urbach tail. The energy E_u determines the rate of the decay of the Urbach tail.

Besides the deposited Si and SiN layers, the substrate also needs to be modelled. For the crystalline silicon substrate and its native oxide layer,

literature values available in the software were used. The exact fused silica substrate was measured and modelled with a Cauchy model by Maximilian Albert. The measured optical response is also influenced by surface and interface roughness, which is represented by an effective layer using the Bruggemann effective medium approximation (EMA). This approach models the roughness as a mixture of two materials with an effective dielectric function that approximates the averaged optical behaviour. In this work, only the surface roughness was taken into account.

Next, the employed fit strategy is described in more detail for each material system, and the results are presented. The mean square error (MSE) is used to evaluate the quality of the fit as it describes the similarity between measured and calculated data [114]:

$$MSE = \sqrt{\frac{1}{2N - M} \sum \left[\left(\frac{\psi_i^{Mod} - \psi_i^{Exp}}{\sigma_{\psi,i}^{Exp}} \right)^2 + \left(\frac{\Delta_i^{Mod} - \Delta_i^{Exp}}{\sigma_{\Delta,i}^{Exp}} \right)^2 \right]} \quad (3.9)$$

M is the number of variables in the model, and σ gives the standard deviation of each experimental data point. N gives the number of ψ and Δ pairs. A lower MSE indicates a better match between model and experimental data, meaning a "better" fit is performed that indicates a better match of calculated optical constants to the actual optical constants. A second consideration is that although the MSE is a good quantity to judge the fit quality, one has to keep in mind that the fitting algorithm changes the fitting parameters based on the initial estimates of the fitting parameters. If they are poorly chosen, the fit can converge towards a local minimum of the MSE and not the global minimum, resulting in useless results. This is particularly bad if the wrong film thickness is assumed, because then the rest of the modelling will not give correct optical properties. Thus, it is important to cross-check the thickness of the thin film with different means, e.g. a profilometer. To mitigate the issue of being trapped in a local minimum, fit parameters can be set as either local or global. If global is chosen, the fitting procedure is repeated by the algorithm for many starting parameters, but this drastically increases the computation time.

In the following section, the analysis of the ellipsometry measurements and the corresponding results are presented. Both materials exhibit absorption within parts of the measured spectrum, which increases the complexity of the

optical modelling. Therefore, a step-by-step fitting procedure was employed to obtain meaningful optical constants.

First, the transparent spectral region is fitted with the Cauchy dispersion equation in order to determine the film thickness. The thickness values obtained from the ellipsometric fits were compared with profilometer measurements to verify the consistency of the model and to prevent unphysical optical constants during the subsequent analysis. Once a reliable thickness was obtained, it was fixed for the remaining analysis.

Second, the optical constants were determined by a point-by-point fit. In contrast to parametric dispersion models, this approach independently varies the refractive index n and the extinction coefficient k at every wavelength separately until the smallest MSE is found. The resulting optical constants may be an adequate result, but they do not necessarily need to be physically correct. Hence, the previously presented dispersion equations are used for fitting to ensure a Kramers-Kronig consistent result over the whole spectral range.

The fused silica substrate is modelled using a Cauchy dispersion model with parameters $A=1.4316$ and $B = 15710 \text{ nm}^2$ as analyzed by Maximilian Albert. For the amorphous silicon layer, fitting with the Cauchy model in the spectral range from 800 nm to 1700 nm yielded a thickness of 590 nm and an MSE value of 31. The comparatively large MSE indicates that the dispersion is not adequately captured by the transparent Cauchy equation. Consequently, more sophisticated oscillator-based models are considered. The complexity of the dispersion model was increased stepwise in order to achieve an accurate description without unnecessarily over-parametrising the fit. Initially, a single Lorentz oscillator model was applied, followed by a Tauc-Lorentz model. However, both approaches still fail to satisfactorily describe the optical behaviour in the low-energy region. Therefore, the model was extended to a Cody-Lorentz equation, which was originally specifically developed for the description of amorphous hydrogenated silicon [112].

Application of the Cody-Lorentz model results in a good agreement with the experimental data and reduces the MSE to 11.8. The obtained fit parameters are summarized in Table 3.7 together with the extracted refractive index and extinction coefficient. Over the spectral range from 500 nm to 1700 nm, the refractive index decreases from 5.0 to 3.4. The extinction coefficient remains small in the near-infrared region and increases from 800 nm towards shorter wavelengths due to the onset of strong interband absorption. The extracted optical bandgap of $E_g=1.5$ agrees well with reported values for hydrogenated

amorphous silicon deposited with PECVD [85].

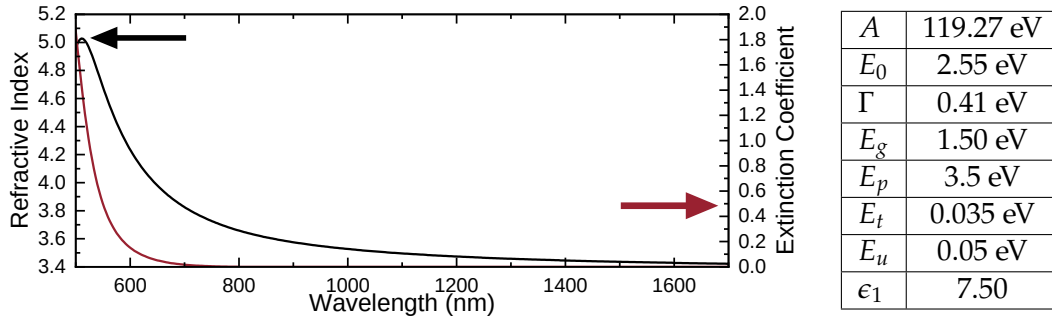


FIGURE 3.7: Refractive index and extinction coefficient of a 600 nm thick amorphous silicon layer grown by plasma-enhanced chemical vapour deposition at 300 °C. Overview of Cody-Lorentz oscillator fit parameters.

The same fitting strategy is also applied to the silicon nitride layer. Silicon nitride deposited by PECVD is commonly hydrogen-containing and can exhibit significant deviations from stoichiometric Si_3N_4 depending on the deposition conditions and precursor gas ratios. In particular, silicon-rich silicon nitride may exhibit absorption within the visible spectral range. Initially, the layer is described by a Cauchy model resulting in an MSE of 6.8 and a film thickness of 1186 nm. The model adequately reproduces the experimental data at longer wavelengths, but failed to describe it at wavelengths below 500 nm. This behaviour indicates the onset of absorption as expected for silicon-rich films. To account for absorption, the model was extended to the Tauc-Lorentz oscillator. This significantly improved the quality of the description, indicated by a lower MSE of 1.6. The resulting fit parameters as well as the dispersion of the refractive index are displayed in Figure 3.8. The refractive index values lie in the range of 1.96 to 2.06 and are consistent with reported values from the literature.

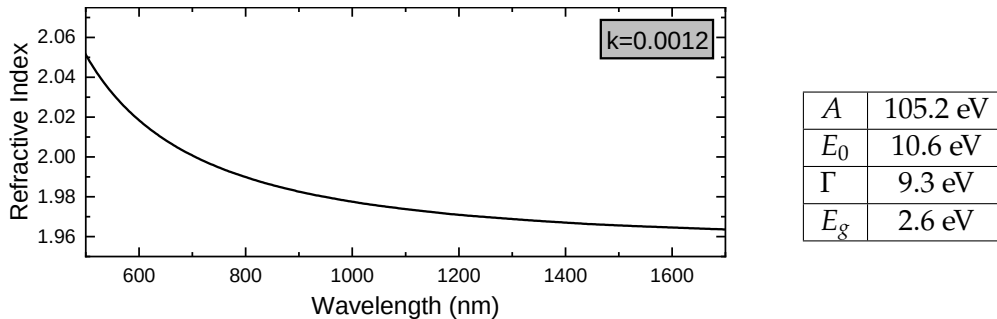


FIGURE 3.8: Refractive index and Extinction coefficient of a 1180 nm thick amorphous silicon nitride layer grown with plasma-enhanced chemical vapour deposition at 300 °C. Overview of Tauc-Lorentz oscillator fit parameters.

Chapter 4

Metasurface-Enabled Multiport Interferometry

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Quantum information processing is based on the generation, manipulation, and detection of quantum states to encode, process and transmit information. Photons are particularly attractive carriers of quantum information due to their low decoherence rates, high propagation speed, and compatibility with existing optical communication infrastructure [115].

A key conceptual breakthrough was provided by Knill, Laflamme, and Milburn in 2001, who showed that efficient quantum computation is possible using only linear optical elements, single-photon sources, and projective measurements [116]. Prior to that work, it was believed that nonlinear interactions were necessary for photonic quantum computing. Their results established linear optics as a viable and scalable framework for the implementation of quantum algorithms. Examples of these linear optical elements include beam splitters and phase shifters, which play a central role in building optical networks. In 1994, it was shown that any arbitrary unitary transformation ($N \times N$) can be decomposed into a sequence of two-mode (2×2) operations. This decomposition implies that, in principle, quantum operations can be realized

by cascading beam splitters and phase shifters. One example of a free-space implementation was demonstrated in [117]. However, scaling free-space networks to a large number of optical modes presents significant challenges as conventional bulk optical elements suffer from limited scalability and miniaturization capabilities. Conventional integrated photonic platforms based on waveguides and directional couplers offer a promising route to overcome the limitations of miniaturization as they enable the realization of MZIs with a more compact footprint [118, 119]. Metasurfaces complement conventional integrated photonics and even extend its functionalities by enabling compact, planar implementations of complex transformations in free-space. Unlike conventional beam splitters and directional couplers, metasurfaces can be engineered to realize multiport beam splitting by distributing light into $N > 3$ spatial modes within a single planar device.

Beyond quantum applications, multiport beam splitters are also of interest in classical optics. They enable increased functionalities such as multiport outputs [120] or parallel signal processing [121].

In this work, we investigate a metasurface-based approach to achieve multiport functionality. Specifically, we demonstrate a free-space multiport metasurface-based interferometer. The interferometer consists of two nominally identical silicon-based metasurfaces, where each metasurface acts as a multiport beam splitter. By controlling the relative phases between the different light paths, we achieve deterministic control of the output intensities, effectively realizing a reconfigurable multiport interferometer. We begin by characterizing the individual metasurfaces using classical light to study their diffraction efficiencies and output distributions. Finally, we extend our study to the quantum regime by replacing the classical source with single-photon emission from a semiconductor quantum dot. We find that the metasurface architecture preserves the quantum statistics and enables both second- and third-order photon correlation measurements within a single device. Together, these results mark an important step toward scalable metasurface-based interferometry for quantum and classical photonic applications as our system in its cascaded configuration emulates a typical interferometer with enhanced functionality and scalability.

This chapter is organized as follows. It begins with a review of relevant literature, highlighting existing approaches to scaling the number of optical modes in photonic networks. Subsequently, the theoretical framework required to analyze and interpret the experimental results is introduced with particular emphasis on key concepts in quantum optics. The chapter then presents

the design and functionality of the metasurface-based system. Finally, the experimental results are discussed, starting with the classical characterization of the device and ending with its operation in the single-photon regime.

This work has been published in ACS Photonics [69]. The work was done in cooperation with the hybrid quantum photonic devices (hqpd) group. The quantum optical experiments were performed with the assistance from Nicolás Claro-Rodríguez and Patricia Kallert. The quantum dot embedded inside a circular Bragg cavity was provided by groups of Tobias Huber-Loyola, Sven Höfling, Armando Rastelli, and Rinaldo Trotta. The simulation of the initial metasurface design was performed by Dr. Ruizhe Zhao in the group of Lingling Huang at Beijing Institute of Technology.

4.1 Metasurface-Based Multiport Optical Transformations

An optical (quantum) network consists of N input and N output modes whose transformations are described by unitary matrices. A foundational result by Reck et al. demonstrated that any $N \times N$ unitary transformation can be decomposed into a sequence of 2×2 unitary operations realized by beam splitters and phase shifters. This decomposition requires $N \cdot (N - 1)/2$ two-mode interferometric elements, yielding a hardware complexity that scales quadratically with the number of optical modes [122]. They established that universal linear optics can, in theory, be constructed from elementary interferometric building blocks. Clements et al. in 2016 showed that by arranging BS and phase shifters in a symmetric rectangular instead of a triangular architecture improves efficiency and reduces the number of required components [123]. Building on this concept, Arends et al. in 2024 provided an algorithm to decompose $N \times N$ unitaries into smaller $m \times m$ with ($m > 2$) subunitaries, laying the groundwork for exploring devices that simultaneously operate on a larger set of modes.

Different platforms and strategies have been explored to achieve multipath and flat-type beam splitters. The most established physical platform for implementing large-scale optical unitaries is integrated photonics. Here, unitary transformations are realized using arrays of Mach-Zehnder interferometers (MZI), consisting of an ensemble of waveguides, directional couplers or multimode interference (MMI) couplers and phase shifters. While integrated

photonics provides a compact alternative to bulk optics, universal implementations of arbitrary unitaries are still typically based on decompositions into 2×2 matrices, as formalized by Reck and Clements. It is important to note that these building blocks however, are not strictly limited to 2×2 building blocks. Higher-dimensional couplers could reduce circuit length in specific architectures, but current scalable photon processors still predominantly rely on cascaded two mode interferometric building blocks [124, 125]. This becomes particularly relevant when comparing integrated photonic architectures to planar platforms such as metasurfaces, which aim to implement higher-order operations in a single planar device rather than through cascaded operations. To overcome these limitations, alternative flat optical platforms such as metasurfaces have been proposed. Compared to directional couplers, they are not restricted to a certain number of optical modes and hence offer a different scaling paradigm compared to waveguide-based approaches. Metasurface-based beam splitters have been realized using both dielectric and plasmonic materials, with and without polarization selectivity, as well as different numbers and spatial configurations of outputs have been shown, highlighting the unprecedented design freedom of metasurfaces [126–130]. Tian et al. demonstrated a metasurface supporting between two and seven output channels with different power distribution. Metasurface-based BS do not necessarily need to operate in transmission as presented by Huang et al. [127]. Their metasurface consists of TiO_2 nanodisks on fused silica placed on a silver mirror and generated four or five diffraction orders in reflection. There are not only experimental realizations but also many theoretical proposals on how flat-type, highly efficient beam splitting can be achieved. These proposals include other material systems than silicon and various numbers of spatial output distributions (1D vs. 2D) as well as numbers ranging from two to even nine outputs [128, 129, 131]. However, most existing implementations correspond to effectively $1 \times N$ devices, where a single input mode is mapped onto multiple outputs functioning as a power splitter and not necessarily as a typical beam splitter [126, 127, 129, 132]. This restricts the direct applicability to full unitary transformations involving multiple coherent input modes. More recent theoretical and experimental work has extended metasurface functionalities into the quantum regime. Metasurfaces are used to generate quantum light or to manipulate quantum states. An example of the former task was demonstrated by Santiago-Cruz et al. [133], who used a gallium arsenide metasurface that supports a quasi-bound state in the continuum resonance to generate two entangled photons from one pump photon by a spontaneous

parametric down-conversion process. Alternatively, a quantum emitter can also be implemented into the metasurface whose Purcell enhancement is used to improve the emission [134, 135]. Metasurfaces are exploited to entangle qubits as first theoretically proposed [136], and later also demonstrated experimentally by different groups [46, 47, 137, 138]. Other demonstrations of metasurfaces in the quantum realm include quantum imaging, coherent perfect absorption, or quantum sensing [139].

Beam splitting in the quantum regime has also been explored, however mostly in theory and experimental realizations are lacking.

One example of such theoretical work was presented by Liu et al. [120], who introduced the concept of parallel beam splitting using a gradient metasurface based on the Pancharatnam–Berry phase. Their design employs a spatial rotation of meta-atoms to generate a linear phase gradient, leading to polarization-dependent diffraction. The PB-phase relies on circular polarization states. If left circularly polarized light is incident onto the metasurface a non-diffracted beam of the same polarization and a diffracted beam (Path +1) of right circular polarized light are produced. If the incident polarization is reversed diffraction to Path -1 occurs. If linear polarization is used, three diffracted beams occur where each one has a different polarization. A key feature of this approach is that different input beams incident simultaneously can interfere in shared output ports, enabling multi-input multi-output interference in a single device. However, this functionality is inherently tied to polarization degree of freedom, leading to a strong coupling between spatial and polarization modes. This represents a fundamental limitation compared to conventional beam splitters, where spatial modes can often be treated independently of polarization. Moreover, the reliance on circular polarization states introduces additional experimental constraints and reduces compatibility with more general input states.

Building on this concept, Liu et al. [140] proposed that the same metasurface platform can be used to implement a two-qubit quantum gate, specifically a controlled-Z (CZ, also called *CNOT* gate) operation. By exploiting the parallel beam splitting mechanism and carefully selecting input paths and polarization states, they showed that a single metasurface can realize a functionality that would conventionally require three bulk cube beam splitters. This concept was experimentally realized by Xun et al. [141] who demonstrated a quantum gate, confirming that metasurfaces can implement non-trivial quantum operations beyond simply passive beam splitting.

Beyond specific gate implementations, a more general theoretical framework was introduced by Yousef et al. [142], who described metasurfaces in terms of quantum graphs. In this picture, diffraction orders correspond to nodes, while correlations between photons define edges, enabling a unified description of multiport quantum interference processes. They present a theoretical framework that allows the implementation of any quantum graph (= quantum network) within a single metasurface. They construct a two-output and four-output metasurfaces. They study Hong-Ou-Mandel interference and demonstrate that different output configurations either result in bunching or antibunching. However, they only use a single input mode.

Taken together, these works illustrate a shift from merely light routing to implementing (unitary) transformations with metasurfaces. However, while metasurfaces have been shown to implement single-input transformations with classical light and specific quantum gates, the behaviour of multi-input high-dimensional interference in metasurface-based multiport systems remains insufficiently explored.

4.2 Physical Principles

The designed metasurface acts as a diffraction grating and can be considered an extension of a standard beam splitter with spatial non-orthogonal input/output modes. The cascade of two such metasurfaces in combination with phase shifters mimics a higher-dimensional Mach-Zehnder interferometer. In the following section, first, the mathematical framework for a standard beam splitter and interferometer are presented. Their behaviour with classical and non-classical light states is discussed to compare it in subsequent chapters with the behaviour of the metasurface. Additionally, the generation of single photons is briefly discussed.

4.2.1 Beam Splitter as Building Blocks for Optical Networks

Beam splitters are important optical elements for both classical and quantum optics. A beam splitter has two input ports 1 and 2, and in the classical picture, the input power is redistributed into output modes 3 and 4. The incident light is split at the internal interface of the cube BS into a transmitted and reflected field described by the transmission and reflection coefficients t and r . The following mathematical description is derived from [143]. The beam splitting

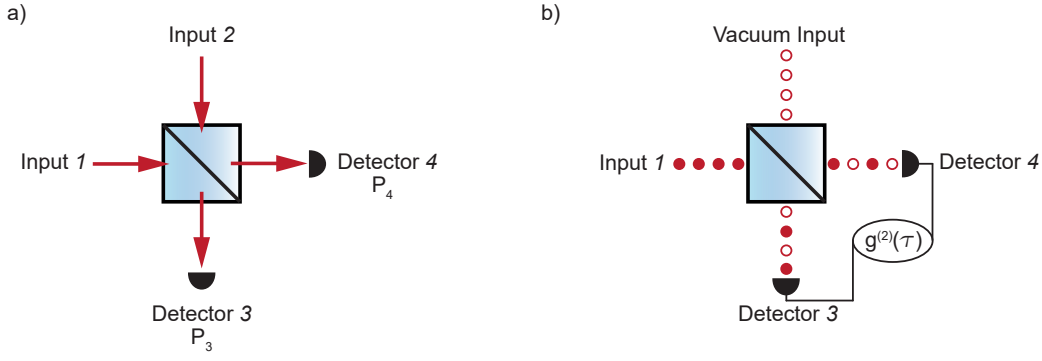


FIGURE 4.1: Beam splitter with (a) classical and (b) quantum input states.

operation is described as a linear transformation by a matrix:

$$\begin{pmatrix} E_3^{\text{out}} \\ E_4^{\text{out}} \end{pmatrix} = \begin{pmatrix} r & t \\ t & -r \end{pmatrix} \begin{pmatrix} E_1^{\text{in}} \\ E_2^{\text{in}} \end{pmatrix} \quad (4.1)$$

In the quantum description, the classical fields are replaced by the corresponding quantum operators so that Eq. 4.1 is transformed into:

$$\begin{pmatrix} \hat{a}_3^{\text{out}} \\ \hat{a}_4^{\text{out}} \end{pmatrix} = \begin{pmatrix} r & t \\ t & -r \end{pmatrix} \begin{pmatrix} \hat{a}_1^{\text{in}} \\ \hat{a}_2^{\text{in}} \end{pmatrix} \quad (4.2)$$

In the quantum picture the creation of an optical mode is described via the creation operator, while the destruction of a mode is described via the annihilation operator. If a single photon is sent into mode 1 the input state is written as $|\psi\rangle = |1\rangle_1|0\rangle_2$. The probability to detect the photon in mode 3 is calculated via $P_3 = {}_2\langle 0|_1\langle 1|\hat{N}_3|1\rangle_1|0\rangle_2 = {}_2\langle 0|_1\langle 1|(r^*\hat{a}_1^\dagger + t^*\hat{a}_2^\dagger)(r\hat{a}_1 + t\hat{a}_2)|1\rangle_1|0\rangle_2 = r^*r = R$, where R is the probability of a single photon from port 1 being detected at D_3 and R is also the reflectance of the beam splitter. The probability to detect the single photon at detector D_4 is given by $P_4 = T$. If a 50:50 BS is used, the probability to detect a photon in either output is 50% so the result is similar to the classical case. Although the beam splitter transforms the two optical modes into a coherent superposition of output modes in the quantum description, measurements of single output intensities alone do not reveal this difference. The non-classical character becomes apparent only through measurements involving correlations between the output modes, such as coincidence detection. A first difference is observed when we consider the probability P_{34} which describes simultaneous detection at output 3 and 4. Classically, this is possible as a fraction of the light T is transmitted to detector 4, while a fraction $R = 1 - T$ is reflected to detector 3, resulting in

the probability for double detection:

$$P_{34}^{\text{classical}} = \frac{R(E_1)^2 T(E_1)^2}{(E_1)^2 (E_1)^2} = RT. \quad (4.3)$$

The result in the quantum picture looks different. The simultaneous measurement is described by the operator $\hat{a}_4 \hat{a}_3$ resulting in the probability of $P_{34}^{\text{quantum}} = {}_2 \langle 0 | {}_1 \langle 1 | (\hat{a}_4 \hat{a}_3)^\dagger (\hat{a}_4 \hat{a}_3) | 1 \rangle_1 | 0 \rangle_2 = 0$ which states that quantum-mechanically double detection of a single photon is not possible which is different from the classical result.

This joint detection probability is also described in the framework of correlation functions developed by Roy J. Glauber that can both be defined for classical and quantum light states [3]. The n -th order correlation function in the quantum picture describing the n -fold photon correlation is defined as:

$$g^{(n)}(\tau) = \frac{\langle \hat{I}(t) \hat{I}(t + \tau_1) \cdots \hat{I}(t + \tau_{n-1}) \rangle}{\langle \hat{I}(t) \rangle^n}, \quad (4.4)$$

The second-order intensity correlation function for zero time delay quantifying the suppression of multiphoton states can be rewritten as:

$$g^{(2)}(0) = \frac{\langle \hat{a}^\dagger \hat{a}^\dagger \hat{a} \hat{a} \rangle}{\langle \hat{a}^\dagger \hat{a} \rangle^2} = \frac{\langle n^2 \rangle - \langle n \rangle}{\langle n \rangle^2} = 1 + \frac{(\Delta n)^2 - \langle n \rangle}{\langle n \rangle^2}. \quad (4.5)$$

Experimentally the value is extracted using the Hanbury Brown and Twiss (HBT) setup that measures coincidences at two single-photon detectors. This concept of coincidence measurements was proposed by Hanbury Brown and Twiss already in 1954 originally for radio astronomy but has been extended to optical signals [144]. The setup consists of a beam splitter and two single-photon detectors to define the photon statistics. For classical light, $g^{(2)}(0) \geq 1$, whereas non-classical light can exhibit anti-bunching, characterized by $g^{(2)}(0) < 1$. In the idealized case of a perfect single-photon Fock state $|1\rangle$, the probability of detecting two photons simultaneously vanishes, resulting in $g^{(2)}(0) = 0$. However, practical single-photon emitters are probabilistic sources, and the measured value of $g^{(2)}(0)$ depends on the emission dynamics, detector resolution, and measurement time window. Consequently, even high-quality single-photon sources may exhibit a finite $g^{(2)}(0)$ due to multiphoton emission events, background photons, detector dark counts, and limited photon statistics. Therefore, while $g^{(2)}(0)$ remains an important metric for characterizing single-photon purity, its interpretation requires consideration of the experimental conditions [145]. The third-order correlation function

quantifying the suppression of simultaneous three-photon events is defined as [146]:

$$g^{(3)}(0) = \frac{\langle \hat{a}^\dagger \hat{a}^\dagger \hat{a}^\dagger \hat{a} \hat{a} \hat{a} \rangle}{\langle \hat{a}^\dagger \hat{a} \rangle^3} = \frac{\langle \hat{n}(\hat{n} - 1)(\hat{n} - 2) \rangle}{\langle n \rangle^3}. \quad (4.6)$$

Third-order correlations are measured with an extension of the standard HBT setup. In this case two beam splitters and three detectors are necessary.

Beam splitters are not only tools to study coincidence probabilities but are also used to entangle photonic modes and are used to study Hong-Ou-Mandel (HOM) interference [147]. Going back to the single-photon incident on the beam splitter described by the state $|\psi\rangle = |1\rangle_1 |0\rangle_2$ one can mathematically demonstrate the entangling capability of the beam splitter. The state can also be written via the creation operator $\hat{a}^\dagger |0\rangle_1 |0\rangle_2$ and the beam splitter transforms the state to $(r\hat{a}^\dagger + t\hat{a}^\dagger) |0\rangle_3 |0\rangle_4 = r |1\rangle_3 |0\rangle_4 + t |0\rangle_3 |1\rangle_4$, which describes a path-entangled state of a photon in path 3 and path 4. Entangled means that the state cannot be factored into a product of two individual states.

HOM interference occurs when two perfect indistinguishable photons impinge on a 50:50 beam splitter from different input ports. Both photons due to their indistinguishability will always take the same output port meaning they bunch and the probability to detect one photon in each output arm is zero. This manifests in a dip in the coincidence measurement spectrum for zero time delay. Using this concept the other way around, the HOM dip provides an experimental measure of photon indistinguishability. By varying the temporal delay between the two photons impinging on a beam splitter, the coincidence detection rate exhibits a minimum when both photons arrive simultaneously. The depth of this dip is quantified by the visibility and is a measure for the temporal, spatial, and spectral overlap [147].

This discussion shows that the beam splitter itself already is non-trivial optical element. By cascading two beam splitters, a Mach-Zehnder interferometer (MZI) can be constructed to study interference. Classically, interference arises from the superposition of electromagnetic field amplitudes. For two fields with amplitudes E_1 and E_2 the intensity is different from simply their simple sum. The intensity is proportional to $I \propto |E_1 + E_2|^2 = |E_1|^2 + |E_2|^2 + 2\text{Re}\{E_1 E_2^*\}$. The last term depends on the relative phases and gives rise to constructive and destructive interference [48]. In quantum optics, interference is observed between probability amplitudes.

A MZI consists of two beam splitters, two mirrors, a phase shifter introducing a relative phase delay ϕ between the two paths before recombination at the

second beam splitter. Depending on whether classical light states, single photons, two distinguishable photons, two indistinguishable photons or even entangled states are used as input states of the MZI, different interesting observations can be made. The interference effect in all cases arises from the same linear unitary transformation implemented by the same interferometer. The difference between classical and quantum interference only originates solely from the choice of input state.

For classical light, the input field can be described by a coherent state $(|\alpha, 0\rangle)$, where $|\alpha|^2$ corresponds to the mean photon number. The interferometer acts linearly on the field amplitudes, resulting in output intensities $I_1 \propto \cos^2(\phi/2)$ and $I_2 \propto \sin^2(\phi/2)$. This behaviour reflects the interference of classical electromagnetic fields and can be fully explained within classical wave optics.

For a single-photon entering input mode a , the action of the interferometer produces the output state $|\psi\rangle = \cos(\phi/2)|1, 0\rangle + i \sin(\phi/2)|0, 1\rangle$ leading to detection probabilities $P_1 = \cos^2(\phi/2)$ and $P_2 = \sin^2(\phi/2)$. So if this measurement is repeated for a large number of events, the same interference as in the classical picture is observed.

For two indistinguishable photons, genuine multiphoton interference occurs. For an input state $|2, 0\rangle$, the output state is $|\psi\rangle = \cos^2(\phi/2)|2, 0\rangle + i\sqrt{2}\cos(\phi/2)\sin(\phi/2)|1, 1\rangle - \sin^2(\phi/2)|0, 2\rangle$. The coincidence term arises from the indistinguishable process where both photons are transmitted and reflected, demonstrating quantum interference beyond the single-photon picture. Using entangled input states, phase sensitivities below the classical shot-noise limit can be achieved, which is of particular interest for quantum metrology [148].

Conventional beam splitters are limited to two output modes, making the realization of high-dimensional interferometers challenging.

Diffraction gratings also provide a way for distributing light into multiple spatial modes. The propagation angles are defined by the grating equation

$$d \sin(\theta_m) = m\lambda, \quad (4.7)$$

where d is the grating period, λ the wavelength, and m the diffraction order. With metasurfaces, spatially varying phase profiles can be engineered that allow the redistribution of light into multiple diffraction orders with tailored intensities. Each diffraction order can be seen as an optical mode, allowing metasurfaces to be viewed as multiport beam splitters.

4.2.2 Creating Single Photons

There are different platforms available to generate single photons. Commonly used platforms include spontaneous parametric down-conversion [149], semiconductor quantum dots [150], colour centers in diamond such as nitrogen-vacancy centers [151, 152] or two-dimensional materials [153, 154]. In spontaneous parametric down-conversion (SPDC), one pump photon is converted into a signal and an idler photon by interacting with the nonlinear material. The process is probabilistic, meaning that photon pairs are generated at random times. A second critical issue is that the probability of creating n pairs scales with the pump intensity to the n th power. Increasing the power thus increases the occurrence of multiphoton emission events [149, 155].

A second, well-established approach to generating single photon is based on epitaxially grown semiconductor quantum dots. They act as artificial atoms with discrete energy levels and can be approximated as two-level systems consisting of a ground state $|g\rangle$ and an excited state $|e\rangle$. There are different schemes to optically excite the quantum dot, for example, with resonant excitation [156, 157], excitation via the biexciton [158–160], phonon-assisted processes [161–164] or the swing up scheme [165, 166]. In a simplified view, in all these schemes an electron is excited to the excited state. The radiative recombination leads to the emission of a photon with a well-defined energy. After emitting a photon, the system returns to the ground state and needs to be re-excited before another photon can be emitted [155]. The QD in our experiment is excited by a phonon-assisted scheme. The laser excitation is energetically blue-detuned from the emitter, so that it is not resonant with the QD emission. The QD is excited by emitting phonons to compensate the energy difference between excitation laser and s-shell transition.

4.3 Design of Beam Splitting Functionality

In the following section, the functionality and design of the metasurface are presented. For the purpose of the experiment, a dielectric metasurface-based multipath beam splitter is designed. Figure 4.2a shows the intended capability of this metasurface. An incoming input beam in_1^0 (in stands for input, the subscript 1 refers to the first metasurface, and the superscript 0 indicates the input channel) is diffracted into three output paths (out^{-1} , out^{+1} and out^{+2}) along the x-direction that correspond to the -1, +1, and +2 diffraction order. Equal intensities of $I_{out^m} = \frac{1}{3}$ are targeted in all three output paths, where

m denoted the number of the diffraction order. The ideal design does not

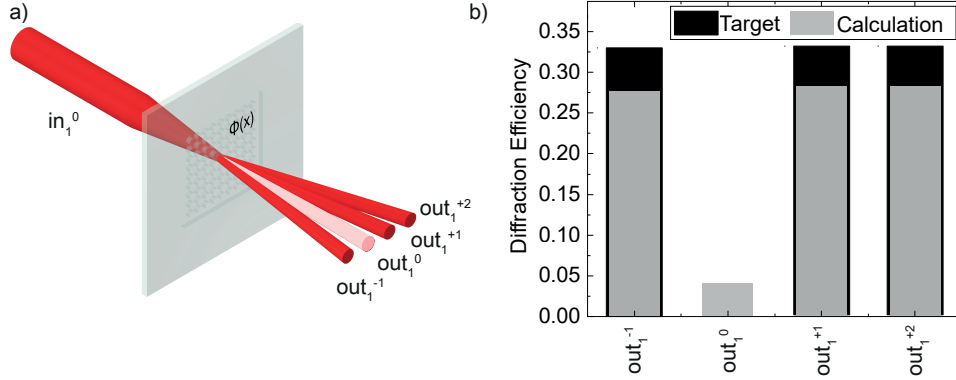


FIGURE 4.2: Design of the metasurface-based multiport beam splitter: (a) Conceptual sketch of the beam splitting functionality of a single metasurface showing the diffraction into four output ports. (b) Comparison of the target and simulated diffraction efficiencies from the designed spatial phase profile.

target the zero-order beam on purpose so that it later can be used as a measure for the efficiency of the metasurface. Light that does not interact with the metasurface is transmitted in its original propagation direction.

As a next step, the spatial phase profile $\Phi(x)$ that meets these demands must be determined. A gradient descent optimization is employed, which minimizes the error function e_m

$$e_m = \sum_{m=1}^3 \text{abs} \left(I_m - \frac{1}{3} \right), \quad (4.8)$$

where I_m refers to the calculated intensities of output path m . If a minimum value of e_m is found, an optimized spatial phase profile is obtained. Each intensity I_m is calculated based on the square of the Fourier coefficients C_x^m , expressed as:

$$C_x^m = \frac{1}{2\pi} \int_0^D e^{i\Phi(x)} e^{i\frac{2\pi m x p}{D}} dx_p \quad (4.9)$$

However, the metasurface can only implement the found spatial phase profile in a discretized manner, where each meta-atom imparts a specific phase shift corresponding to its position. Hence, the integral is replaced by a sum:

$$C_x^m = \frac{1}{2\pi} \sum_0^D e^{i\Phi(x)} e^{i\frac{2\pi m x p}{D}} \quad (4.10)$$

where x_p represents the coordinate of the meta-atoms in the periodic supercell. Each pixel is $p = 400 \text{ nm}$ in size, giving a total supercell length $D = n \cdot p = 22 \cdot 400 \text{ nm} = 8.8 \mu\text{m}$. The resulting diffraction angle depends

Antenna index	1	2	3	4	5	6
Size (nm)	150×155	150×150	140×145	140×145	140×145	140×140
Antenna index	7	8	9	10	11	12
Size (nm)	140×140	135×135	120×125	110×115	110×115	100×105
Antenna index	13	14	15	16	17	18
Size (nm)	100×100	105×110	115×120	125×125	125×125	125×125
Antenna index	19	20	21	22		
Size (nm)	125×125	125×125	115×120	155×155		

TABLE 4.1: Size of every meta-atom within the supercell.

on the length of the supercell and is calculated with the grating equation Eq. 4.7 $\theta = \arcsin(\frac{\lambda}{D}) = 5^\circ$. The larger the total supercell the smaller the diffraction angle, however this also increases the size of the metasurface and thus fabrication times. If the diffraction angle is too large, the collection of the diffracted beams becomes difficult.

During optimization the spatial phase profile $\Phi(x)$ is iteratively varied until the minimum value of the error function e_m is found, meaning the best target intensity distribution is achieved. The generated spatial phase profile after convergence of the algorithm is displayed in Fig. 4.8a. This optimized discretized spatial phase profile yields diffraction efficiencies of $I_{-1,+1,+2} = 0.28$ and $I_0 = 0.04$ as calculated from the Beam Propagation Method.

The initial design of the metasurface was provided by Dr. Ruizhe Zhao from the Beijing Institute of Technology. The propagation phase was chosen as the method to encode distinct phase values to the metasurface. The phase difference arises from different lateral parameters, different meta-atom heights or a change in the refractive index. Varying the lateral dimensions of each meta-atom is the simplest approach considering nanofabrication complexity. To implement the designed spatial phase profile to the metasurface, a mapping between lateral dimensions and achieved phase difference is needed, which is done by numerical simulations of different geometries. As the material amorphous silicon on a fused silica substrate is selected. The individual meta-atoms have a rectangular shape. The numerical simulations are performed with the goal of finding the right phase values while simultaneously providing high transmission values. The lateral dimensions of every meta-atom in the supercell are presented in table 4.1. The height of every meta-atom is 600 nm and the individual meta-atoms are spaced by a periodicity of 400 nm.

For the purpose of this thesis, additional simulations of the metasurface were performed, which are described in detail in the following section. The aim is

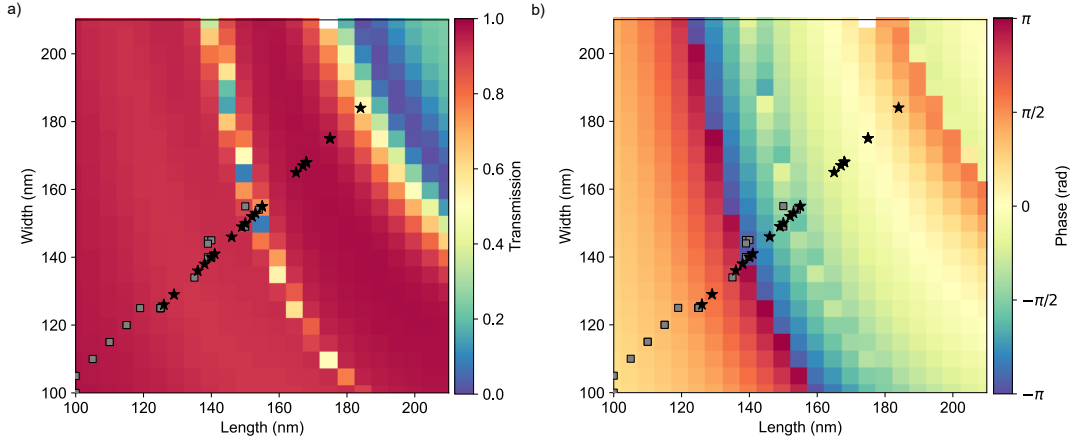


FIGURE 4.3: Simulated transmission and phase change for a range of lateral dimensions of the meta-atom. The incident wavelength, the height of the meta-atom, and the periodicity remain fixed at $\lambda=785$ nm, $h=600$ nm, and $p=400$ nm, respectively. Grey rectangles show the dimensions of the initial simulations, while black stars indicate the sizes of the fabricated metasurface.

to investigate the sensitivity of the designed dimensions to discuss tolerances and limitations of the design. A full-wave 3D finite-element method that solves the time-harmonic Maxwell equations in the frequency domain is used. The simulation domain consists of a fused silica substrate with a height of ($h=3\text{ }\mu\text{m}$ and a silicon nanopost that is surrounded by air ($h=3\text{ }\mu\text{m}$). In the $x-y$ -plane periodic boundary conditions are used and in the z -direction a perfectly matched layer is used. The refractive index $n = 3.69$ and the extinction coefficient $k = 0.003$ of the silicon nanopost was measured by ellipsometry (for further details see Chapter 3). The nanopost is illuminated from the top using a port with a linearly polarized electric field along the x -direction. The transmission is evaluated using the S_{21} -parameter and the phase change is calculated from this parameter as $\phi = \arg(S_{21})$.

Based on this simulation setup, a systematic parameter sweep is performed to evaluate how the lateral dimensions of the nanopost influence both transmission and phase. The antenna pitch and the height remain fixed at 400 nm and 600 nm, respectively, while the lateral dimensions of the nanoposts are varied. Two considerations need to be taken into account to find suitable parameters. First, if the antenna pitch exceeds 541 nm additional unwanted diffraction orders occur because then the individual unit-cell supports higher diffraction orders. Second, it is undesirable that the meta-atom supports Mie resonances as they also lead to a reduction of the transmittance. Therefore, the individual meta-atom will have a small width and a high aspect ratio. In Figure 4.3, the influence of the length and width of the antenna on transmission and phase

change is depicted. The antenna length and width were varied from 100 nm to 210 nm in 5 nm steps, and corresponding phase and transmission values are extracted from the simulations. The transmission is shown in Fig. 4.3a. Low transmission values are indicated in blue, while high values are indicated by red. Near-unity transmission is observed for a large range of meta-atom sizes. While high transmission can be achieved over a broad range of geometries, the phase response exhibits stronger variations. The phase map demonstrates a more modulated behaviour than the transmission due to the appearance of Mie resonances for larger geometries. Based on this behaviour, suitable geometries are selected to construct a look-up table that links each required phase value to a specific nanopost dimension. The designed nanopost sizes are indicated in Fig. 4.3 as grey rectangles.

The designed metasurface is realized using electron beam lithography and the etching-based process described in Chapter 3. To assess how closely the fabricated structure matches the intended design, the metasurface is analyzed using scanning electron microscopy (SEM). A SEM micrograph (Fig. 4.4) of the fabricated metasurface is used for this analysis. The highlighted region indicates the supercell consisting of 22 meta-atoms that is repeated to form the complete metasurface of $400\text{ }\mu\text{m} \times 400\text{ }\mu\text{m}$. The image reveals that the meta-atoms deviate from the intended rectangular geometry, exhibiting slightly rounded corners despite applying proximity effect correction. To quantify the lateral dimensions, the SEM micrograph is analyzed using a Python-based routine. Each image row is converted into a line profile of pixel intensity. Due to the strong contrast between the fused silica substrate and the silicon meta-atoms, the resulting profile resembles a step function. A threshold value of 200 (approximately 80% of the maximum grayscale value) is used to identify the boundaries of individual structures. From this, the widths are extracted by counting pixels and converting them using the pixel size (10.87 nm), yielding meta-atom dimensions in the range of 120 nm to 185 nm. The exact measured sizes are illustrated as stars in Fig. 4.3. The tilted SEM image further indicates the presence of a residual chromium layer on top of the nanoposts, although it was intended to be fully removed during wet etching. Consequently, the measured dimensions represent an upper bound, as slight underetching of the mask implies that the actual silicon structures are smaller.

As discussed in Chapter 3, proximity effect correction is essential for achieving well-defined rectangular meta-atoms. However, it also introduces a trade-off between pattern fidelity, file size and writing time. During the optimization of

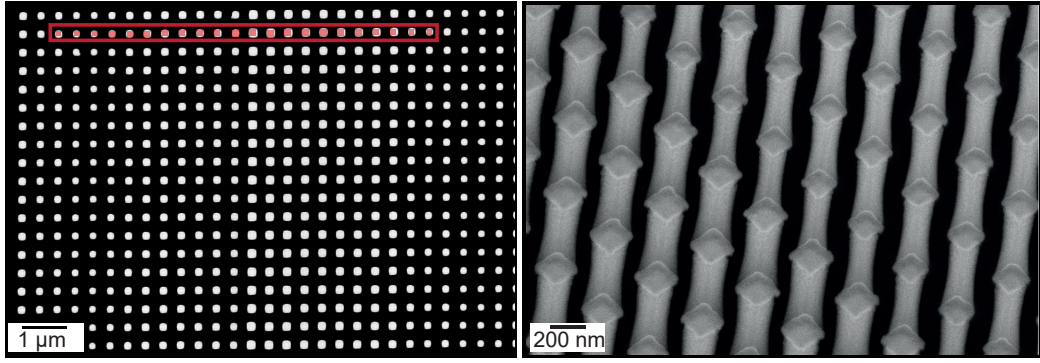


FIGURE 4.4: SEM images of the fabricated metasurface (top and tilted view).

the lithography routine, several observations were made. The first observation is that the increased file size leads to reproducibility issues. The applied proximity effect correction (PEC) significantly increases the file size (up to 1.8 GB compared to 0.18 GB) due to an increased number of vertices as the meta-atoms are subdivided into smaller iso-dose segments. During the lithography process, sometimes the corrected doses were not applied, resulting in circular-shaped meta-atoms instead of rectangular ones as demonstrated in Fig. 4.5. This occurred randomly, but at more than one occasion, sometimes even within a single fabrication run resulting in two very different samples that were fabricated within one sample run. As a solution to this problem, the $400\text{ }\mu\text{m} \times 400\text{ }\mu\text{m}$ large metasurface was segmented into smaller parts, each being $8.8\text{ }\mu\text{m} \times 8.8\text{ }\mu\text{m}$ in size. One supercell has a length of $8.8\text{ }\mu\text{m}$ and was repeated 22 times in the y -direction. However, this segmentation was only possible because the metasurface consists of periodically repeated supercells. The positionlist (PLS), which is written during lithography no longer contains only one entry, but instead contains 2000 entries to build the complete metasurface. This procedure avoided the issue of the doses not being applied correctly. Additionally, it also decreased the writing time to around three hours instead of ten hours. A plausible explanation for the initial issue is a memory limitations of the pattern generator, as smaller segments reduce the number of elements stored in the buffer simultaneously. Although segmentation can introduce stitching errors, no significant artifacts were observed here, since the unit-cell period (400 nm) is large compared to the stitching accuracy ($\approx 10\text{ nm}$). As the lithography process seems to be to some extent a product of luck and deviations from the ideal rectangular geometry remain, it is interesting to assess the sensitivity of the optical response to variations in meta-atom geometry and shape. In particular, the impact of the lateral geometry, incident wavelengths and meta-atom height is analyzed. Only the

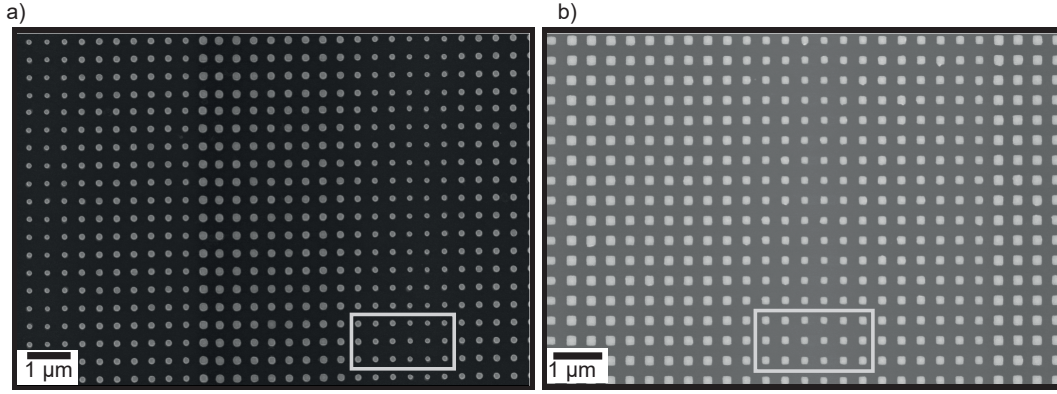


FIGURE 4.5: Comparison of two lithography results: (a) In the absence of properly applied PEC, the insufficient local dose leads to underexposed and rounded corners with reduced size. (b) Correct application of the PEC results in well-defined, rectangular meta-atoms.

observation of the difference in phase itself is not meaningful, therefore, the diffraction behaviour is simulated with the Beam Propagation Method for the extracted spatial phase profiles for different meta-atom parameters.

First the influence of the lateral geometry is investigated by comparing circular and rectangular cross-sections. Figure 4.6 compares the calculated transmittance and phases as a function of the width/diameter for rectangular and circular meta-atoms, referred to as nanoposts and nanopillars, respectively. Both geometries are evaluated for the design wavelengths of 785 nm, a height of 600 nm, and a periodicity of 400 nm. The grey area indicates the range of designed sizes of the meta-atoms. Within the designed range, the transmittance is nearly identical for both geometries. For the nanopost geometry, a dip in the spectrum for a width of 155 nm and 190 nm is visible indicating an electric quadrupole and a magnetic dipole Mie resonance. For the nanopillar, both resonances are shifted to larger diameters. The insets in Figure 4.6 show the electric field profiles in xz -direction. For smaller widths, e.g. 90 nm no mode is confined within the meta-atom. However, the phase response exhibits significant differences. While the nanopost geometry provides almost complete phase coverage from $-\pi$ to π , the nanopillar geometry only covers approximately the range from 0 to π . Therefore, deviations from a perfect rectangular shape will result in a modified spatial phase profile encoded in the metasurface.

The influence of variations in meta-atom height is shown in Figure 4.7b. A height variation of ± 10 nm around the nominal value of 600 nm has only a minor effect on the transmittance and phase response. Noticeable changes occur only for larger lateral dimensions, where the resonant response becomes

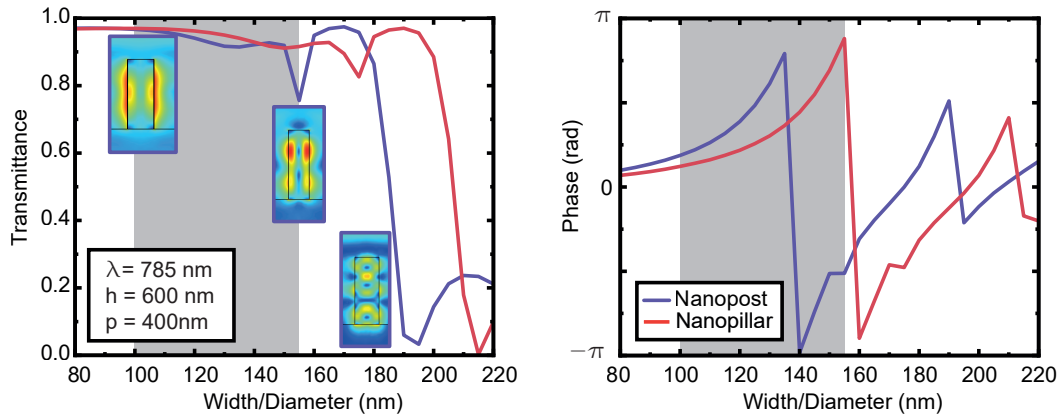


FIGURE 4.6: Comparison of transmittance and propagation phase for a metasurface composed of nanopillars and nanoposts. The different geometries lead to different optical response, affecting both the transmittance and the propagation phase. As a result, the choice of meta-atom design and fabricated size influence the achievable phase coverage.

more pronounced. Since the investigated height variation corresponds to typical thickness deviations during the deposition of silicon layers using PECVD, the propagation phase approach is relatively robust against vertical fabrication uncertainties.

Besides geometrical variations, the incident wavelength provides an additional degree of freedom for modifying the optical response. Figure 4.7a compares the transmittance and propagation phase for different incident wavelengths. The transmission remains nearly unchanged for small lateral dimensions, while the position of the Mie resonance shifts for larger dimensions. The phase response in contrast exhibits an approximately linear shift for different illumination wavelengths. This behaviour demonstrates that the incident wavelength can serve as a post-fabrication tuning parameter, allowing to compensate for deviations in the lateral dimensions. Overall, this analysis presents both advantages and disadvantages of propagation phase encoding. The phase response is sensitive to lateral geometry variations and deviations from the intended meta-atom shape which can modify the encoded phase profile. In contrast, variations in meta-atom height have only a limited influence. Furthermore, the wavelength dependence of the propagation phase provides an opportunity for adjusting the optical response after fabrication. To assess how these effects influence the overall functionality of the metasurface, the resulting diffraction behaviour of every phase mask is computed with a Beam Propagation Method. Using this method, the propagation of a light beam passing through a spatial phase profile can be simulated and the diffraction efficiencies are extracted. As a first step, an initial beam profile (here: Gaussian

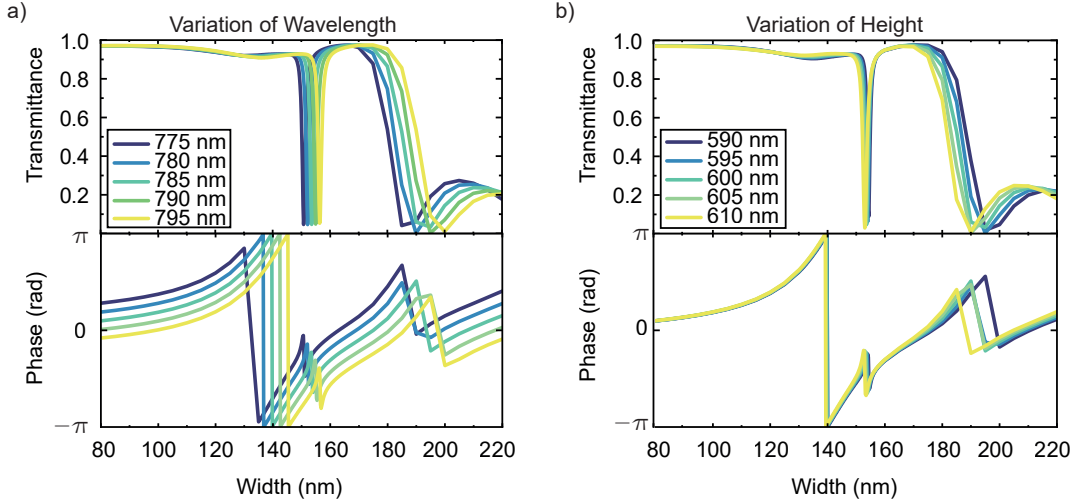


FIGURE 4.7: Comparison of transmittance and accumulated propagation phase for different wavelengths of the incident light and for meta-atoms with different heights. (a) The transmittance remains approximately constant for different wavelengths, while the accumulated propagation phase exhibits a linear wavelength-dependent shift. (b) A ± 10 nm variation in meta-atom height affects the transmittance and phase only for lateral dimensions larger than approximately 180 nm.

beam) is defined. The beam is then propagated along the z -direction before it is sent through the phase mask $\phi_{\text{Metasurface}}$. After the phase mask, the beam is again propagated in free space.

The Gaussian input beam is given by $E_{\text{gauss}} = E_0 \cdot \exp(\frac{-R^2}{w_0^2})$, where $E_0 = 1$, R is the radius and w_0 is the beam waist. The Gaussian input is multiplied with the phase mask $E = E_{\text{Gauss}} \exp(i\phi_{\text{Metasurface}})$ to give the phase-modulated field directly after the phase mask. In my calculation, the phase mask is represented by 22 values for every meta-atom in the supercell that was found in the simulation of the phase profile. The free space propagation over the distance d is calculated in the spatial frequency domain. Therefore, the Fourier transform of the field $\tilde{E}(f_x, f_y)$ is calculated. The propagation is given by $H(f_x, f_y) = \exp(2\pi i d \sqrt{\frac{1}{\lambda^2} - f_x^2 - f_y^2})$. The propagated field is obtained by the Fourier transform of $E_{\text{propagated}} = \mathcal{FFT}(\tilde{E}(f_x, f_y) \cdot H(f_x, f_y))$. The experiment records the k -space image of the diffraction pattern, hence $\tilde{E}(f_x, f_y) \cdot H(f_x, f_y)$ is analyzed in the simulation.

Figure 4.8a compares the spatial phase profile of one supercell and the obtained diffraction pattern in k -space for a nanopost, a nanopillar and the fabricated metasurface. This comparison allows for a direct evaluation of how design choices and fabrication imperfections impact the diffraction efficiency. The top panel displays the spatial phase that each of the 22 meta-atoms encodes and the lower panel shows the $x - y$ -cut of the k -space diffraction pattern where

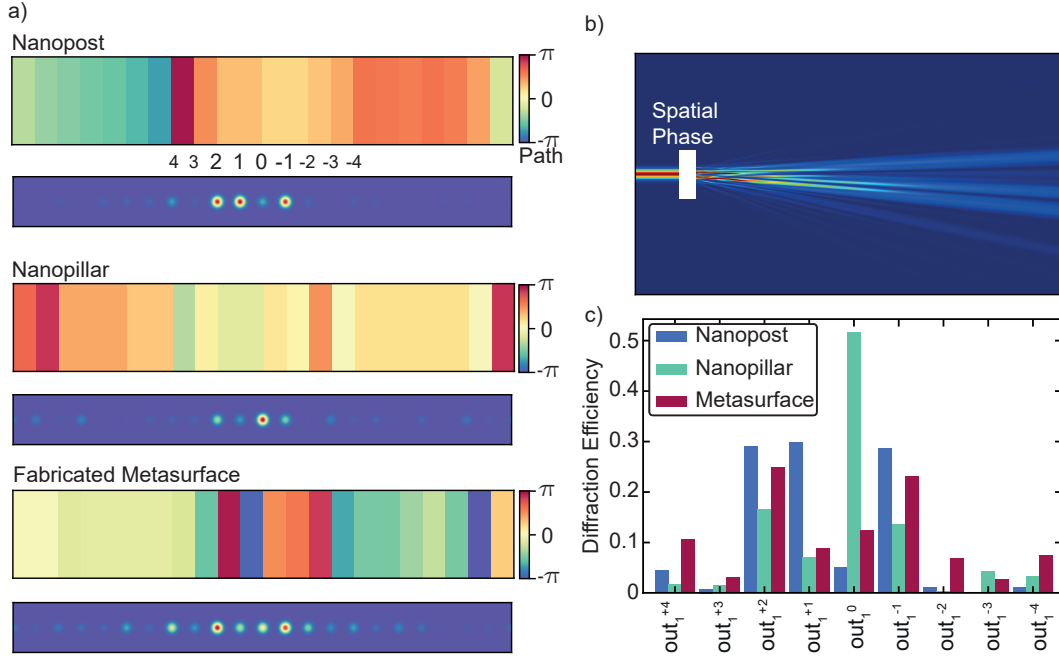


FIGURE 4.8: Robustness analysis of metasurface design based on the propagation phase. (a) The spatial phase profile and resulting k-space diffraction pattern are shown for the designed dimension assuming a rectangular nanopost and a circular nanopillar. Additionally, both plots are calculated for the fabricated metasurface dimensions.

the diffraction spots of orders -4 to $+4$ are labelled. Figure 4.8b shows the spatial propagation of the incident beam that is diffracted by the metasurface (visualized by the white box) and the propagation in z -direction. For this figure the designed phase profile is used. The diffraction efficiencies of the three different cases evaluated from panel a are shown in Figure 4.8c. Comparing the diffraction efficiencies of the nanopost with the nanopillar demonstrates the importance of the correct shape of the meta-atom. The nanopillar ensemble does not efficiently diffract the beam as the non-diffracted zero order has an efficiency of 0.51 and the other output paths have lower efficiency than 0.2. The phase profile assumed for the *fabricated metasurface* can only provide an approximation to the encoded phase profile because only one supercell is used for the BPM calculation and the phase profile was calculated for perfectly rectangular corners. Hence, the calculated diffraction pattern will deviate from the experimentally measured results.

Nevertheless, this analysis demonstrates that while metasurfaces based on the propagation phase offer high design flexibility, their performance is highly sensitive to fabrication inaccuracies, making precise control of the meta-atom geometry essential for achieving the desired optical functionality. The sensitivity to the shape can be reduced by designing circular-shaped nanopillars as

rounded corners are easier to fabricate than sharp ones.

4.4 Beam Splitting with Classical Light

In this section, the experimental results obtained using a classical light source are presented. We begin by describing the experimental setup, followed by a discussion of the results. First, we characterize the diffraction efficiency of each of the two metasurfaces individually. They are then cascaded to build a high-dimensional multipath interferometer which is analyzed in terms of phase-dependent intensity modulation.

4.4.1 Experimental Realization

For the individual characterization as well as the investigation of the cascaded system, the same experimental setup is used. It consists of a light source, the individual or cascaded metasurfaces, and a detection unit based on a $4f$ -imaging system and a CCD camera. A schematic of the whole setup is shown in Fig. 4.9. As the metasurface phase profile was optimised for a wavelength of 785 nm, a laser diode with a central wavelength of 785 nm (Thorlabs LP785-20) is used for all classical measurements. The emitted light is collimated using a fibre collimator with a fixed-focus aspheric lens with a focal length of $f=11$ mm (Thorlabs F220FC-780). The incident polarization is controlled by a combination of a linear polarizer (LP) and a half-wave plate (HWP). A lens with a focal length of $f=300$ mm focuses the beam to match the size of the metasurface. At the first metasurface (MS1), the diffracted beams are generated. Two identical achromatic lenses with a focal distance of $f=35$ mm (AC254-035-B-ML) relay the diffracted beams onto the second metasurface at the same angles as the output angles of MS1. Between the lenses the phase of the individual beams can be manipulated as the light passes through glass slides that can be rotated to change the optical path lengths. At the second metasurface (MS2), the beams interfere to create new phase-dependent outputs. The outputs are then collected by a microscope objective (MO) (20x, NA=0.6). A $4f$ -imaging system composed of two achromatic lenses ($f=100$ mm, Thorlabs AC508-100-B-ML) is used to image the back focal plane (BFP) of the MO onto a CCD camera. In this configuration, the individual diffraction orders appear as distinct spots, allowing the diffraction efficiency of each output channel to be determined. The first lens of the $4f$ -system (indicated by the grey outline) is mounted on a magnetic holder for easy

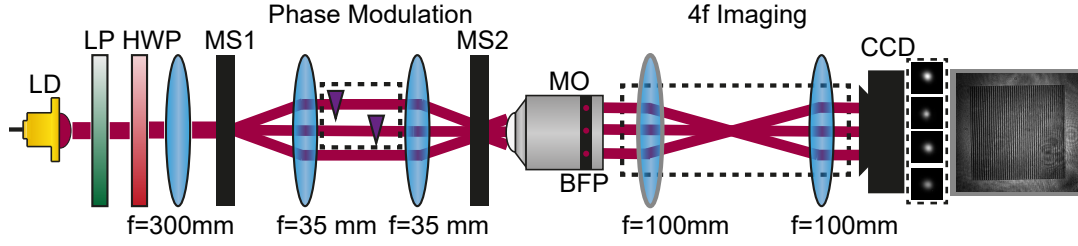


FIGURE 4.9: Schematics of the classical characterization setup: By removing the lens indicated by the grey outline, the metasurface is directly imaged onto the CCD. When both lenses behind the microscope objective (MO) are included, the back focal plane (BFP) of the objective is imaged instead allowing visualization of the individual diffraction spots.

removal. When this lens is removed, the real-space image of the metasurface is projected onto the camera (Andor Solis Zyla). An example of such an image is shown on the right-hand side of Fig. 4.9. For the determination of the diffraction efficiencies of the individual metasurface, only a single metasurface is mounted at the position of MS2 and the glass slides are removed from the setup.

For data analysis, images acquired with the $4f$ - configuration are processed as follows. Either the diffraction efficiency DE_m (relative efficiency) or the transmission T_m (absolute efficiency) of every output path m can be determined. For the analysis of the transmission, first a reference measurement only through the glass substrate is acquired. This serves as the reference value I_0 . Then the transmission T_m is calculated:

$$T_m = \frac{I_m - I_{\text{background}}}{I_0 - I_{\text{background}}} \quad (4.11)$$

The diffraction efficiency is analyzed as follows:

$$DE_m = \frac{I_m - I_{\text{background}}}{\sum (I_m - I_{\text{background}})} \quad (4.12)$$

For each diffraction order, the intensity I_m is obtained by integrating over a region of interest of 50×50 pixels centred on the corresponding spot. A background region of the same size is selected in a dark area of the image to determine the background signal, which is subtracted from all measured intensities.

4.4.2 Characterization of Single Metasurface

The experimentally measured diffraction efficiencies for the two fabricated metasurfaces are shown in Fig. 4.10. Four dominant diffraction spots with

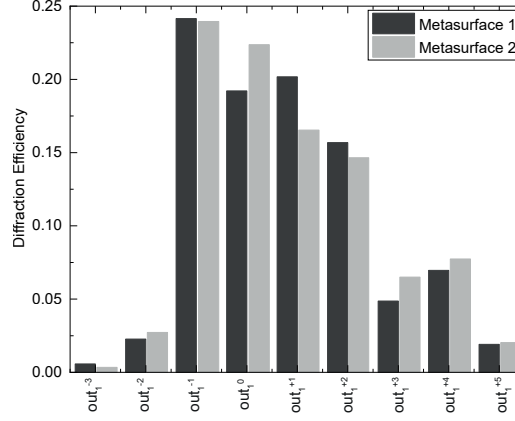


FIGURE 4.10: Comparison of the relative diffraction efficiencies measured for both individual metasurfaces.

diffraction efficiencies from 0.15 to 0.25 are observed. Additionally, a smaller fraction of light is also diffracted to higher diffraction orders. The deviations arise from the imperfect rectangular shape of every meta-atom, the remaining Cr on top of the antenna and the increased dimension of every antenna which alter the spatial phase profile of the metasurface that results in modified diffraction patterns as discussed in the previous section. The additional path out_m^0 is simply used as an additional mode for the subsequent experiments. The splitting ratios between the desired diffraction orders are not the only relevant parameters for the experiment, but also the overall transmission efficiency and associated losses are of interest. For the selected diffraction channels $-1, 0, +1$, and $+2$, the combined transmission efficiency is approximately 50% for each metasurface. The remaining fraction of the incident power is distributed among higher diffraction orders and reflected light. Although the losses reduce the available optical power in the desired diffraction orders, the generated modes still maintain sufficient efficiency for the proof-of-principle demonstration and the implementation of the metasurfaces as a high-dimensional interferometer.

4.4.3 High-dimensional Interferometer made from Cascaded Metasurfaces

Next, the cascade of the two metasurfaces is studied. The general concept of the cascaded metasurfaces building the interferometer is illustrated in Fig. 4.11. The first metasurface acts as the multiport beam splitter creating four output beams and the second one is used to create the coherent superposition of its inputs. As the different output paths of the first metasurface serve as the inputs for the second metasurface e.g. out_1^0 is renamed to in_2^0 . Because of the

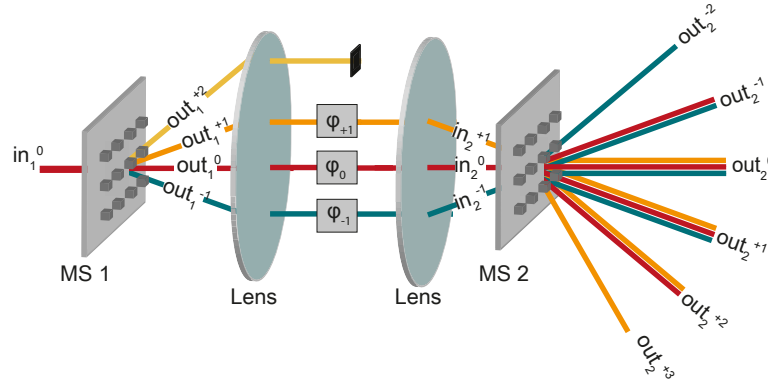


FIGURE 4.11: Conceptual illustration of the cascaded metasurface to study phase-controlled interference.

principle of optical reciprocity, the cascade of exactly the same metasurface twice will lead to the recombination into a single beam if all beams impinge on the second metasurface with the appropriate relative phase relation. However, experimentally, the generated beams have different path lengths, such that the correct relative phase relation is not achieved. As a result, the new input paths do not recombine into one beam but each incident beam generates new output paths. We exploit this behaviour to form an $N \times M$ interferometer. If the light beam is incident at an angle, the resulting diffraction pattern shifts by the incident angle according to the law of momentum conservation, leading to an increase in the number of output paths. Consequently, the interferometer has more output paths than inputs. This configuration results in the coherent superposition of multiple diffracted beams in certain output paths, as illustrated in Fig. 4.11. It is important to note that the photons cannot travel to every possible output port which leads to a different number of interfering beams per output port. Phase shifters φ_0 and φ_{+1} are inserted in the corresponding beam paths, and the phase-dependent transmission of the cascaded system is measured. To simplify the experimental configuration, the

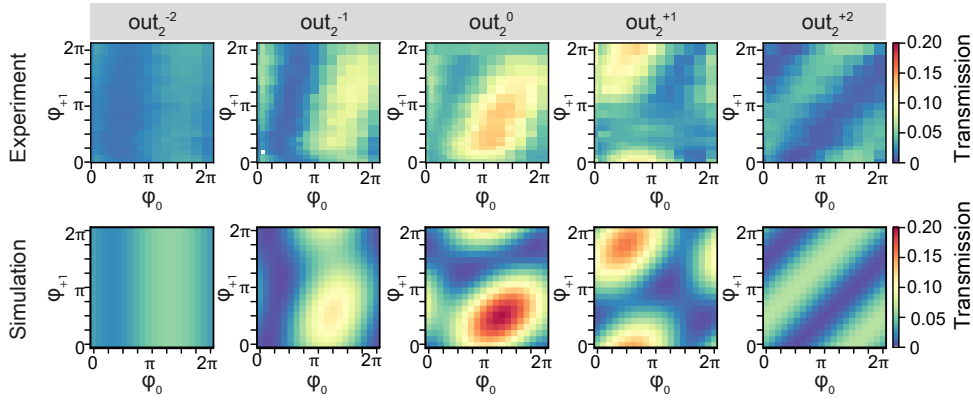


FIGURE 4.12: Experimentally measured (top row) and calculated (bottom row) transmission maps for different phase settings. Good agreement between experiment and simulation is observed, confirming the phase-dependent modulation of the output intensity arising from interference between different input channels.

channel out_1^{+2} is blocked. For each combination of the phase values φ_0 and φ_{+1} , the transmission after the second metasurface for the different output orders is recorded. The resulting two-dimensional transmission maps are displayed in Fig. 4.12 and show the transmission in output path $\text{out}_2^{-2,-1,0,+1,+2}$ as a function of both phases φ_0 and φ_{+1} .

The measured transmission maps exhibit pronounced modulation of the output intensities for all output modes as the relative phases are varied. Depending on the phase, constructive interference enhances the transmission while destructive interference suppresses it. As a result, the relative phase provides a deterministic control parameter that enables active switching and continuous tuning of the power distribution among the output paths. However, the achievable interference visibility is limited by the finite efficiency of the metasurface and the associated optical losses.

The characteristic structure of each transmission map is directly linked to the number of interfering beams contributing to a given output channel. For example, the diagonal interference pattern observed in channel out_2^{+2} indicates that this output depends simultaneously on both phases φ_0 and φ_{+1} . Overall, the periodic redistribution of optical power among the output paths demonstrates coherent interference controlled purely by the relative phase. The cascaded metasurfaces operate as a phase-tunable, multiport interferometer analogous to a Mach-Zehnder interferometer, but realized in a planar geometry. This highlights the metasurface's ability to manipulate the output configurations across multiple modes within a compact footprint. The metasurface-based network allows for straightforward up-scaling as the number of modes can

readily be increased. The dimensionality of this design can be increased by including the fourth beam that was blocked for the experiments or a new metasurface with a greater number of diffraction orders can be designed and fabricated.

4.4.4 Analytical Calculation of the Transmission

The theoretically expected output intensities are calculated using a matrix-based model, where each matrix describes the transformation of the electric field at a specific stage of the experimental setup. The transmission after the second metasurface is expressed as

$$|[E_{\text{out}}]|^2 = |[MS_2] \cdot [\text{Phase}] \cdot [MS_1] \cdot [E_{\text{in}}]|^2 \quad (4.13)$$

where the vector $[E_{\text{in}}]$ represents the incident electric field amplitude, which is normalized to unity. The matrix $[MS_1]$ represents the first metasurface and describes how the incident field is redistributed into the four output modes $\text{out}_1^{+2,+1,0,-1}$ for an input entering through in_1^0 . It contains four elements corresponding to the transmission coefficients t_m^{MS1} . Upon diffraction, each beam gains an additional phase term $e^{i\phi_m}$ that is specific to that respective path. The phase matrix accounts for the phase shifts φ_1 and φ_0 applied to the respective channels out_1^{+1} and out_1^0 . Entries equal to unity mean that no external phase shift is applied. When the four beams impinge on the second metasurface, a total of seven output paths out_1^{+4} to out_1^{-2} are generated. This transformation is described by matrix $[MS_2]$, which maps the four inputs to seven outputs. Each beam is split according to its respective transmission coefficient t_m and each beam acquires an additional phase $e^{i\phi_m}$. For the phase maps shown in Fig. 4.12 the absolute square of the electric field vector $[E_{\text{out}}]$ is calculated to obtain the transmission of each channel. The full matrix expression describing the designed metasurface system is given by:

$$\begin{bmatrix} E_{\text{out}+4} \\ E_{\text{out}+3} \\ E_{\text{out}+2} \\ E_{\text{out}+1} \\ E_{\text{out}0} \\ E_{\text{out}-1} \\ E_{\text{out}-2} \end{bmatrix} = \begin{bmatrix} t_{+2} \cdot e^{i\phi_{+2}} & 0 & 0 & 0 \\ t_{+1} \cdot e^{i\phi_{+1}} & t_{+2} \cdot e^{i\phi_{+2}} & 0 & 0 \\ t_0 & t_{+1} \cdot e^{i\phi_{+1}} & t_{+2} \cdot e^{i\phi_{+2}} & 0 \\ t_{-1} \cdot e^{i\phi_{-1}} & t_0 & t_{+1} \cdot e^{i\phi_{+1}} & t_{+2} \cdot e^{i\phi_{+2}} \\ 0 & t_{-1} \cdot e^{i\phi_{-1}} & t_0 & t_{+1} \cdot e^{i\phi_{+1}} \\ 0 & 0 & t_{-1} \cdot e^{i\phi_{-1}} & t_0 \\ 0 & 0 & 0 & t_{-1} \cdot e^{i\phi_{-1}} \end{bmatrix} \cdot \begin{bmatrix} 1 & 0 & 0 & 0 \\ 0 & e^{i\varphi_{+1}} & 0 & 0 \\ 0 & 0 & e^{i\varphi_0} & 0 \\ 0 & 0 & 0 & 1 \end{bmatrix} \cdot \begin{bmatrix} t_{+2}^{\text{MS1}} \cdot e^{i\phi_{+2}} \\ t_{+1}^{\text{MS1}} \cdot e^{i\phi_{+1}} \\ t_0^{\text{MS1}} \\ t_{-1}^{\text{MS1}} \cdot e^{i\phi_{-1}} \end{bmatrix} \cdot [E_{\text{in}}] \quad (4.14)$$

This matrix model allows for several qualitative predictions. First, some output paths receive contributions from only a single beam. In this case, no phase-dependent modulation of the transmission is expected. One example is E_{out_2} which is only generated by in_2^{-1} . Second, other output paths are generated by the interference of two beams. In such cases, the transmission depends on the relative phases between the contributing modes. For example, the output path out_2^{-1} is generated by the beams propagating through the inputs in_2^{-1} and in_2^0 . However, only the latter path is externally phase-modulated. Consequently, the transmission depends solely on the phase $e^{i\varphi_0}$, which results in a one-dimensional modulation pattern in the transmission map. In contrast, the output path out_2^{+2} is formed by interference of beams from in_2^0 and in_2^{+1} . Since both of these paths are phase-modulated, the transmission depends on both $e^{i\varphi_0}$ and $e^{i\varphi_{+1}}$, leading to the observed diagonal interference pattern. However, the diffraction efficiency measurement of a single metasurface shows that diffraction order beyond +2 to -1 are also generated by the metasurface, although with lower efficiency. The experimentally measured transmission map for out_2^{-2} confirms the contribution of those modes to the overall behaviour of the cascaded system as the map shows a modulation indicating phase-dependent transmission and showing that those lower orders cannot be universally neglected.

In the following paragraph the procedure used to determine a suitable matrix describing the experimentally observed transformation is described step by step. The simplest and broadest description is to use the designed values for $[\text{MS}_1]$ and $[\text{MS}_2]$ as transmission coefficients t_m in both matrices. The designed coefficients are $t_{-1,+1,+2} = \sqrt{0.28}$ and $t_0 = \sqrt{0.04}$, while all other contributions are neglected. The calculated phase maps are shown in Figure 4.13. More specifically, matrix $[\text{MS}_1]$ and matrix $[\text{MS}_2]$ read as follows:

$$[\text{MS}_1] = \begin{bmatrix} \sqrt{0.28} \\ \sqrt{0.28} \\ \sqrt{0.04} \\ \sqrt{0.28} \end{bmatrix} \quad \text{and} \quad [\text{MS}_2] = \begin{bmatrix} 0 & 0 & 0 & 0 \\ 0 & \sqrt{0.28} & 0 & 0 \\ 0 & \sqrt{0.28} & \sqrt{0.28} & 0 \\ 0 & \sqrt{0.04} & \sqrt{0.28} & \sqrt{0.28} \\ 0 & \sqrt{0.28} & \sqrt{0.04} & \sqrt{0.28} \\ 0 & 0 & \sqrt{0.28} & \sqrt{0.04} \\ 0 & 0 & 0 & \sqrt{0.28} \end{bmatrix} \quad (4.15)$$

The schematic illustration on the left side of the following figures explains how the different input beams are distributed and interfere in their respective output paths. Output paths out_2^{-2} and out_2^{+3} exhibit no phase dependence,

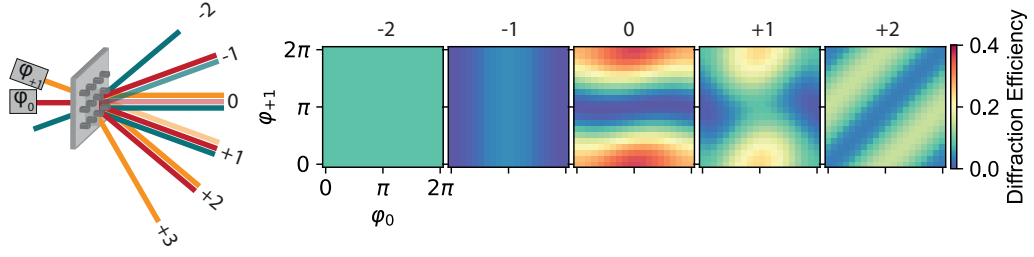


FIGURE 4.13: Phase-dependent transmission calculated from the designed transmission coefficients.

as only a single optical path contributes in each case, resulting in a constant transmission. In output paths out^{-1} , the input beams in^0 and in^{-1} interfere, while only the phase φ_0 is varied resulting in a one-dimensional modulation. In output channel out^0 , three beams interfere with each other and both phases φ_{+1} and φ_0 are varied. However, the intensity of input in^0 is comparatively weak, which explains why the modulation is dominated by in^{+1} as indicated by the predominantly vertical modulation pattern. In output path out^{+1} , three beams again interfere. However, in this case the input intensity of in^{+1} is weaker. This explains why the orientation of the phasemap now is in the exactly opposite direction. In output out^{+2} only the beams in^0 and in^{+1} interfere and both inputs have the same intensity, resulting in the pronounced diagonal phase dependence.

When comparing the simulated phase maps with the experimental data, significant discrepancies are observed. Only the output out^{+2} shows reasonable agreement, while all other outputs differ. To address this, the model is extended by replacing the designed transmission values with the experimentally measured values as presented in Table 4.2. Additionally, the -2. diffraction order is included in the calculation, such that each input beam now contributes to paths $\text{out}^{-2,-1,0,+1,+2}$.

Path	Transmission MS1	Transmission MS2
-2	0.01371	0.01718
-1	0.14577	0.15094
0	0.11596	0.14098
+1	0.12177	0.10421
+2	0.09467	0.09233

TABLE 4.2: Measured transmission values for both metasurfaces

The result is shown in Fig. 4.14. With these modifications, phase-dependent modulation is now also observed in output channel out^{-2} . While the overall phase dependence is described more accurately, noticeable deviations from

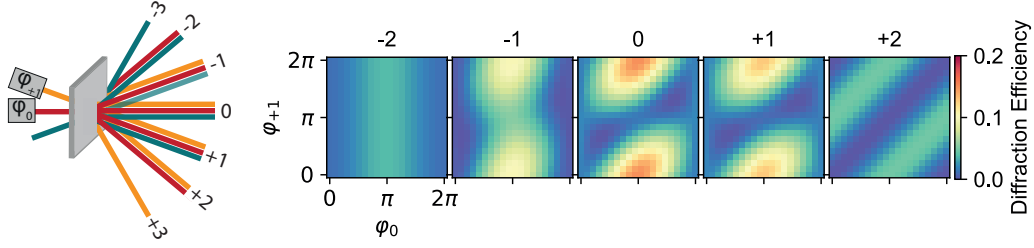


FIGURE 4.14: For the calculation of the transmission the contributions from -2 to +2 are taken into account.

the experimental data remain, indicating that additional physical effects must be taken into account. As a final refinement, we consider the complex phase contribution $e^{i\phi_m}$ of each diffraction order, where ϕ_m represents the phase associated with the corresponding Fourier component of the metasurface transmission function C_m^x . This leads to the modified expression:

$$\begin{aligned}
 [\text{MS}_1] &= \begin{bmatrix} t_{+2}^{\text{MS1}} e^{i\phi_{+2}} \\ t_{+1}^{\text{MS1}} e^{i\phi_{+1}} \\ t_0^{\text{MS1}} \\ t_{-1}^{\text{MS1}} e^{i\phi_{-1}} \end{bmatrix}, \quad [\text{MS}_2] = \begin{bmatrix} t_{+2} e^{i\phi_{+2}} & 0 & 0 & 0 \\ t_{+1} e^{i\phi_{+1}} & t_{+2} e^{i\phi_{+2}} & 0 & 0 \\ t_0 & t_{+1} e^{i\phi_{+1}} & t_{+2} e^{i\phi_{+2}} & 0 \\ t_{-1} e^{i\phi_{-1}} & t_0 & t_{+1} e^{i\phi_{+1}} & t_{+2} e^{i\phi_{+2}} \\ 0 & t_{-1} e^{i\phi_{-1}} & t_0 & t_{+1} e^{i\phi_{+1}} \\ 0 & 0 & t_{-1} e^{i\phi_{-1}} & t_0 \\ 0 & 0 & 0 & t_{-1} e^{i\phi_{-1}} \end{bmatrix}
 \end{aligned}
 \tag{4.16}$$

In Figure 4.14, the calculated transmission pattern in out^{+1} and out^0 is nearly identical. This pattern is also observed in the experiment albeit phase shifted. As the same inputs generate the paths out^{+1} and out^0 this phase shift can only be explained if, upon diffraction, different phases ϕ_{+1} and ϕ_0 is acquired for both inputs.

To determine the phase values ϕ_{+2} , ϕ_{+1} , and ϕ_{-1} , a numerical optimization routine was implemented in Python. The algorithm calculates phase maps for a range of phase combinations and compares them to the experimental data. The optimal phase values are identified by maximizing the similarity across all five output channels simultaneously. The comparison between the experimentally measured phase maps and the calculated results is shown in Fig. 4.11. Good agreement is observed, demonstrating that the inclusion of diffraction-induced phase terms is essential to accurately reproduce the experimental behaviour.

This formalism accounts for both amplitude redistribution and phase-dependent interference, illustrating how the relative phases between different outputs can deterministically steer light in the cascaded metasurface system. However, this approach to model the interferometric behaviour is phenomenological and does not provide a fully rigorous description of quantum optical phenomena. For this, the matrix describing the lossless beam splitting behaviour must be unitary [167]. Several approaches exist that allow for the measurement of the unitary matrix describing an $N \times N$ linear optical network. Anthony Laing and Jeremy L. O'Brien presented a method in 2012 relying on non-classical interference to characterize the unitarity of an optical network [168]. Rahimi-Keshari et al. presented a method in 2012 that relies only on standard laser sources and intensity measurements to directly measure this matrix. Since their method relies on classical light sources it is easier to implement experimentally. The entries of a unitary matrix M are generally complex numbers $M_{jk} = r_{jk}e^{i\theta_{jk}}$ and it has been shown that for $N > 3$ the knowledge of r_{jk} is not sufficient to determine all missing phases [167], therefore, all phases $e^{i\theta_{jk}}$ and transmission coefficients r_{jk} need to be measured. To achieve this, two different measurements need to be performed. To determine r_{jk} , a coherent state with known intensity I is sent to input mode j while the other input states are in the vacuum state and the transmission in every output path k is evaluated as $r_{jk} = \sqrt{I_k/I}$. Second, to determine the phase a coherent state needs to be split by a 50:50 beam splitter and the phase ϕ between both inputs needs to be controlled. One input is sent to one input serving as the reference and the second mode with the varying phase is sent into input mode j . The intensity I_k is recorded as a function of the incident phase ϕ and the phase of the matrix element $e^{i\theta_{jk}}$ is obtained as $\theta_{jk} = 2\pi - \phi$. This measurement needs to be repeated for all input combinations. With the obtained matrix for a $N \times N$ linear network now the behaviour of indistinguishable and distinguishable photons can be predicted and effects such as high-dimensional HOM interference can be studied [167].

4.5 Interferometry with a Quantum Light Source

In the following section, results obtained with a quantum light source are presented and compared to measurements performed with classical light. First, we repeat the experiment with a single metasurface and demonstrate its capability to measure second- and third-order photon correlations. In a second step, the cascaded system is employed to demonstrate single-photon interference.

4.5.1 Experimental Setup

The experimental setup that employs the single metasurface as a multiport beam splitter is nearly identical to the one used for classical measurements, but it is incorporated into a larger experimental platform consisting of several interconnected setups. These different units of the experimental network are described below and they are shown in Figure 4.15a-e. The different components to conduct the experiment are a μ -Photoluminescence setup, a spectrometer, a transmission monochromator, the HBT setup using the metasurface instead of a conventional cube beam splitter, and the cascaded metasurfaces.

At the heart of the experiment is the generation of single photons with a semiconductor GaAs quantum dot. The GaAs layer was grown by molecular beam epitaxy and the quantum dots are embedded inside circular Bragg cavities (CBR) to enhance the photon extraction efficiency. Without the CBR, total internal reflection at the semiconductor-vacuum interface decreases the extraction efficiency. However, embedding the quantum dot inside this cavity overcomes these limitations by engineering a resonant cavity mode that efficiently funnels the emitted photons out of the semiconductor material. The concentric Bragg rings act as a reflector that couples the in-plane emission into a defined vertical radiation mode with a tailored far-field profile. According to Fermi's golden rule, the spontaneous emission rate is proportional to the product of emitter-field coupling strength and the optical density of states at the cavity frequency [169]. The resonant cavity mode enhances the local density of optical states at its resonance frequency that is engineered to be identical to the quantum dot transition frequency which increases the spontaneous emission rate through the Purcell effect. As a result, a larger fraction of the emitted photons is directed toward the collection optics which improves the extraction efficiency. The sample was fabricated by the research groups of Huber-Loyola, Höfling, Rastelli, and Trotta. More detailed information on the

sample itself as well as characterisation measurements can be found in [170]. Single photons are generated via the process of photoluminescence (PL). Luminescence generally describes the process of a material spontaneously emitting radiation which originates when the material relaxes from an excited state into its ground state. In the case of photoluminescence, this excitation is induced optically through photon absorption [171]. The photoluminescence setup itself is modular and consists of an excitation unit, cryogenic microscope, and detection and analysis units, which can be flexibly interconnected using optical fibers.

For optical excitation of the QD, different laser sources can be employed and sent to the PL-setup. For above-band excitation a continuous-wave green laser diode with a wavelength of 522 nm is used. In the experiments presented here, the QD is excited via a phonon-assisted excitation scheme [162, 164] which requires a near-resonant pulsed laser. For this purpose, a femtosecond mode-locked Ti:sapphire laser (MIRA, Coherent) with a repetition rate of 80 Hz is used. Due to its broadband emission, spectral filtering with a pulse shaper is necessary, and an excitation wavelength of ≈ 794 nm is selected to excite the quantum dot. Additionally, a narrow bandwidth diode laser (Toptica DL Pro) was employed for alignment purposes only. Its wavelength is tuned to match the QD emission, enabling precise alignment of the fiber couplers. Each laser source is coupled into a single-mode fiber and directed to the PL-setup. For out-coupling, a fiber collimator is chosen such that the beam diameter is approximately 2.8 mm to match the entrance aperture of the microscope objective. The resonant excitation laser is guided into the cryostat via a 90:10 beam splitter, while the green laser is coupled using a dichroic mirror.

The GaAs QD sample is mounted inside a closed-cycle cryostat (Attodry 800 by attocube) and cooled to 5 K. The sample is mounted on piezoelectric actuators to adjust the focus and move the sample. An imaging unit is integrated into the setup. By inserting a flip-mounted pellicle into the beam path, the sample can be illuminated with a fiber-coupled LED and imaged onto a camera. This allows simultaneous use of laser excitation and wide-field illumination to align the CBR.

The excitation laser is focused onto the sample using a microscope objective (attocube, 50x, NA 0.81), which also collects the emitted photoluminescence. The residual excitation light is filtered using a dichroic mirror and a notch filter before the signal is coupled into a fiber and routed to different analysis or detection units. Spectral analysis of the signal is performed using a spectrometer

4.5. Interferometry with a Quantum Light Source

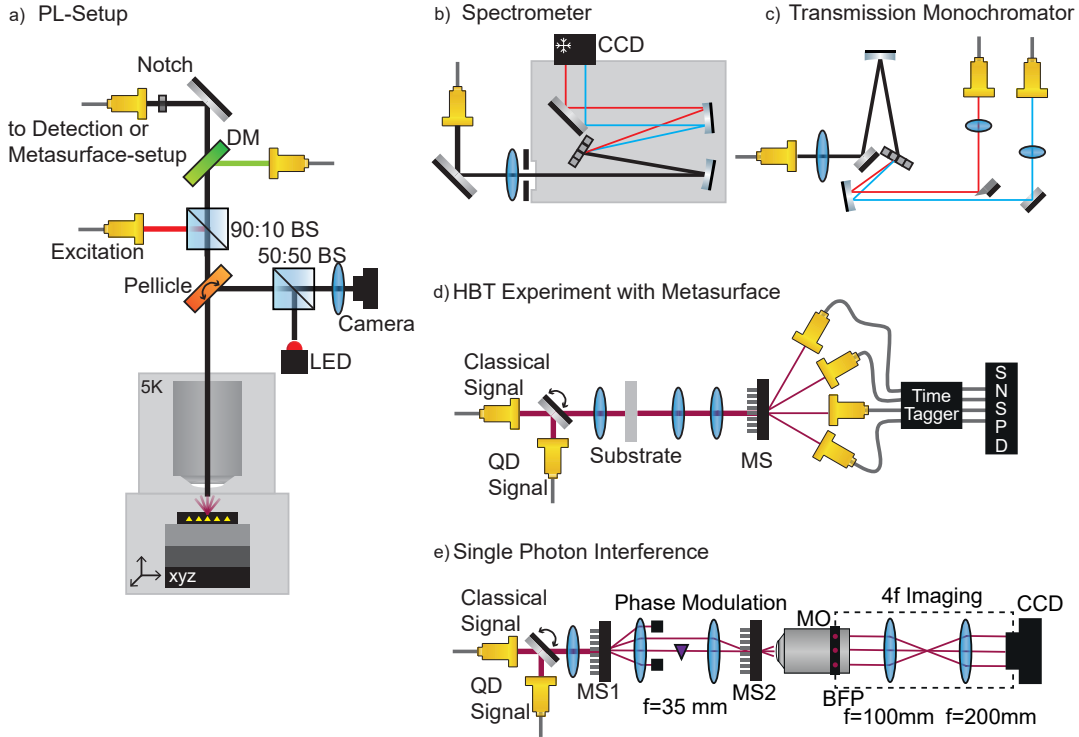


FIGURE 4.15: Schematic overview of the experimental setups used for quantum optical measurements: a) Photoluminescence (PL) setup combining multiple excitation sources; the emitted single photons are directed to various analysis modules and an imaging system. b) Spectral analysis of the emitted photons using a spectrometer consisting of a diffraction grating and a pixelated CCD detector. c) Transmission monochromator for additional spectral filtering with a bandwidth of 19 GHz. d) Metasurface-based setup acting as a higher-dimensional extension of a Hanbury Brown and Twiss experiment, enabling access to higher-order photon correlations within a single device. e) Cascade of two metasurfaces used to investigate single-photon interference effects.

(Princeton Instruments). The system consists of a set of parabolic mirrors and a grating, which spatially separates the individual spectral components of the QD emission. These are detected with a liquid nitrogen cooled pixelated CCD (Princeton Instruments PyLoN). A measured spectrum is shown in Figure 4.16a showing both the QD emission and a residual contribution of the excitation laser. For additional spectral filtering of the signal, a transmission monochromator (TM) is used. The signal is expanded with a lens and a mirror before it is sent onto the transmission grating. The different spectral components of the signal are spatially separated and focused by a second parabolic mirror. At the focal point D-shaped mirrors are mounted on translation stages to separate two distinct lines. In both separated paths the signal is collimated with a lens and again fiber coupled. After this additional filtering, the signal is either directed to a HBT setup for correlation measurement using superconducting

nanowire single-photon detectors to characterize the single-photon emission or first sent to the metasurface setup for further experiments.

Two different kinds of experiments are conducted with the metasurface. The first experiment uses a single metasurface and repeats the HBT experiment but instead of a cube BS the metasurface is used for beam splitting allowing to analyze higher order correlations with only a single device. The setup itself was not constructed from scratch, but the setup from the classical measurements was extended and adapted for the quantum optical measurements (see Figure 4.15d/e). Instead of using a CCD to detect the signal, the four outputs are fiber coupled. Both classical laser signal and QD emission can be coupled into the setup via two fiber couplers and switching between the two is enabled by a mirror on a magnetic mount. As the same setup is used for the experiment with both the single- and dual-metasurface experiments the first three lenses remain in the setup, but the single metasurface is mounted at the second position. There the incident light is split into the different paths $out^{+2,+1,0,-1}$ which are coupled into fibers. The four different paths are then routed to the SNSPDs.

The cascade of both metasurfaces is used to study single-photon interference. The first metasurface is mounted in the first position where the paths $out^{+2,+1,0,-1}$ are generated. Two lenses with focal length of 35 mm are used to redirect the light onto the second metasurface as in the experiment with the classical light. However, now all paths except out_1^{+1} and out_1^0 are blocked. To change the phase between the two paths, a glass slide on a rotation stage is placed in path in_2^0 . The signal that interacts with the second metasurface is collected with the microscope objective and imaged with a $4f$ -imaging setup onto a CMOS camera (Omegon Pro veTEC 533M).

4.5.2 Characterisation of the Quantum Dot

The GaAs quantum dot sample contains many quantum dots and many resonators, so as a first step a bright quantum dot with strong anti-bunching needs to be selected on the sample. The brightness can be evaluated using the count rate on the spectrometer or at the single-photon detectors, while anti-bunching is characterized using a HBT setup. To record the PL spectrum of the QD and identify its transitions, the sample is excited with the 522 nm laser above-band. In this spectrum, a bright transition corresponding to a charged exciton was identified. For our subsequent measurements, we use this transition but excite it with a pulsed red laser in a phonon-assisted scheme

[164] while simultaneously using a small amount of the green CW laser to neutralize the charge environment.

After the identification of a bright emission line, the single-photon emission from the quantum dot is characterized by measuring the second-order intensity correlation function $g^{(2)}(\tau)$ of the generated light with a HBT setup. The photons are first spectrally filtered by the transmission monochromator and then coupled into a 50:50 fibre-based beam splitter (BS). The two outputs are connected to two SNSPDs in combination with a time tagger (Swabian Instruments). The time tagger stores the arrival time of every detection event with picosecond resolution, enabling a time-resolved correlation analysis between both channels.

The recorded time tagger files are analyzed in Python using a library that was provided by the manufacturer with predefined analysis functions. In particular, the function `TimeTagger.Correlation(channel 1, channel 2, bin width, number of bins)` is used to calculate the temporal correlations between the photon detection events. For the correlation measurement a bin width of 8 ps is chosen and 100000 time bins are evaluated, corresponding to a total analyzed delay window of 800 ns. In the correlation analysis one channel is defined as the reference channel ("start") and the code determines whether a detection event occurs in the second channel for a given time delay τ . Repeating this for all registered detection events yields the number of coincidence counts as a function of the time delay of detection events at the detectors.

The coincidence histogram is depicted in the bottom panel of Figure 4.16a, where the coincidence events are displayed as a function of the time delay. Since the QD is excited with a pulsed laser of a 80 MHz repetition rate, the coincidence histogram exhibits a periodic sequence of peaks separated by the pulse period of the laser. These peaks arise from the periodic excitation and correspond to photon detection events originating from different excitation cycles. The coincidence histogram is generated by correlating the arrival time of each detected photon with all subsequent detection events. Therefore, photons emitted during one excitation cycle are compared with photons detected from the same or other excitation cycles, resulting in peaks separated by the laser repetition period. At zero time delay, the central peak is strongly suppressed compared to the neighbouring peaks. This suppression is the characteristic feature of anti-bunching, demonstrating the absence of simultaneous multi-photon emission. To quantify the degree of anti-bunching, the value of the second-order correlation function at zero time delay is calculated from the

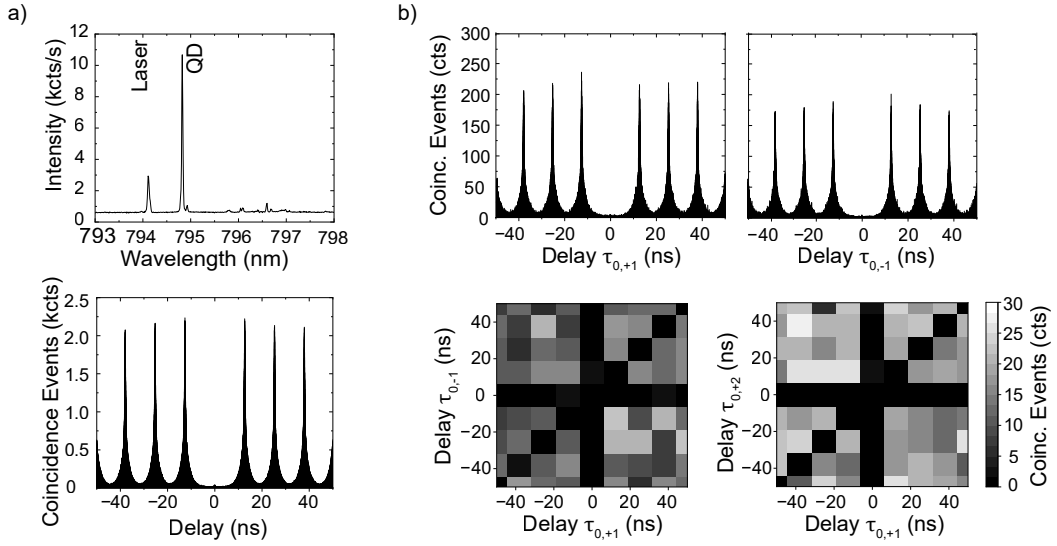


FIGURE 4.16: Quantum measurement performed with a metasurface-based interferometer: (a) Emission spectrum of the quantum dot together with the second-order correlation measurement confirming single-photon emission through antibunching. (b) Experimentally measured second- and third-order correlations demonstrating that the metasurface preserves quantum coherence and nonclassical photon correlations.

integrated peak areas according to

$$g^{(2)}(0) = \frac{2 \cdot A_0}{A_{\text{left}} + A_{\text{right}}}, \quad (4.17)$$

where A_0 denotes the integrated counts of the central peak. An integration window of 2 ns is chosen. A_{left} and A_{right} are the integrated counts of the neighbouring side peaks to the central peak. Using this method gives a value of $g^{(2)}(0) = 0.017 \pm 0.0003$ confirming strong anti-bunching, demonstrating that the emission is dominated by true single-photon events that cannot arise from classical light sources. The uncertainty of the measured $g^{(2)}(0)$ value is determined by error propagation based on the statistical fluctuations of the recorded photon counts, assuming Poissonian counting statistics. Due to the long acquisition time and the resulting high number of detected events, the statistical contribution to the uncertainty is relatively small. However, this value represents only the statistical uncertainty of the measurement. Additional systematic contributions may arise from detector dark counts or residual background emission from the excitation laser. These effects are not included in the presented uncertainty estimate and may lead to an underestimation of the total measurement uncertainty.

4.5.3 Correlation Measurement with a Single Metasurface

Building on the previously described HBT experiment, the fibre-based 50:50 beam splitter is replaced by the metasurface, which acts as a multiport beam splitter. The overall detection and data acquisition remain unchanged, but this time all four output channels are connected to four channels of the time tagger and SNSPDs. Consequently, the correlation analysis is performed with the same python-based workflow using the previously introduced `TimeTagger.Correlation()` function for the analysis of the second-order correlation function. However, this time, all possible combinations of the four channels are analyzed. The time tagger library also has a predefined function for the analysis of the third-order correlation function named `TimeTagger.Histogram2D(channel 1, channel 2, channel 3)` which calculates the coincidence events for three channels as a function of two time delays τ_1 and τ_2 .

In contrast to the two-mode fibre beam splitter, the metasurface redistributes the incident single-photon field into four different output ports. This enables parallel correlation measurements between multiple detector pairs within a single measurement and time-tagging dataset. For each pair of detector channels, the second-order correlation function is calculated using the same method as described above. Figure 4.16b shows two representative second-order intensity correlations measured between output pairs (0, +1) and (0, -1). Both measurements show nearly background-free anti-bunching with an average degree of second-order coherence $g^{(2)}(0) = 0.011 \pm 0.0005$ for all six measurements. This confirms that the metasurface preserves the quantum nature and coherence of single-photon emission. This value is smaller than the one measured with the conventional beam splitter, but as the $g^{(2)}(0)$ value is an intrinsic property of the quantum dot it should not depend on whether the photons are split by the conventional cube beam splitter or the metasurface. The multiport nature of the metasurface further allows an extension of the correlation analysis beyond the second-order and we extend the analysis to third-order intensity correlations $g^{(3)}(\tau_1, \tau_2)$. Using the same time-tagged dataset, we extract the coincidence detection events in three channels. The resulting two-dimensional histogram is shown in Fig. 4.16b for the output path combinations (0,+1,-1) and (0,+1,+2). The data exhibits strongly suppressed multiphoton emission probabilities, visible as three characteristic lines of reduced coincidence counts: i) the first (0) and second (+1) photon arrive at the same time (the vertical line), ii) the first (0) and third (-1) photon arrive at the same time (the horizontal line) and iii) the second (+1) and third (-1)

photon arrive simultaneously (the diagonal feature). These lines reflect the photon antibunching extended to third order, confirming the nonclassical state of light. Importantly, all correlations are extracted from the same time-tagged dataset without modification of the acquisition scheme, highlighting the scalability of the metasurface-based approach. Compared to conventional 2×2 -mode beam splitters, where the measurement of $g^{(n)}(\tau_1, \dots, \tau_n)$ requires $n - 1$ beam splitters, the metasurface provides a compact platform for parallel multi-channel correlation measurements simplifying the experimental setup for higher-order correlation experiments.

4.5.4 Single Photon Interference

Building on these results, we investigate the interference of single photons using the cascaded metasurfaces to build a phase-controlled interferometer. The first metasurface splits the incident light field of the single photon into the four distinct orders. Two of these outputs are selected and directed onto the second metasurface with the help of two lenses. The remaining channels are blocked in order to reduce the system to a two-path interferometer. At the second metasurface, each input generates four output paths. The input beam in_2^0 redistributes the photon field into $\text{out}_2^{-1,0,+1,+2}$, while in_2^{+1} redistributes it into the outputs $\text{out}_2^{0,+1,+2,+3}$. Thus, superposition in modes out_2^0 , out_2^{+1} , and out_2^{+2} is achieved. A phase shifter placed in one of the paths introduces a tunable relative phase φ_{+1} before they are recombined on the second metasurface. The resulting spatial intensity distribution is recorded with the camera. From these images, the diffraction efficiencies of the individual output channels are extracted as a function of the applied phase shift.

Figure 4.17 shows the resulting phase-dependent response for output channels $\text{out}_2^{+3,+2,+1,0,-1}$. As expected, the intensity in the output paths out_2^0 , out_2^{+1} , and out_2^{+2} oscillates sinusoidally with the phase, demonstrating interference between the optical paths. The oscillations reflect phase-dependent redistribution of the optical field among the output modes. Importantly, this behaviour is observed although only a single photon is present in the interferometer as demonstrated by the low value of $g^{(2)}(0)$. This indicates that the interference arises from the superposition of single-photon probability amplitudes rather than from classical intensity mixing.

At the second metasurface, the relative phase between the input modes determines constructive and destructive interference conditions, resulting in the observed modulation of the diffraction efficiencies across the output ports.

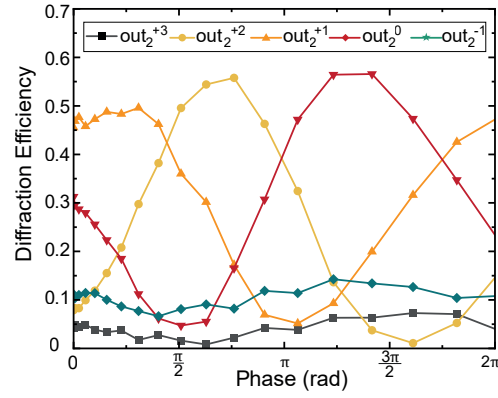


FIGURE 4.17: Phase-controlled redistribution of the light reveals coherent interference at the single-photon level.

This confirms that the metasurface cascade enables controlled interference between spatial modes. The dimensionality of the system can be increased by using more than two outputs of the first metasurface as inputs for the second.

4.6 Conclusion and Outlook

In this work, we have designed, fabricated, and experimentally investigated a metasurface-based multiport beam splitter. The beam splitter operates as a diffraction grating, but with specifically designed output orders and diffraction efficiencies. The simulated spatial phase profile was implemented using the propagation phase which operates by locally varying the lateral dimensions of every meta-atom in the supercell built from 22 meta-atoms. A key advantage of this approach is its polarization-independent operation, as the propagation phase mechanism does not rely on geometric anisotropy or polarization-dependent resonances.

The fabricated metasurface was first characterized structurally using scanning electron microscopy. The SEM micrograph reveals meta-atoms with enlarged lateral dimensions and rounded corners. A sensitivity analysis was performed with the help of the Beam Propagation Method where first the propagation phase for different lateral dimensions, height, and shape of the meta-atoms was simulated. From these simulations, the spatial phase profile was constructed and diffraction efficiencies have been simulated by BPM. This analysis revealed that even small deviations in geometry lead to modified diffraction behaviour. The diffraction efficiencies for all outputs of the fabricated metasurfaces were characterized with classical light and provided the basis for constructing the cascaded interferometric system.

We implemented a high-dimensional Mach-Zehnder interferometer by cascading two metasurfaces and introducing controllable relative phase shifts between optical paths. The classical interference measurements demonstrated clear phase-dependent transmission, confirming multi-path interference within the metasurface optical network. This network can be used to route light in a phase-dependent manner to different output configurations. Additionally, a phenomenological matrix model was developed to calculate the phase-dependent transmission.

Motivated by recent theoretical work on the decomposition of arbitrary unitary transformations into multimode sub-unitary blocks [172], the metasurface and the cascade of metasurfaces were studied in the quantum regime. In particular, we investigated whether the metasurface preserves essential quantum properties such as single-photon purity. Using single-photon emission from a semiconductor quantum dot, we used a single metasurface to measure second- and third-order intensity correlations with a single device and a

single measurement. The observed antibunching demonstrates that the metasurface preserves the single-photon nature of the input state. Furthermore, the metasurface platform enables higher-order correlation measurements within a single device. Additionally, the system was extended to the cascaded metasurface-based interferometer, and phase-controlled single-photon interference was demonstrated.

The metasurface concept is scalable and can be extended to a larger number of modes, enabling increasingly complex optical transformations within a single device. While metasurfaces do not rely on in-plane waveguiding, their integration into existing photonic architecture is possible for example through direct fabrication on fiber end-facets [173, 174].

A next step toward a more rigorous mathematical description is the full reconstruction of the metasurface matrix after the measurement technique proposed by Rahimi et al. [167]. This approach would move beyond the current phenomenological model allowing us to describe both classical and quantum interference including Hong-Ou-Mandel type effects.

From a quantum-optical viewpoint, an important question is whether the metasurface preserves non-classical correlations and entanglement between optical modes. This can be addressed by extending the setup to multiphoton input states which are incident on the metasurface from different spatial inputs. Then interference measurements with indistinguishable photons can be performed. Additionally, a CHSH inequality measurement can be performed to confirm if the metasurface preserves entanglement.

Overall, this work establishes metasurfaces as a possible approach to scale optical networks to large dimensions for both classical and quantum applications.

Chapter 5

Unidirectional High-Harmonic Generation from Hybrid Nanoresonators

5.1	Simulation of the Direction-Dependent Extinction Cross Section	91
5.2	Experimental Demonstration of the Generation of Harmonics	93
5.3	Conclusion and Outlook	100

Conventionally, high-harmonic generation (HHG) is associated with the generation of attosecond light pulses in gases or plasmas [175, 176]. In this case, strong laser fields interact with the medium in a highly nonlinear way, leading to the emission of many odd-order harmonics. Under suitable conditions, these harmonics can form a broad spectrum or even a quasi-continuum, which is required for the generation of ultrashort attosecond pulses. Gas-phase HHG typically operates in the nonperturbative regime, meaning that the harmonic intensities do not follow the power-law scaling expected from perturbative nonlinear optics [177]. HHG based on gas media is well developed, and its generation process is well understood by theory [178]. Compared with gases, solid-state systems offer the advantage of much higher atomic density and periodic lattice structures, which provide additional degrees of freedom for controlling the excitation dynamics and the emitted harmonic signal [179]. More recently, solids such as ZnO [180], GaSe [181], and SiO₂ [182], as well as thin films [183, 184] and metasurfaces [39, 185] have been explored for the generation of HHG. Particularly in these nanophotonic systems, the term HHG is often used in a broader sense. In many cases, the observed process is still

closer to a nonlinear frequency conversion in the perturbative regime or the weakly nonperturbative regime. Strong local field enhancement can enable the generation of higher-order harmonics from subwavelength structures, but typically only a few harmonic orders are observed [185]. Even though the spectral range is more limited compared with gas-phase HHG, the term HHG is widely used to describe nonlinear emission beyond the lowest harmonic orders.

The generation of high harmonics poses several practical challenges. In bulk materials, strong absorption at shorter wavelengths in some material systems can reduce conversion efficiency [186]. In addition, phase matching, i.e., momentum conservation between the pump and generated fields, must be satisfied for a coherent build-up of the harmonic radiation over the propagation distance in the bulk material [187]. Furthermore, efficient shaping and manipulation of ultraviolet radiation remains difficult due to the lack of low-loss optical components, since conventional materials often exhibit strong absorption in this spectral range [188]. Metasurfaces can mitigate several of these limitations by confining the interaction to subwavelength thickness. This reduces propagation losses and relaxes phase-matching constraints. At the same time, the typically small interaction volume limits the overall conversion efficiency. This limitation can be compensated by engineering resonant modes that provide strong field enhancement inside the resonator to increase the nonlinear conversion. Metasurfaces offer a high degree of design freedom, allowing the emitted wavefront to be shaped by imprinting a spatial phase distribution onto the wave. The emission direction can also be engineered, which is relevant for our study of directional harmonic generation. The generation of high harmonics has been studied in different metasurface platforms. Plasmon-enhanced high-harmonic generation by placing gold nanoantennas on top of a silicon substrate was demonstrated [189]. However, plasmonic structures offer strong field enhancement but suffer from intrinsic ohmic losses. This has motivated a shift towards dielectric platforms [37]. HHG in dielectric materials has been explored in amorphous silicon metasurfaces with quasi-bound state in the continuum resonance [39, 190], a free-standing crystalline silicon metasurface membrane [191], a gallium phosphide metasurface [185], and a single aluminium gallium arsenide nanoresonator [192].

In this chapter, we investigate direction-dependent high-harmonic generation (HHG) in asymmetric hybrid nanopillar resonators based on bianisotropic mode coupling. In contrast to conventional metasurface implementations, a

single circular-shaped subwavelength nanopillar is used to achieve nonlinear frequency conversion. Under excitation at frequency ω , the nanoresonator generates higher harmonics at 3ω , 5ω , and 7ω due to the centrosymmetric crystal structure of silicon and silicon nitride. The nonlinear response strongly depends on the direction of illumination due to the asymmetric nanopillar geometry, which enables bianisotropic mode coupling. The presented study extends previous concepts by enabling direction-dependent HHG in a single nanoresonator system. Such asymmetric nonlinear optical responses are of particular interest for all-optical information processing, where direction-dependent signal routing and nonlinear functionalities are required. In addition to directional control, the simultaneous generation of third-, fifth-, and seventh-order harmonics provides multiple information channels within a single nanoscale device.

This project was carried out in collaboration with the group of Sergey Kruk. The nanopillar fabrication was performed by me based on the design and simulations performed by Albert Mathew. The experimental setup was designed, assembled, and aligned during my research stay at the Australian National University (ANU) in Canberra in collaboration with Piyush Jangid (ANU) and Yves Köppeler (UPB). During this period, direction-dependent HHG was experimentally observed for several nanopillar geometries and pump wavelengths. Following this research stay, systematic characterization of nanopillars was completed by Albert Mathew and Piyush Jangid. The manuscript for publication is currently under preparation.

5.1 Simulation of the Direction-Dependent Extinction Cross Section

The objective of this study is to achieve asymmetric control over high-harmonic emission, schematically shown in Figure 5.1a. Under excitation with a laser of frequency ω , the third 3ω , fifth 5ω , and seventh harmonic 7ω of the excitation frequency are generated simultaneously. Due to the asymmetric resonator geometry, the efficiency of harmonic generation depends strongly on the illumination direction. While forward excitation results in efficient HHG, excitation from the substrate side leads to strongly suppressed harmonic emission for a specific nanopillar geometry and pump wavelength. For other combinations of nanopillar diameter and pump wavelength, illumination

from the backside leads to stronger HHG.

The structural asymmetry in the design leads to Ω -type bianisotropy, which

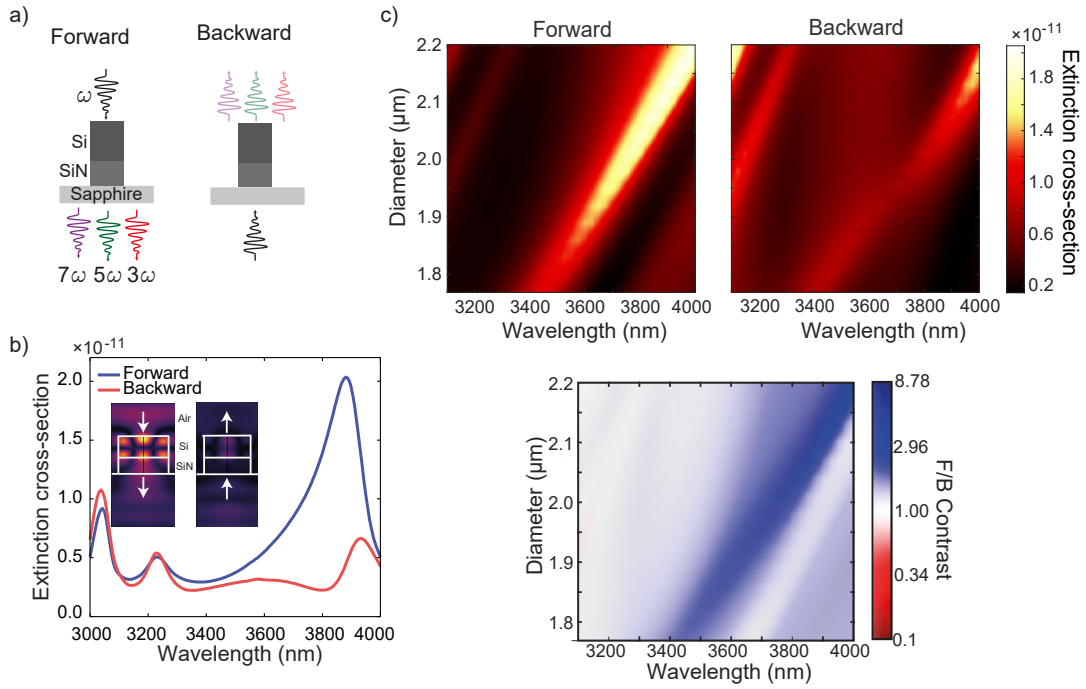


FIGURE 5.1: (a) Schematics of nonreciprocity in an asymmetric silicon-silicon nitride nanopillar (b) Simulated extinction cross section for a nanoresonator with a diameter of $2.1 \mu\text{m}$ under forward (blue graph) and backward (red graph) illumination. The inset shows the electric field confinement for an excitation wavelength of 3875 nm . (c) Extinction cross section as a function of pump wavelength and diameter. From these maps, the Forward/Backward contrast is calculated. The different resonance behaviour under forward and backward illumination is clearly identifiable.

describes the coupling between the electric and magnetic Mie modes. As a consequence, the mode composition of the resonator depends on the illumination direction, resulting in direction-dependent near-field enhancement inside the resonator. Under strong optical excitation, the nonlinear response thus is direction-dependent due to the different field enhancement inside the resonator.

To find suitable nanopillar parameters, the extinction cross section of the asymmetric nanopillar is simulated using COMSOL Multiphysics by Albert Mathew (ANU). The extinction cross section is a measure of how strongly the nanostructure removes energy from the incident electromagnetic wave. It represents an effective area which quantifies the total loss of incident power due to scattering and absorption by the nanoparticle. A large value indicates resonant excitation of optical modes in the nanoresonator, for example, the occurrence of Mie modes resulting in strong near-field confinement inside the

resonator.

The simulated extinction cross section is displayed in Figure 5.1b as a function of pump wavelength for forward (blue graph) and backward (red graph) illumination for a nanopillar with a diameter of 2100 nm. In both cases, the extinction cross section exhibits a local maximum at 3050 nm and 3240 nm, while only the spectrum for forward illumination displays a global maximum at 3875 nm. These peaks in the spectrum indicate Mie resonances. The cross section for backward illumination shows a local maximum that is spectrally red-shifted compared to 3875 nm and significantly reduced in strength.

The insets in Figure 5.1b show the electric field confinement inside the single nanoresonator at a pump wavelength of 3875 nm for forward and backward illumination. Under forward illumination, the electric field is strongly enhanced and confined inside the silicon layer, indicating resonant excitation. In contrast, backward illumination produces weaker field confinement, consistent with the absence of resonant features in the extinction cross section.

Next, the cross section is evaluated as a function of pump wavelengths and diameters, as shown in Figure 5.1c. Increasing the resonator diameter shifts the resonant mode towards longer wavelengths for both forward and backward illumination, consistent with geometric scaling of Mie resonances. To quantify directional asymmetry, a forward-to-backward (F/B) contrast was calculated from the ratio of the extinction cross section. The contrast map for the extinction cross section is shown in the bottom panel of Figure 5.1c. Regions of high F/B contrast correspond to excitation conditions for which the resonator exhibits stronger enhancement under forward illumination, whereas low contrast values indicate enhanced response under backward illumination. The largest contrast for each wavelength and diameter follows a diagonal line along the map, demonstrating that the condition for the strongest directional asymmetry shifts systematically with resonator diameter and excitation wavelength. Based on this behaviour, nanopillars with different diameters were fabricated to investigate both resonant and off-resonant nonlinear responses experimentally.

5.2 Experimental Demonstration of the Generation of Harmonics

The sample consists of a sapphire substrate, a 900 nm thick SiN layer, and a 1210 nm thick Si layer. Nanopillars with diameters from 1770 nm to 2200 nm

were patterned with electron beam lithography and subsequently transferred into the material stack via reactive-ion etching. Sapphire was chosen as substrate material due to its transparency in both the MIR spectral range and the ultraviolet spectral range, in contrast to fused silica, which exhibits strong absorption at the laser pump wavelength and the shorter wavelengths of high-harmonic emission [193, 194].

The experimental setup used for nonlinear characterization is shown schematically in Figure 5.2a and consists of three main components, which are a laser as the excitation source, the nanopillar sample, and a spectrometer for wavelength-dependent signal detection. As the laser source for excitation, an optical parametric amplifier (MIROPA, Hotlight Systems) with a tunable idler wavelength from 2550 nm to 4250 nm is used. The OPA is pumped by a pulsed laser (Femtolux, Ekspla) operating at 1030 nm. The excitation laser is focused

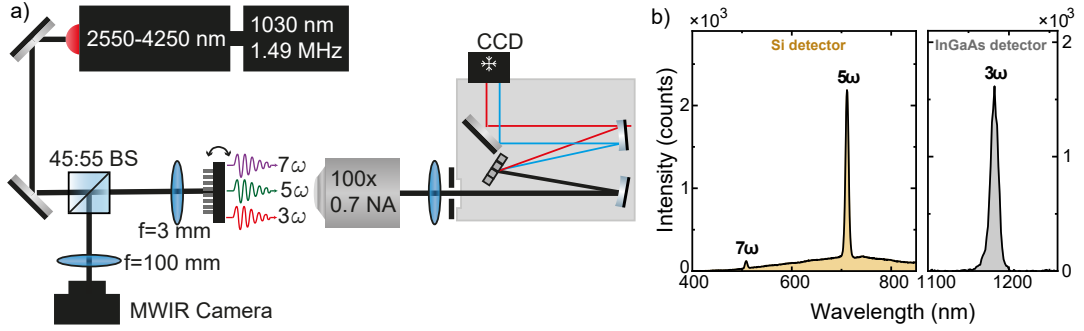


FIGURE 5.2: (a) Experimental setup (b) The representative measured spectra with two different spectrometers show the generation of the 3., 5., and 7. harmonic wavelength.

onto a single nanopillar with a CaF_2 lens of 3 mm focal length. The radius of a focused Gaussian beam is given by $w'_0 = \lambda f / (\pi w_0)$, where λ denotes the wavelength of the light source, f denotes the focal length of the lens, and w_0 is the radius of the input beam. For an incident beam radius of 2 mm, the resulting focal spot diameter is approximately $2w'_0 = 3.1 \mu\text{m}$, enabling excitation of a single nanopillar. To ensure that the laser spot illuminates the chosen nanopillar, a mid-wave infrared (MWIR) camera is employed in a reflection geometry. A removable beam splitter mounted on a magnetic holder is placed before the lens in the beam path. The reflected light is redirected and imaged with a $f = 100 \text{ mm}$ lens made of CaF_2 onto the camera (NIT Tachykon 16k). The sample is mounted on a three-axis translation stage for precise positioning. As the metasurface needs to be measured under frontside and backside illumination, the same stage is used a second time on the opposite side of the beam path so that the illumination direction can be easily changed

by mounting the sample on the different translation stage. The generated harmonic radiation is collected in transmission with a microscope objective (Mitutoyo Plan Apo NIR 100 $\times$, 0.7 NA) and is directed to a spectrometer. For detection, two different spectrometers need to be used. The first one is a Peltier-cooled InGaAs-based detector (Ocean Optics NIQ Quest) with a spectral range of 900 nm to 1700 nm for the detection of the third harmonic. For the detection of the fifth and seventh harmonics a silicon-based detector is used (Teledyne Princeton Instruments, HRS-750 spectrograph). A representative spectrum for a pump wavelength of 3560 nm and frontside illumination is shown in Figure 5.2b. Harmonic peaks are observed at 1186 nm, 712 nm and 509 nm, corresponding to the third, fifth, and seventh harmonics of the excitation wavelengths, respectively. Consistent with the centrosymmetric crystal structure of silicon and silicon nitride, only odd-order harmonics are observed. The spectra of the fifth and seventh harmonics exhibit a broadband background contribution. As expected for higher-order nonlinear processes, the conversion efficiency decreases with harmonic order, resulting in a substantially weaker seventh harmonic emission compared to the fifth harmonic. This trend is consistent with the perturbative scaling of nonlinear polarization. However, to verify the exact scaling, it is necessary to measure the generated signal as a function of excitation power.

The nonlinear response is systematically investigated by varying the pump wavelength and the nanopillar diameter from 1.77 μm to 2.2 μm . The excitation power was fixed at 10 mW throughout all measurements. Higher excitation powers resulted in gradual degradation of the nanopillars, likely caused by thermal damage.

Figure 5.3 summarizes the measured third harmonic generation (THG) response as a function of pump wavelength and nanopillar diameter. Figure 5.3a compares representative THG spectra for two nanopillars measured under frontside and backside illumination. For a nanopillar diameter of $d = 1780$ nm excited at 3605 nm, the THG intensity under frontside illumination exceeds that obtained under backside excitation by approximately a factor of 30. In contrast, for a nanopillar with diameter $d = 2140$ nm pumped at 3184 nm, the THG signal under backside illumination is enhanced by approximately one order of magnitude relative to frontside excitation. Comparison with the simulated extinction spectra reveals that these parameter combinations coincide with resonant conditions for the respective illumination directions. The enhanced nonlinear emission therefore follows the resonant field-enhancement induced by the direction-dependent excitation of the nanoresonator.

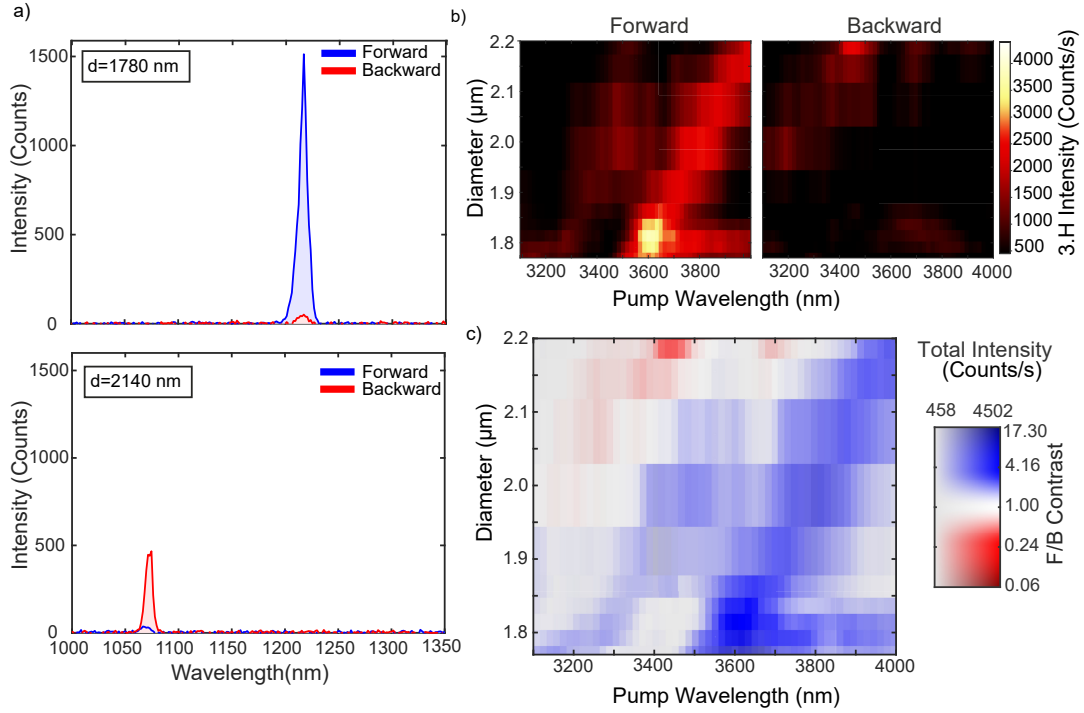


FIGURE 5.3: Measured third-harmonic generation (THG) response of Si-SiN nanopillars. (a) Representative THG spectra for two nanopillars under forward (F) and backward (B) illumination. For a nanopillar with a smaller diameter, THG is enhanced under forward illumination, whereas a larger nanopillar exhibits stronger emission under backward illumination, demonstrating direction-dependent nonlinear response. (b) Measured THG intensity as a function of pump wavelength and nanopillar diameter for forward and backward illumination. Distinct regions of maximum THG intensity are observed for the two illumination directions, indicating different resonant excitation conditions. (c) Forward-to-backward contrast map, calculated from the ratio of the measured THG intensities in (b), highlighting regions of enhanced direction-dependent nonlinear generation.

Panel b compares the THG intensity under forward and backside illumination as a function of pump wavelength and nanoresonator diameter. The corresponding forward-to-backward-contrast $F/B = I_{\text{forward}}/I_{\text{backward}}$ is shown in Figure 5.3c. The measured THG maxima closely follow the resonant regions predicted by the simulated extinction cross section, confirming that nonlinear enhancement is governed by resonant field confinement inside the nanopillar. Whereas the simulations of the extinction cross sections demonstrate maxima for several parameter combinations, the measured THG signal indicates a clear maximum for a pump wavelength of 3605 nm and a diameter of 1790 nm. The THG signal generated for backward illumination is significantly weaker compared to the opposite case. Here, the maximum is observed for a diameter of 2140 nm and a pump wavelength of 3184 nm.

Next, the contrast is evaluated by calculating the ratio of THG intensity for

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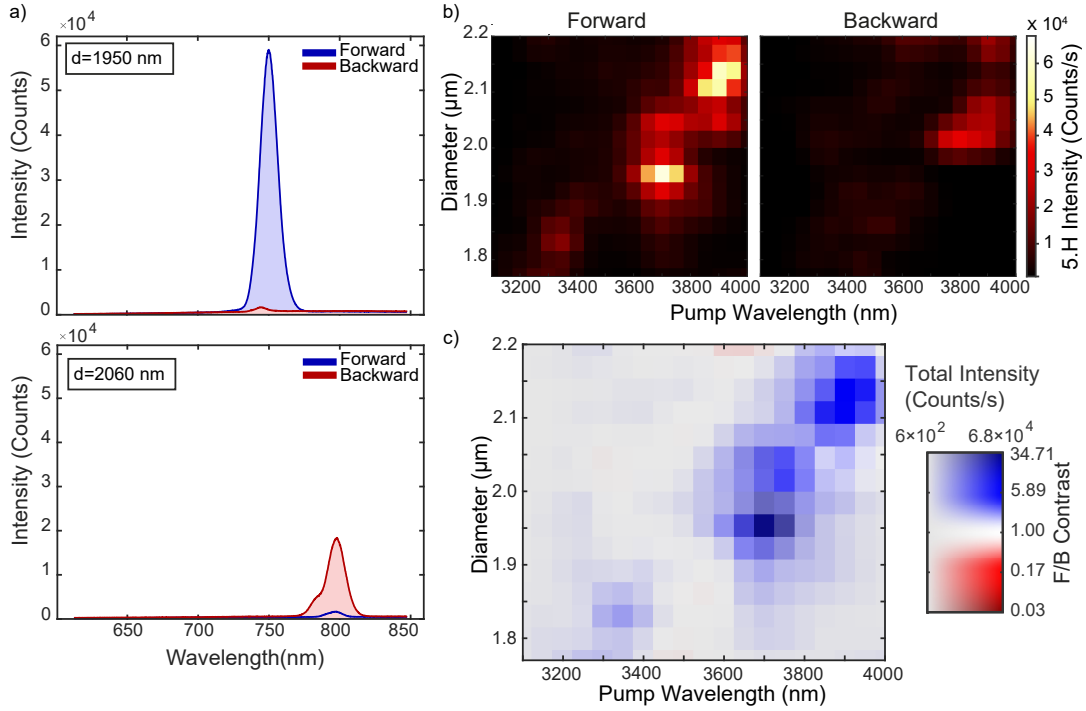


FIGURE 5.4: Measured fifth-harmonic generation (5HG) response of Si-SiN nanopillars. (a) Representative 5HG spectra for two nanopillars under forward (F) and backward (B) illumination. For a nanopillar with a smaller diameter, 5HG is enhanced under forward illumination, whereas a larger nanopillar exhibits stronger emission under backward illumination, demonstrating direction-dependent nonlinear response. (b) Measured 5HG intensity as a function of pump wavelength and nanopillar diameter for forward and backward illumination. Distinct regions of maximum 5HG intensity are observed for the two illumination directions, indicating different resonant excitation conditions. (c) Forward-to-backward contrast map, calculated from the ratio of the measured 5HG intensities in (b), highlighting regions of enhanced direction-dependent nonlinear generation.

frontside and backside illumination. The contrast map reveals three distinct regimes. First, at shorter pump wavelengths and larger resonator diameters, backside illumination results in stronger THG emission, corresponding to regions of low F/B contrast. Second, for larger excitation wavelengths, a pronounced region of enhanced frontside THG emerges, coinciding with the resonant enhancement regime predicted by simulations. Finally, regions of weak THG emission for both excitation directions exhibit low contrast due to the absence of efficient resonant excitation. The diagonal trajectory of the observed features across the parameter space reflects the geometric scaling of the resonant mode. This demonstrates that the direction-dependent nonlinear response can be systematically engineered through resonator geometry and excitation wavelengths. An analogous analysis is performed for the fifth harmonic generation, with the results summarized in Figure 5.4.

Panel a compares the spectra for two different nanopillars. For a nanopillar

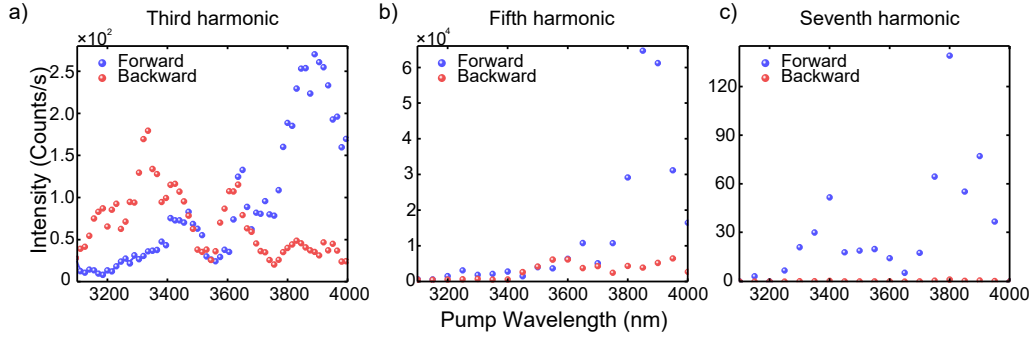


FIGURE 5.5: Comparison of third-, fifth-, and seventh-order harmonic signal under forward and backward illumination.

diameter of $d = 1950$ nm excited at $\lambda = 3700$ nm, the fifth-harmonic signal reaches its maximum under frontside illumination, yielding an F/B contrast of approximately 55. Conversely, for $d = 2060$ nm and $\lambda = 3900$ nm, the strongest signal is observed under backside excitation, corresponding to a B/F contrast of approximately 13. Panel b shows the measured signal for forward and backward illumination as a function of pump wavelength and nanopillar diameter, from which the F/B contrast map is calculated. Regions where the signal generated under forward or backward illumination is dominant can be identified, and the resulting contrast map demonstrates pronounced features similar to the THG case. In comparison to the THG signal, the highest observed signal is red-shifted towards longer pump wavelengths, but still a pronounced contrast is observed in the resonant region for pump wavelengths of 3500 nm to 4000 nm. The shift in pump wavelength observed for the largest 3. and 5. harmonic signals could be attributed to the different position of Mie resonances at the third- and fifth-harmonic wavelengths.

Figure 5.5 compares the third, fifth, and seventh harmonic signals for a nanopillar with a diameter of $d = 2100$ nm as a function of the pump wavelength. Only the third-harmonic signal exhibits well-defined maxima for both forward and backward illumination. For higher-order harmonics, the backward signal is weak so the F/B contrast is dominated by the near-absence of the backward signal rather than by a spectral shift of the maximum signal. The strongest signal for forward illumination is observed for a pump wavelength of 3900 nm, while for backward illumination the strongest signal is observed at 3350 nm. For the fifth and seventh harmonics, the maximum for the strongest forward signal coincides with that of the THG signal at ≈ 3900 nm. In contrast to the THG signal, no 5. harmonic signal under backward illumination is observed for pump wavelengths of 3100 nm to 3400 nm and only a weak signal is observed for larger pump wavelengths, indicating a shift towards longer pump

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wavelengths for the maximum contrast. The maximum seventh harmonic signal under forward illumination is observed for a pump wavelength of 3800 nm and a second local maximum is observed around 3400 nm. Despite an integration time of 120 s, no measurable seventh-harmonic signal was detected under backside illumination.

5.3 Conclusion and Outlook

In this work, we demonstrated nonlinear high-harmonic generation in individual hybrid silicon-silicon nitride nanopillars supporting Mie-type resonances. These structures enable efficient generation of odd-order harmonics up to the seventh order under mid-infrared excitation. Importantly, the harmonic emission is strongly dependent on the illumination direction. This asymmetric behaviour arises from geometry-induced bianisotropic mode coupling, which leads to different near-field enhancement under frontside and backside excitation. As a result, both the efficiency of harmonic generation and the spectral position of maximum nonlinear conversion can be controlled via resonator geometry and excitation wavelength. Compared with conventional approaches relying on bulk materials or extended metasurfaces, the presented approach demonstrates that a single subwavelength resonator can already provide sufficient field confinement to generate a measurable high-order nonlinear signal. The observed dependence of the harmonic signal strength on resonant conditions confirms that the nonlinear conversion is governed by Mie resonances and the field enhancement they provide, rather than by bulk propagation.

Having demonstrated the generation of high harmonics in an individual resonator, the next natural step would be to transition from single pillars back to periodic metasurfaces. This would allow additional control over the spatial phase, amplitude, and polarization of the generated fields. This would enable functional nonlinear optical components such as beam-steering elements, metalenses, or metaholograms.

An important next step is improving the robustness of the nanopillar. The present results indicate that illumination at elevated excitation power results in thermal damage to the sample. Addressing this issue would be beneficial, as higher excitation powers are required to observe even higher-order harmonics. The current deposition process based on plasma-enhanced chemical vapour deposition offers several opportunities for optimization. Increasing the deposition temperature may yield a thermally more stable material. Additionally, adjusting the precursor flow rates may modify the hydrogen incorporation into the Si layer or increase the nitrogen content in SiN. Alternatively, replacing the current materials with more thermally stable alternatives may further enhance the robustness of the nanopillar.

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