



Spatial and temporal light shaping with metasurfaces

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Abstract

This work looks into metasurfaces with three distinct applications.

Chapter 3 covers temporal shaping of laser pulses. By facilitating three successive meta-surface interactions on a single glass substrate by means of reflective surfaces, a miniature 4f pulse shaper is realized. After introducing angular dispersion, different dispersions are applied to incident laser pulses by encoding of wavelength dependent phase information on the second metasurface interaction. The dispersion properties of the laser pulse are experimentally characterized via Second Harmonic Generation Frequency Resolved Optical Gating measurements (SHG FROG).

In chapter 4, the relation between the rotational symmetry of nano-particles to the surrounding lattice symmetry by use of the nonlinear geometrical phase is explored. A model to describe the allowed phase terms is introduced and experimentally verified by various rotationally symmetry meta-atoms placed on a silver mirror in square or hexagonal lattice configuration.

Lastly, chapter 5 covers non-reciprocal metasurfaces for direction dependent hologram generation. By realization of a plasmonic three layer system with intermediate polarization filtering, three different holograms are generated by different input-output combinations of linear polarizations. Depending on the direction of incidence, the required combination of linear polarizations to retrieve a specific hologram changes.

Kurzfassung

Diese Arbeit beschäftigt sich mit Metaoberflächen in drei unterschiedlichen Anwendungen.

Kapitel 3 befasst sich mit der zeitlichen Formung von Lichtpulsen. Durch drei sukzessive Metaoberflächeninteraktionen auf einem einzigen Glassubstrat, ermöglicht durch reflektive Oberflächen, wird ein miniaturisierter 4f Pulsformer realisiert. Nach Erzeugung von Winkeldispersion werden durch wellenlängenabhängige Phaseninformation unterschiedliche Dispersionen in Laserpulsen erzeugt. Diese Dispersion wird experimentell durch Second Harmonic Generation Frequency Resolved Optical Gating Messungen (SHG FROG) charakterisiert.

In Kapitel 4 wird unter Verwendung der nichtlinearen geometrischen Phase der Zusammenhang zwischen der Rotationssymmetrie von Nanostrukturen zur umliegenden Gittersymmetrie untersucht. Ein Modell zur Beschreibung der erlaubten Phasenterme wird vorgestellt und experimentell durch verschiedenen rotationssymmetrische Metaatome, die auf einem Silberspiegel platziert sind, in quadratischer und hexagonaler Gitteranordnung verifiziert.

Zuletzt befasst sich Kapitel 5 mit nicht-reziproken Metaoberflächen zur Erzeugung von richtungsabhängigen Hologrammen. Durch ein plasmonisches Dreilagensystem mit intermediärer Polarisationsfilterung werden drei Kombinationen aus einfallender und detektierter Polarisation verwendet, um Hologramme zu erzeugen. Die dafür benötigte Polarisationskombination unterscheidet sich hierbei, wenn das Dreilagensystem in Vor- oder Rückwärtsrichtung beleuchtet wird.

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In the past century, technological advancement led to an astonishing miniaturization of computational components. Machines that initially required an entire room to fit severely shrank in size while leaving past generations completely in the dust regarding their performance. To support the increasing demand for data traffic that developed in parallel with computational capability, previous transoceanic telephone cables were replaced by optical fibers, allowing for operating at telecommunication bands. The advantages were way lower signal degradation, which directly boosts latency as the signal requires fewer repeaters, in addition to a much larger bandwidth.

While signal transmission over vast distances is covered by optical fibers, data processing and storage is still performed electronically. This naturally introduces an interface where optical signals need to be transformed to electrical signals and vice versa. This so called electro-optical bottleneck [1] introduces latency through repeated conversions and consume large amounts of energy [2]. In order to overcome this limitation, current research is interested in integrated photonics and optical computing architecture that allows to convert more parts of the data processing chain to the optical domain. In order to keep up with already established infrastructure, these optical components need to undergo the same evolution as their electronic counterparts; towards high efficiency and small footprints.

While it is certainly one enticing driving force, it is not just the computation architecture that benefits from miniaturized optical setups. The benefit extends to cameras, LiDAR, portable laser systems, optical neural networks, and many more. However, conventional optical elements, such as lenses or waveplates, rely on phase accumulation across macroscopic distances. Combine this with e.g. multi-lens systems that require achromatic performance or dispersion effects and these setups quickly get bulky, heavy and expensive. An early attempt to decouple the optical function of a component from its bulk volume is the Fresnel lens, invented to facilitate collimating optics in light houses [3]. But for truly compact optical components, conventional fabrication methods do not seem to suffice and require a change in fabrication platform.

Luckily, with decades of experience in fabricating electronic circuits, we stand before a mature nanofabrication architecture that heavily encourages exploration of the so called flat-optics regime. Over the past two decades, a class of materials has emerged that can fill the role of miniaturized optical components. Depending on the design, these materials have shown to exhibit all kinds of optical properties and are broadly classified as “metamaterials”.

Metamaterial

/ˌmɛtəˈmætiəriəl/

noun

A synthetic composite material engineered to display properties not usually found in natural materials. [4]

In general, metamaterials specify as artificial 3D structures whose functionality extends beyond the bulk material properties that they are constructed from. Most of these materials focus on the manipulation of waves, but they are by no means limited to only the optical regime. While metamaterials address electromagnetic waves, ranging from radiowaves up to UV light, they also extend to applications concerning acoustic waves and even seismic waves. In that regard, metamaterials have established themselves at the absolute pinnacle of scattering control in the form of amplitude and phase manipulation. Less prevalent applications include mechanical and thermal metamaterials.

Due to the predominantly planar nature of existing nanofabrication technologies, many metamaterial concepts are naturally translated to 2D implementations. Categorized as metasurfaces, these materials often consist of 2D arrays of plasmonic or dielectric metaatoms with subwavelength dimensions and periodicity, placed on a dielectric substrate. For the most part, individual metaatoms are designed to exhibit certain transmission/reflection properties as well as phase manipulation of certain frequencies or polarization states. Depending on their planar arrangement, a variety of applications become accessible.

In the radiowave regime, metasurfaces are mainly considered to bend, absorb or enhance signals. Special attention is paid to their expected impact on the sixth generation of mobile communication technology (6G) [5]. By dynamically modulating these materials, such reconfigurable intelligent surfaces (RISs) are expected to enable large and energy efficient data transfer. As 6G is considered for higher frequency bands that are however prone to absorption, beam steering metasurfaces are also considered to be used as “smart mirrors” that efficiently guide cellular signals around obstacles [6] or provide frequency conversion functionality that bridge the gap between wide range fiber-optical communication and free space signal transmission [7].

With respect to the control of electromagnetic waves from NIR up to UV radiation, metasurfaces have been used to realize all kinds of applications: beam-steering [8], holography [9], imaging [10], nonlinear frequency generation [11], and many more. A lot of the realized components are already up to par with their bulky counterparts. While early metasurface research primarily focused on replicating the functionality of individual optical components, current research shows an increasing interest in complete, compact optical systems. In part, research steers towards convenient on-chip integration that is compatible with current CMOS-fabrication standards [12] while other applications facilitate miniaturization of e.g. LiDAR [13] or AR systems [14]. With recent advances in computational design methods, inverse-design based structures allow for increasingly complex functionalities and are gravitating towards optical neural networks [15].

The following work contributes to this development by investigating metasurface-based approaches for compact pulse shaping, beam steering in combination with nonlinear optical generation, and nonreciprocal wave manipulation. Together, these topics address light's temporal, spatial, frequency and directional properties via metasurfaces.

2.1 Metals

2.1.1 Optical properties of gold and silver

Metals cover more than two thirds of elements in the periodic table and along their shiny appearance, other characteristic properties are good ductility as well as high electric conductivity and often good thermal conductivity [16]. In contrast to dielectric materials, some electrons in metals tend to be weakly bound to the positive ions of the bulk material. This allows for easy induction of electrical currents when an external electric field is applied and influences the dielectric function $\epsilon(\omega) = \epsilon_0 \epsilon_r(\omega)$ of the system. ϵ_0 corresponds to the vacuum permittivity and $\epsilon_r(\omega)$ is the relative permittivity.

In the Drude-model, the weakly bound electrons can be approximated as a free electron gas against a fixed background of positive metal ions. In order to derive an expression for the relative permittivity $\epsilon_r(\omega)$ under these conditions, one considers a harmonic external driving force and constructs the following equation of motion for the electrons [17]:

$$m\ddot{x} + m\gamma\dot{x} = -eE(t) \quad (2.1)$$

$\dot{x}$ and $\ddot{x}$ are the first and second time derivative of the electron's displacement x , respectively. γ is the damping factor of the system, e the elementary charge and m the electron's optical mass. $E(t) = E_0 e^{-i\omega t}$ is the harmonic time dependence of the applied electric field. This equation of motion contains the assumption, that the electron movement is only influenced by collisions with the positively charged metal ions, neglecting other factors like Coloumb interaction between electron-electron interactions in between collisions. The average time between two collisions is given by $\tau = 1/\gamma$ and is also called the relaxation time. At room temperature weakly bound electrons in metals have a relaxation time in the order of $\tau = 10^{-14}$ s [18], whereas lattice impurities as well as lattice vibrations (phonons) tend to decrease this relaxation time [19]. As an example, at temperatures of $T = [100, 0, -196]^\circ\text{C}$ gold shows relaxation times of $\tau = [2.1, 3, 12] \cdot 10^{-14}$ s, respectively [16]. Note that equation 2.1 does not contain a restoring force and the free electrons are only driven by the external field.

In order to connect this expression to the dielectric function, one relates the sum of the displaced electrons to the induced polarization as such:

$$P = -Nex = \epsilon_0(\epsilon_r - 1)E. \quad (2.2)$$

N describes the electron density, as the total polarization scales with the amount and distance of displaced charges. Typically, electron densities of metals are in the range of $N = 5 \cdot 10^{22} / \text{cm}^3$ [16]. By solving equation 2.1 for $x(t)$ and connecting it to the induced polarization P , the dielectric function eventually yields

$$\epsilon_r = \epsilon_\infty - \frac{\omega_p^2}{\omega^2 + i\gamma\omega} . \quad (2.3)$$

$\omega_p = \sqrt{\frac{Ne^2}{\epsilon_0 m}}$ is the plasma frequency of the electron gas. ϵ_∞ corresponds to the permittivity when ω goes to infinity and is related to the impact of the bound electrons from the positive ions of the metal (background material). Note that the Drude model itself gives $\epsilon_\infty = 1$, corresponding to immobile, non-polarizable bound electrons in the lattice. In addition, ϵ connects to the complex refractive index $\tilde{n}$ via:

$$\epsilon_r = \tilde{n}^2 = (n + i\kappa)^2 . \quad (2.4)$$

With n as the real-valued refractive index and κ as the absorption-/extinction coefficient. The electric field propagation behavior of an electromagnetic wave inside such a medium is governed by the Helmholtz equation

$$-\nabla^2 E = k^2 E , \quad k = \omega \sqrt{\mu\epsilon} . \quad (2.5)$$

$\mu = \mu_0 \mu_r$ is the permeability of the material, while we assume $\mu_r = 1$ here. Looking at $E = E_0 e^{ikr}$, we can see that for real, positive ϵ_r , i.e. at large frequencies, the electric field keeps its oscillatory behavior inside the medium, meaning that transmission inside the system is allowed for $\omega > \omega_p$. On the other hand, real and negative ϵ_r , i.e. small frequencies, cause the argument of the exponential function to become real and negative valued, signifying rapid damping of the wave as it propagates into the medium when $\omega < \omega_p$. An electric field amplitude that penetrates into the material quickly diminishes over a few tens of nanometers with the $\frac{1}{e}$ -field amplitude factor marking the so called skin layer. For gold/silver at a wavelength of 600 nm, this skin layer depth is 31 nm/24 nm, respectively [17]. As a consequence, frequencies below the plasma frequency are strongly reflected by metals. As an example, the plasma frequency of silver correspond to 328 nm [20] and is found between 137 nm [20, 21] - 147 nm [21] for gold. Note that the 137 nm are not explicitly stated by Johnson and Christy, but are extracted from their provided data by Olmon et al.

The provided plasma frequencies might surprise; if frequencies below ω_p are to be reflected, why does gold not reflect the entire visible spectrum, akin to silver? What gives gold its characteristic color are the interband transitions of the material and these are not captured by the Drude model. However, below both the plasma frequency and the interband transition, the Drude model is sufficient to describe the conductive properties of metals, typically ranging from the near-infrared to the microwave regime. Gold has a strong interband transition around 500 nm, enabling excitation of the electrons in the d-orbital to the sp-orbital. The corresponding absorption reaching up till 680 nm and confines the

validity of the Drude model to wavelengths above 680 nm, instead of the plasma frequency [21]. These resonance frequencies are considered in the Drude-Lorentz model and extend the equation of motion of the electrons to [18]:

$$m\ddot{x} + m\gamma\dot{x} + m\omega_0^2x = -eE(t) . \quad (2.6)$$

The added term now introduces a restoring force given by the bound electrons of the lattice ions, featuring a resonance frequency ω_0 . The resulting ϵ_r then shows an additional $-\omega_0^2$ in its denominator. In the case that only the plasma frequency dependence is observed, this resonance frequency is equal to zero (see equation 2.3). For a more accurate description that considers multiple resonance frequencies, the relative permittivity of a bulk metal gives [22]

$$\epsilon_r = \epsilon_\infty - \sum_i \left(\frac{\omega_{p,i}^2 A_i}{\omega^2 - \omega_i^2 + i\gamma_i\omega} \right) . \quad (2.7)$$

A_i and γ_i refer to the amplitude and damping of the i 'th resonance. Note that the plasma frequency is also term dependent, as it captures the density of the electrons as well as their effective mass considered for a respective resonance. Figure 2.1 shows a dielectric function for arbitrarily chosen values, featuring a plasma frequency of 1 and two additional resonances at frequencies $\omega = 2$ and $\omega = 4$ with amplitudes of 2 and 4, respectively. γ is fixed at 0.05 for all resonances.

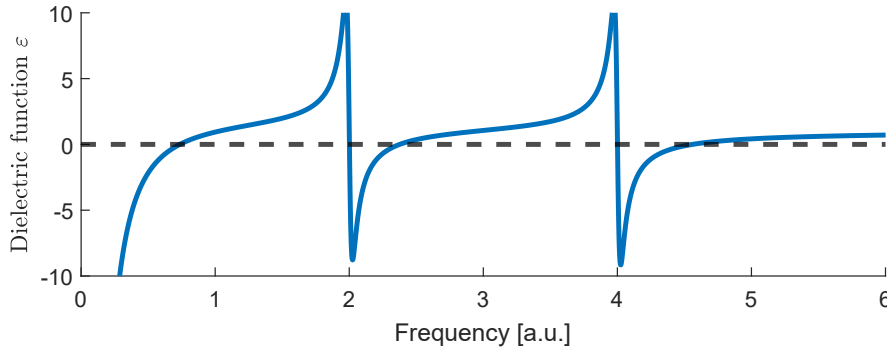


Figure 2.1: Dielectric function corresponding to the Drude-Lorentz model with plasma frequency $\omega_p = 1$ as well as two resonance frequencies at $\omega = 2$ and $\omega = 4$.

This work mainly uses gold and silver as metals for the presented metasurfaces. The refractive index values of these metals that were used in the following chapters are taken from P. B. Johnson and R. W. Christy, shown in figure 2.2 for a wavelength range from 350 nm - 1350 nm [20]. The applications for these different metals are limited to wavelengths above 760 nm in the case of gold, and above 400 nm in the case of silver.

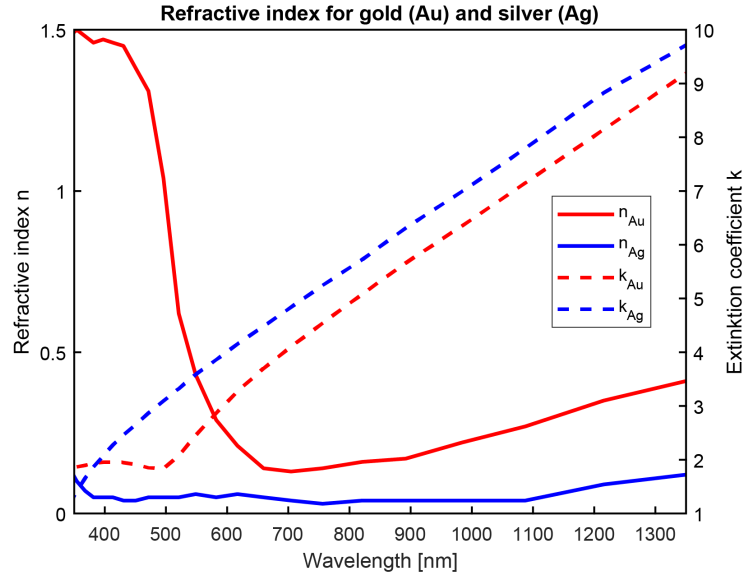


Figure 2.2: Refractive index and extinction coefficient for gold and silver for a wavelength range from 350 nm - 1350 nm provided by Johnson and Christy [20].

2.1.2 Optical properties of metal nano-particles

The plasma frequency refers to a collective oscillation of a cloud of electrons (volume plasmons) and represents a longitudinal wave in the bulk material. As a consequence of its oscillation direction, it can not be excited with an incident transversal electromagnetic wave [18]. As of now, the presented optical behavior does not give a lot to work with. Volume plasmons can not be coupled to using light and the resonance frequencies considered in the Lorentz-extension of the Drude model are fixed bulk properties of the material.

For this reason, metasurfaces make use of sub-wavelength plasmonic nano-particles. In the form of localized surface plasmon polaritons (LSPPs), collective electron oscillations on the physical boundaries of metal nano-particles allow for resonant excitation of nano-particles. These modes can be excited by light fields and facilitate surface currents akin to an antenna. Hence, the resonance frequency of these structures is heavily dependent on the geometrical extend of the particles. Because of the finite extend of the particle, the displaced charges induce a dipole moment p

$$p = \epsilon_0 \epsilon_m \alpha E_0 . \quad (2.8)$$

ϵ_m gives the permittivity of the surrounding dielectric material and α corresponds to the polarizability of the plasmonic particle. One of the few analytically solvable examples of plasmonic particles placed inside a dielectric medium is a metallic sphere. By choosing a diameter d much smaller than the incident wavelength, one can approximate a constant phase of the applied electric field amplitude across the particle, therefore allowing to separate the time dependence of the problem. The polarizability of such a system is given

by [18]

$$\alpha = 4\pi R^3 \frac{\epsilon - \epsilon_m}{\epsilon + 2\epsilon_m} \quad (2.9)$$

with R as the radius of the nano-particle. This expression allows convenient connection between the macroscopic dielectric function ϵ of the metallic/plasmonic material and its microscopic properties. Note that a resonant enhancement occurs when the denominator is minimized. For a slowly varying imaginary part of $\epsilon(\omega)$ this occurs at $\text{Re}[\epsilon(\omega)] = -2\epsilon_m$, which is called the Fröhlich condition [18]. This requirement is typically fulfilled near resonances of the corresponding bulk-metal. When the dielectric constant of the host medium is increased, i.e. its refractive index increases, the Fröhlich condition is fulfilled at smaller frequencies, meaning that the resonant behavior is red-shifted. An example of this behavior is illustrated on the dielectric function shown in figure 2.1 and can be seen in figure 2.3 for dielectric constants of $\epsilon_m = [1, 2, 3]$.

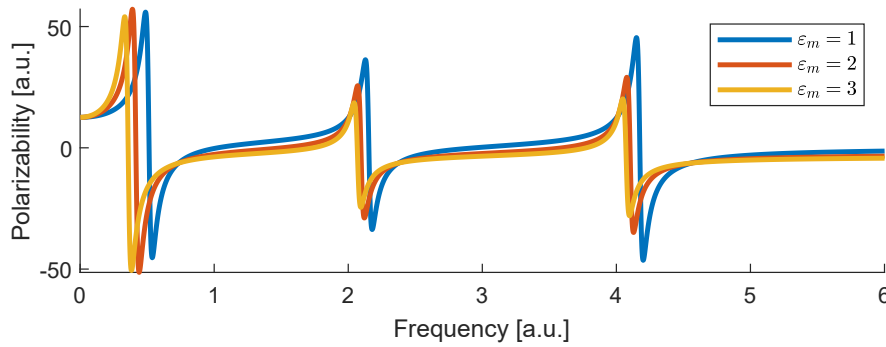


Figure 2.3: Polarizability of a sphere with radius $R = 1$ for different dielectric constants $\epsilon_m = [1, 2, 3]$. As the dielectric constant of the host medium is increased, the maximum polarizability redshifts towards more negative ϵ .

This dependence of the ambient environment makes plasmonic nano-particles popular candidates for optical sensing. For a spherical nano-particle of subwavelength dimensions, the presented polarizability does give a good understanding of its optical properties. While we now see, that the surrounding refractive index can cause resonance shifts in the LSPPs, the provided expression in equation 2.10 does not yet show why the resonance wavelength is dependent on the size of the particle. The provided radius of the sphere is only a global scaling factor and besides that, we only made the assumption, that the nano-particle is smaller than the incident wavelength. The size-dependence, although intuitively comprehensible when compared to charge oscillation in antennas, is better understood, when abstracting the above equation to an ellipsoid with three different major axes a_1, a_2, a_3 . In this case, the polarizability evaluates to [18]

$$\alpha = 4\pi a_1 a_2 a_3 \frac{\epsilon - \epsilon_m}{3\epsilon_m + 3L_i(\epsilon - \epsilon_m)} \quad (2.10)$$

While I will not cover it in detail, L_i represents a geometrical factor between 0 and 1,

corresponding to the i 'th major axis of the ellipsoid. In addition, the sum over these geometrical factors evaluates to $\sum_i L_i = 1$. We can see, that it influences the denominator of the polarizability and can therefore induce a resonance shift. In the case of the sphere, the three geometrical factors evaluate to $1/3$, respectively. For structures that are elongated along one major axis relative to the rest, one finds that the corresponding geometrical factor decreases. Taking a geometrical factor of $1/6$, the denominator evaluates to $\epsilon + 2.5\epsilon_m$. As a consequence, ϵ requires a greater negative magnitude to fulfill the Fröhlich condition. Looking at figure 2.1, this causes a resonance redshift, as one expects from an antenna with increasing length.

Overall, by fabricating small nano-particles the optical response of the underlying material can be tampered with by restricting the possible electron displacement and generally, for more complex shaped structures numerical calculations are used to determine the corresponding optical properties. As a consequence of their shape and host medium, the resonance frequencies and scattering of these particles can be engineered.

As a downside to plasmonic structures, collisions with the lattice ions dissipate the kinetic energy of the LSPs, causing Ohmic losses and heating the structures. Ultimately, this property is the limiting factor in their laser induced damage threshold - a shortcoming that is not shared by their dielectric counterpart. Additionally, as the nano-particles resemble a small antenna, the radiated signal from the induced dipole radiates in both halves of free space, making the total transmission after resonant excitation less efficient. And lastly, note that the phase between the driving field and the electrons oscillations reaches a maximum of π when passing the resonances towards larger frequencies, with a $\pi/2$ phase delay at resonance; a full 2π -phase in the presented form is not possible, which limits wavefront shaping capabilities. However, solutions to the latter issue will be introduced as they are needed in the following chapters.

2.2 Dielectrics

2.2.1 Optical properties of amorphous silicon

The only dielectric material that constitutes nano-antennas within the confines of the following chapters is amorphous silicon (a-Si:H). Deposited via plasma enhanced chemical vapor deposition (PECVD) using argon (Ar) diluted silan gas (SiH_4) at 300°C , the process does not include any doping, leaving the resulting silicon non-conducting at room temperature. In general, the dielectric function can also be described by the Drude-Lorentz model shown in equation 2.7. The main difference to metals is that the free electron contribution in non-conduction materials evaluates to zero.

The dielectric function/refractive index of the deposited “bulk” silicon is highly dependent on the fabrication parameters during PECVD. As a consequence, the linear optical properties are not taken from literature but are instead measured via ellipsometry (the measurement was conducted by Rebecca Aschwanden). Figure 2.4 shows the refractive

index and extinction coefficient of a deposited amorphous silicon thin film (thickness 600 nm) that will be used in the simulations of this work.

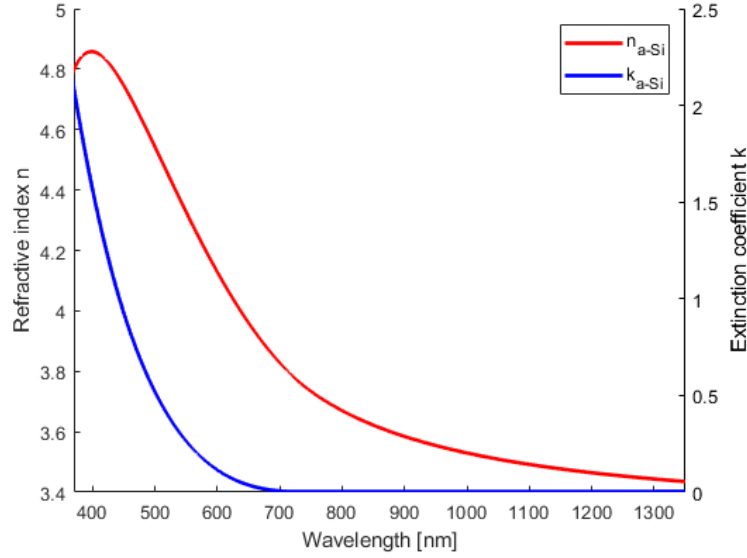


Figure 2.4: Refractive index and extinction coefficient for PECVD-deposited amorphous silicon for a wavelength range from 350 nm - 1350 nm, measured via ellipsometry by Rebecca Aschwanden.

The deposited films feature a large refractive index and quite low losses at wavelengths above 700 nm, falling within a factor of two of reported literature on the extinction coefficient of crystalline silicon [23]. Often, due to the disordered nature of the silicon, momentum conservation restrictions in the material relax, turning it from an otherwise indirect bandgap material to a direct bandgap semiconductor. The extinction coefficients can differ wildly depending on the fabrication procedure and the intended use; the data provided by Pierce et al. shows losses that are a factor of 30 larger than the measured values here [24] which are desirable when using silicon in photo-detecting devices. As building blocks for metasurfaces, a high refractive index and low losses are however an ideal place to be in.

2.2.2 Optical properties of dielectric nano-particles

The large refractive index and relatively low losses of the deposited a-Si makes it ideal to function as nano-resonators in metasurfaces. In contrast to metals that mainly interact with the electric field of light, dielectric structures extend this regime to an equal capability to magnetic fields. While metals only cover a π -phase shift due to one present resonance driven by the electric field, the addition of magnetic modes enables a full 2π -phase coverage in dielectric nano-structures. In addition, dielectric nano-particles do not suffer from

heating due to Ohmic losses and therefore exhibit a much higher laser induced damage threshold. While there are different ways that can be exploited to resonantly excite dielectric nano-structures, here I will briefly cover the most widely used: Mie-like resonances.

For dielectric particles that are much smaller than the wavelength of incident light, the polarizability is also governed by equation 2.10. In this quasi-static regime, the induced polarization is primarily dipolar.

In the case that the optical size of the nano-particle is roughly equal the incident wavelength, scattering of incident plane waves from a spherical, non-absorbing and non-magnetic particle in a homogeneous background medium are described via the Mie theory [25]. The optical extend of the nano-particle in combination with its high refractive index contrast between to the surrounding medium allows to confine both electric and magnetic fields down to subwavelength scales. The fields inside and outside of the sphere are developed into an infinite series of vector spherical harmonics, capturing the angular and radial behavior of the scattered waves. Depending on the shape, size and refractive index of the nano-particle as well as the incident wavelength, various electromagnetic multipole modes - these covering dipoles, quadrupoles etc. - contribute to the scattering of said particle. The scattering efficiency, a measure for the interaction strength between a particle and the incident wavelength, of a small dielectric sphere with radius R is defined as [25]

$$Q_{\text{sca}} = \frac{2}{k^2 R^2} \sum_{n=1}^{\infty} (2n+1) (|a_n|^2 + |b_n|^2) . \quad (2.11)$$

k is the wavenumber, n the evaluated order of the mode and a_n/b_n correspond to the Mie coefficients of n -th order for electric/magnetic fields, respectively. Enforcing continuity on the tangential field components at the sphere's boundary allows calculation of these Mie coefficients. a_1 corresponds to an electric dipole, whereas b_2 represents a magnetic quadrupole etc. A particular resonance is marked by a maximized Mie coefficient and one often finds that one particular field contribution dominates the scattering behavior of a particle at one particular wavelength. Besides spherical or cylindrical particles, more complex structures often rely on numerical calculations to determine their scattering behavior.

More sophisticated designs tailor the contribution of multiple modes to engineer the optical behavior of nano-particles. In Huygens metasurfaces, balancing of electric and magnetic dipoles in a nano-resonator is utilized to suppress back scattering from the structures and allow almost perfect transmission through the structures [26]. Other excitation like quasi-BIC modes [27] or Fano-resonances [28] can involve coupling of multiple modes to realize huge quality factors that yield strong field enhancement useful for nonlinear optical applications.

Compact metasurface-based pulse shaping

3

3.1 Introduction

Ultrashort laser pulses, these being of picosecond time scales or less, play an increasingly important role in today's research and cover a wide range of applications. Typically, the modes inside a laser cavity all have random phases with respect to each other, and can even be subject to thermal fluctuations, ultimately resulting in a near constant output power. However, by controlling the relative phases of the longitudinal modes in a laser cavity (mode-locking) across a large bandwidth, a pulse train can be created through constructive interference. These high power bursts with times scales down to a few femtoseconds, have established themselves as useful tools across several disciplines.

In 1999, Ahmed Zewail received the nobel prize in chemistry for using ultrashort laser pulses to study chemical reactions on equally short timescales, thus opening the field of femtochemistry. One of the most common techniques born from this field is the pump-probe spectroscopy; by quickly exciting an optical system - such as a molecule - and casting a probe pulse with a well defined temporal separation from the excitation pulse, the time- and population-dynamics of the system can be gauged by observing properties like the delay dependent transmission or atomic bonding dynamics [29]. This measurement protocol requires pulse lengths much shorter than the typical (de-)population time of the system's energy levels. In a similar manner, quantum dots can also be excited by precisely tuning an individual pulse's energy and spectral phase. Ultrashort laser pulses are also highly beneficial for nonlinear optical processes, as these scale in a non-linear fashion with the present electric field amplitude; by concentrating the energy into a small time frame, the conversion efficiency can increase by several orders of magnitude.

Needless to say that some applications heavily benefit from having precise knowledge about the temporal and spectral properties of the laser pulses used in the experiment. I wouldn't necessarily be able to provide a nonlinear conversion efficiency if I didn't know the temporal pulse width used in the nonlinear optical experiments. I also wouldn't learn about the excitation or reaction dynamics of certain molecules if I didn't know about the spectral phase of the laser pulses - let alone optimizing them, as the spectral phase can impact the excitation behavior of a system [30, 31]. Luckily, a literal zoo of measurement techniques have established themselves in order to retrieve the spectral phase and/or temporal shape of laser pulses. These cover FROG (Frequency-Resolved Optical Gating), SPIDER (Spectral Phase Interferometry for Direct Electric-field Reconstruction), TADPOLE (Temporal Analysis by Dispersing a Pair Of Light E-fields), GRENOUILLE (apparently

“frog” in french, GRating-Eliminated No-nonsense Observation of Ultrafast Incident Laser Light E-fields), DOVE (Direct Optical Vector Extraction), MICE (Mutual Interferometric Characterization of independent Electric fields) and SPICE (bugs often found in spices..., Spectral Phase Interferometry for Complex Electric fields). From these, FROG and SPIDER seem to be the most prominent techniques used.

While knowledge about the laser pulse properties is crucial, control over these seems just as important. The necessity of which might arise from a requirement for exotic pulse shapes or simply to compensate material dispersion leading to unwanted pulse broadening.

Since material dispersion can have the unwanted effect of broadening laser pulses, one might think that piecing together different bulk materials with various thicknesses might allow to create custom spectral phases. However, you are often at the mercy of a slowly varying wavelength dependent refractive index in a lot of materials - enough to be a headache in some experimental setups, but not enough to conveniently allow realization of exotic pulse shapes. The main disadvantage here is, that there are little ways of modulating the material dispersion on the bulk level. This is where the advantage of artificial materials, like metasurfaces come into play. In 2021, Ossianer et al. designed a silicon based metasurface to apply a wavelength dependent phase profile for pulse shaping [32]. By adjusting the geometrical dimensions of periodically arranged sub-wavelength silicon cylinders, the mostly linear dispersion relation within an 80 nm bandwidth could be controlled. Their presented design, using 610 nm high silicon nanoposts, is able to compensate the equivalent of ≈ 2 mm of silica glass by introducing anomalous dispersion. This could prove useful if applied to individual optical components by constantly re-compressing the incident laser pulses. However, higher order dispersion contributions are not really accessible - a shortcoming which is shared by pulse compressors that mainly change the second order dispersion responsible for the pulse width. The total dispersion from a single metasurface is ultimately limited by the thickness of the nano antennas. Nevertheless, the realized dispersion per unit length is over 3000 times higher than that of silica glass, showing the potential of dispersion engineered materials.

Another more bulky but way more flexible approach involves the Fourier transform pulse shaping. By using a grating to introduce angular dispersion, subsequent focusing of the diffracted light causes a spatial separation of the used spectrum in the focal plane. By placing a phase mask into the focal plane, one can achieve a wavelength dependent phase modulation. Additional introduction of another lens and grating re-collimates the spectrum into laser pulses with modulated temporal properties. For the most part, spatial light modulators (SLMs) are used as a phase mask, as they allow for adjustable phase profiles. They are already capable of realizing wavelength multiplexed holograms [33] and show update rates of more than 100 kHz [34]. Additionally, the SLM allows to incorporate the grating and lens functionality as well, enabling a more compact design [35]. On the other hand, the disadvantages are high cost and limited pixel size/resolution. While not programmable, metasurfaces have also been used as phase masks in Fourier transform pulse shaping setups [36, 37]. Additionally, they can be optimized for NIR applications which is unfavorable for SLMs that use indium tin oxide (ITO) contacts. Divitt et al. realized a phase

and amplitude modulating metasurface by fabricating cuboid shaped silicon nanoantennas on silica glass to manipulate the phase of the incident spectrum while placing rotated wire-grid polarizers on the opposite side of the glass substrate to achieve amplitude modulation, thereby nicely separating two functionalities with simple structures [38]. As metasurfaces have established themselves as potent flat-optical materials, more and more research is done on compact devices [39, 40]. These applications include a direct combination of metasurfaces with detecting CMOS detectors to realize spectral image sensors via fabrication of bandpass filters [41] to more complex, folded metasurface based devices [42, 43]. In 2018, Faraji-Dana et al. demonstrated a compact folded spectrometer [44]. Their device facilitates successive metasurface interactions on a single piece of glass by using metasurface based gratings in combination with mirror layers on both sides of a glass substrate. Incoming light is spectrally separated within their device across a total of three metasurfaces and coupled out of their device through an aperture, allowing for combination with detecting electronic. This particular publication is also what inspired the realization of a compact metasurface based pulse-shaping device. The following chapter aims to combine the compactness of metasurface based devices with the Fourier transform pulse shaping, allowing for an arbitrary wavelength dependent phase profile.

3.2 Theory

3.2.1 Pulse shaping

In order to generate laser pulses to begin with, the overlapping of monochromatic waves with different wavelengths is required. By putting together either a variety of discrete wavelengths or a continuous distribution of them, the temporal shape of the resulting pulse is determined by the spectral amplitude $A(\omega)$ and the spectral phase $\phi(\omega)$. To determine the electric field $E(t)$ a Fourier transform of the electric field into frequency domain is performed.

$$E(\omega) = A(\omega)e^{i\phi(\omega)} \quad (3.1)$$

$$E(t) = \frac{1}{2\pi} \int_{-\infty}^{+\infty} A(\omega)e^{-(i\omega t - \phi(\omega))} d\omega \quad (3.2)$$

While mathematically, there is an infinite amount of possible pulse-forms one could realize, in optics, often a Gaussian pulse shape is considered in simulations and experiments. In an idealized case, the spectral amplitude of a Gaussian pulse is described as

$$E(\omega) = \sqrt{\frac{\pi}{2 \ln(2)}} \tau \exp \left[-\frac{\tau^2 (\omega - \omega_0)^2}{8 \ln(2)} \right] \quad (3.3)$$

while the spectral phase $\phi(\omega)$ is considered constant. Here, τ is the Full Width at Half Maximum (FWHM) of $E(t)$ and ω_0 is the carrier frequency corresponding to the center

wavelength. The temporal shape of such a spectral distribution is

$$E(t) = \exp \left[-8 \ln(2) \left(\frac{t}{\tau} \right)^2 \right] \quad (3.4)$$

This shape is a consequence of the constructive interference of the entire spectrum around a single point in time - or space - rapidly decreasing in amplitude (and intensity) around said point. As time passes, the point in space where the entire spectrum constructively interferes changes position, resulting in the propagation of the Gaussian pulse. This also means, that the propagation of this pulse is not explicitly dependent on the speed of light c but rather the speed at which the point of constructive interference changes its position in space. In vacuum, where the propagation speed for the entire spectrum is the same (c), the propagation speed of the Gaussian pulse is also c while the temporal shape is constantly maintained. And similarly, if exposed to an optical medium with a constant refractive index n , while the propagation speed of the individual wavelengths as well as the optical pulse is now reduced to $\frac{c}{n}$, the temporal shape still remains the same. As soon as chromatic dispersion - the presence of a wavelength dependent refractive index - is introduced, the propagation speed and the temporal shape are affected as a consequence.

To describe the effect of chromatic dispersion in more detail, we can extend the general frequency-dependent phase $\phi(\omega)$ to a dependency on the spatial coordinate x .

$$\phi(x, \omega) = k(x) \cdot x = \frac{x}{c} \cdot \omega \cdot n(\omega) \quad (3.5)$$

Furthermore, the phase-profile $\phi(\omega)$ can be expressed as a Taylor-series which allows for more convenient separation of the influence of different material-dispersions on the propagation behavior and shape of laser pulses [45].

$$\phi(\omega) = \sum_{m=0}^{\infty} \frac{(\omega - \omega_0)^m}{m!} \cdot \left(\frac{\delta^m}{\delta \omega^m} \phi(\omega) \right)_{\omega=\omega_0} = \sum_{m=0}^{\infty} \frac{(\omega - \omega_0)^m}{m!} D_m \quad (3.6)$$

m corresponds to the order of the Taylor-expansion, ω_0 to the carrier frequency and D_m is the dispersion coefficient [46]. By inserting the formula 3.5 into D_m , we get

$$D_m = \frac{x}{c} \cdot \left(\frac{\delta^m (\omega \cdot n(\omega))}{\delta \omega^m} \right)_{\omega=\omega_0}. \quad (3.7)$$

By evaluating individual dispersion coefficients, different influences can be attributed to them. D_0 represents the *phase* propagation of the carrier frequency and is further expressed as

$$D_0 = \frac{x}{c} \cdot \omega_0 \cdot n_0. \quad (3.8)$$

With $n_0 = n(\omega_0)$. D_1 gives information about the propagation speed (group velocity v_G) of the entire wave-package by giving the delay time t_G of the pulse after propagating through a certain material thickness x . This dispersion coefficient is also often referred to as group

delay (GD).

$$D_1 = \frac{x}{c} \cdot \frac{\delta(\omega \cdot n(\omega))}{\delta\omega} = x \cdot \frac{\delta k}{\delta\omega} = \frac{x}{v_G} = t_G \quad (3.9)$$

The group velocity v_G of the wave package is described as

$$v_G = \frac{d\omega}{dk}. \quad (3.10)$$

D_2 describes the broadening of a laser pulse (depending on the assumed laser pulse shape) and is called group delay dispersion (GDD).

$$D_2 = \frac{x}{c} \cdot \frac{\delta^2(\omega \cdot n(\omega))}{\delta\omega^2} = x \cdot k'' \quad (3.11)$$

Note that GDD refers to the total experienced second order dispersion, while there also exists the group velocity dispersion (GVD) that corresponds to GDD per unit length. This means, that GDD is GVD times the propagated distance. The spectral and temporal shape of a Gaussian pulse without dispersion can be expressed with the time-bandwidth product. This measure uses the temporal τ and spectral λ FWHM for a given pulse. For a time-bandwidth limited Gaussian pulse - a pulse with no applied chromatic dispersion - the time-bandwidth product is $\tau \cdot \lambda \approx 0.441$, allowing for easy conversion from the temporal to the spectral FWHM and vice versa. When exposing such a time-bandwidth-limited Gaussian pulse to a certain amount of GDD, the resulting temporal FWHM $\tau(x)$ can be approximated as [46]

$$\tau(x) = \tau \cdot \sqrt{1 + \left(4 \cdot \ln(2) \cdot \frac{D_2}{\tau^2}\right)^2}. \quad (3.12)$$

Additionally, the carrier frequency $\omega(t)$ of the pulse changes linearly over time. This effect is called spectral chirp.

$$\omega(t) = \frac{\delta(\omega_0 \cdot t)}{\delta t} = \omega_0 + \frac{D_2}{\left(\frac{\tau}{2\sqrt{\ln(2)}}\right)^4 + D_2^2} \cdot t \quad (3.13)$$

$\frac{\tau}{2\sqrt{\ln(2)}}$ corresponds to the $\frac{1}{e}$ -width of the intensity profile. While D_2 has units s^2 , conventionally, the dispersion coefficients D_m are given in units of ps^m or fs^m .

3.2.2 FROG measurement and retrieval

In order to characterize the spectral phase of ultrashort laser pulses, we will use the frequency-resolved optical gating (FROG) method invented by Rick Trebino and Daniel J. Kane [47]. This method resembles an autocorrelation (or auto-spectrogram) measurement enabled through the generation of second harmonic. The optical gating is done with the to-be-characterized pulse whereas the use of a well-known reference pulse is also possible

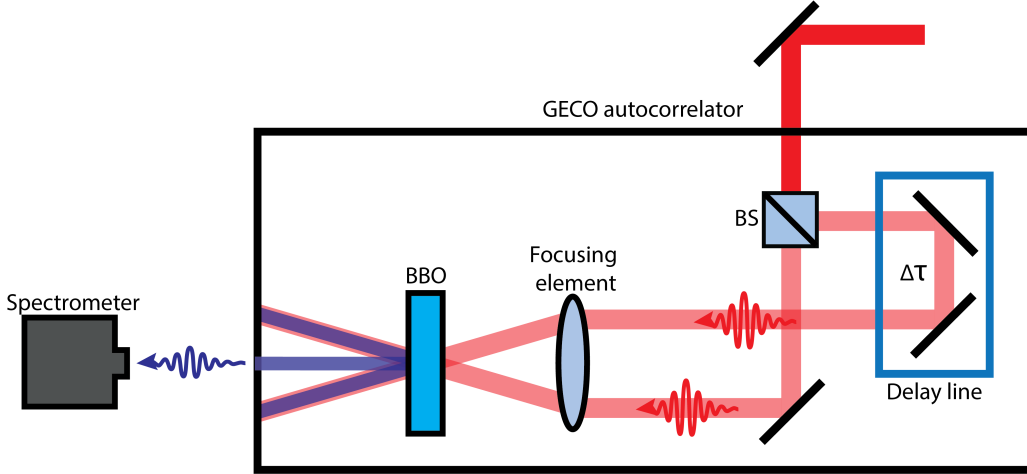


Figure 3.1: Schematic SHG FROG experimental setup. Two beam paths with a relative time delay τ are focused into a nonlinear optical crystal. Depending on the temporal overlap of the two laser pulses, SH is generated and coupled to a spectrometer.

(used in XFROG). A schematic of an SHG FROG setup is shown in figure 3.1. After passing a beam splitter, one beam path experiences a time delay τ via a delay line. Afterwards, both beam paths pass a lens or a concave mirror which focuses them into a nonlinear optical crystal. Inside the nonlinear optical medium, both pulses can generate SH by interacting with themselves, with their initial propagation direction being maintained. Additionally, both pulses can interact with each other - provided that the temporal delay between the two paths is small enough - and generate SH. Since the two incident beams are non-collinear, the propagation direction of any SHG signal is dependent on the wave vector sum of the two contributing beams. This way, the time delay dependent gating can be filtered from the other two SHG signals.

$$\mathbf{k}_3 = \mathbf{k}_1 + \mathbf{k}_2 \quad (3.14)$$

The autocorrelation signal is then coupled to a spectrometer. Thus, the resulting intensity distribution I_{SHG_FROG} (also called a FROG trace) is measured over frequency ω and delay time τ and can be described via the Fourier-transforms of the two input pulses.

$$I_{SHG_FROG}(\omega, t) = \left| \int_{-\infty}^{\infty} E(t)E(t - \tau)e^{-i\omega t} \right|^2 \quad (3.15)$$

Since the FROG trace only captures the spectral intensity and does not directly allow access to the electric field's phase, typically an iterative phase-retrieval algorithm based on generalized projections is used [48, 49]. By alternately projecting on two different conditions, convergence to a solution that fulfills both can be ensured. One condition is that the measured spectral intensity I_{FROG} is proportional to the squared electric field $E_{sig}(\omega, \tau)$ of the SHG signal.

$$I_{FROG} \sim |E_{sig}(\omega, \tau)|^2 \quad (3.16)$$

The other condition connects this SHG signal field to the fundamental fields involved in the measurement, enforcing the second condition

$$E_{sig}(t, \tau) = E(t) \cdot E(t - \tau) \quad (3.17)$$

The implementation of such an algorithm (adapted from Daniel Kanes work [48]) was done by Philip Georgi and the optimization loop of the algorithm is shown in figure 3.2.

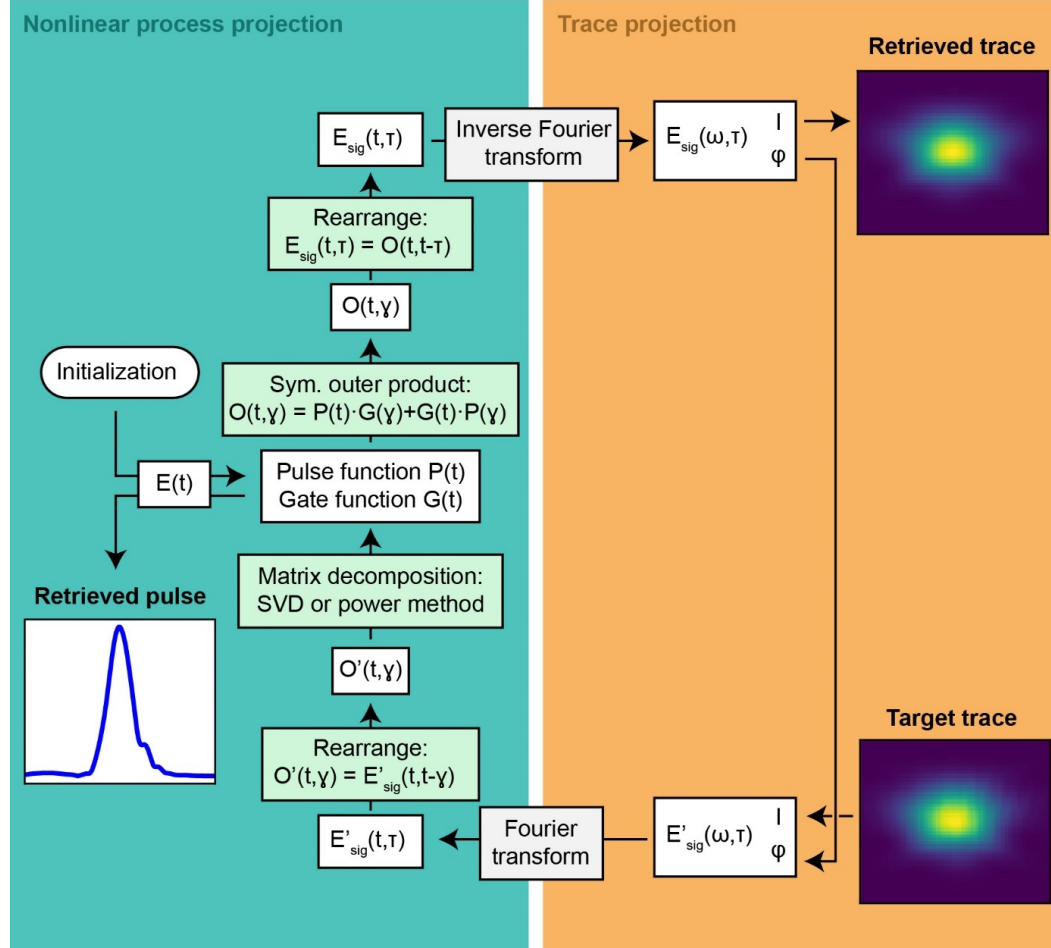


Figure 3.2: Iterative algorithm to retrieve pump field phase information. The signal field $E'_{sig}(t, \tau)$ is rearranged into an outer product $O'(t, \gamma)$ via coordinate transformation. By finding the eigenvalue of this rank-1 matrix, decomposition into a pulse- and gate- function ($P(t), G(t)$) becomes possible. Recalculating the signal field and applying a Fourier-transform retrieves the corresponding FROG trace. Replacing the intensity profile with the measured SHG FROG trace and Fourier-transforming back to the time dependent signal field $E'_{sig}(t, \tau)$ completes an iteration cycle. After convergence, the goal is to retrieve the pulse dispersion from the pulse-/gate- function. [50]

The idea of the algorithm is to decompose the time and delay dependent SHG electric field amplitude $E_{\text{sig}}(t, \tau)$ into a pulse- $P(t)$ and a gate- $G(t)$ function while also enforcing equality of these two fields. Hence, the signal field is generally expressed as

$$E'_{\text{sig}}(t, \tau) = P(t)G(t - \tau) \quad (3.18)$$

For now, let's assume we know the signal field $E'_{\text{sig}}(t, \tau)$ (shown in the bottom left of figure 3.2). We can arrive at the outer product $O'(t, \gamma)$ by transforming the signal fields dependence of τ to $\gamma = t - \tau$. On a discretized time-grid, the outer product can be written as the dyadic vector product of P and G:

$$O(t, \gamma) = P(t) G(\gamma)^T \quad (3.19)$$

An outer product results in a rank-1 matrix and finding an eigenvector can be done somewhat differently than standard matrix operations that otherwise involve formulating a characteristic equation, finding the determinant of the matrix, solving for the eigenvalues and solving a system of linear equations for the eigenvectors. In this case, a rank-1 matrix only has one eigenvalue and the eigenvector directly relates to the relevant pulse-/gate-function. This allows to use the power method (power iteration) to decompose the outer product. Generally, the power method repeatedly multiplies a random, normalized starting-vector onto a matrix M , ultimately converging to the most dominant (largest absolute value) eigenvalue (up to a constant factor) of the matrix. We can find the eigenvector of the pulse- and gate- function from the matrices

$$M_P = OO^T \quad (3.20)$$

$$M_G = O^T O \quad (3.21)$$

Assuming that the algorithm converged, the retrieved P- and G- function can be evaluated to extract the linear fields dispersion. Otherwise, the algorithm proceeds and the signal field $E_{\text{sig}}(t, \tau)$ is reconstructed from the pulse- and gate-function after forming the outer product. The inverse Fourier-transform of this signal field generates the retrieved FROG trace. Afterwards, the trace intensity is replaced with the measured one - making use of condition 1, see equation 3.16 - whereas the calculated phase remains. Afterwards, the signal field $E'_{\text{sig}}(t, \tau)$ is calculated via Fourier-transform and rearranged into the outer product from which the eigenvectors for P and G are acquired via the power method. Note that symmetry between the pulse- and gate- function can be enforced, by redefining the outer product to

$$O_{\text{sym}}(t, \gamma) = P(t)G(\gamma)^T + G(\gamma)P(t)^T \quad (3.22)$$

eventually resulting in equal eigenvectors for the two functions. The algorithm is initialized by guessing the fundamental electric field $E(t)$. As the algorithm cannot distinguish between positive and negative dispersion - due to the nature of the SHG FROG trace - besides a zero-dispersion input field, the algorithm is observed to be robust against the initialization choice. However, whenever a reasonable initialization/educated guess can be made, it tends to speed up the process. Here, we can make use of the trace's marginals for

the initial guess, as proposed by Rana Jafari et al [51]. These being the integral over either the frequency or delay, thus giving the delay marginal $M(\tau)$ or the frequency marginal $M(\omega)$, respectively.

$$M(\omega) = \int_{-\infty}^{+\infty} I_{\text{FROG}}(\omega, \tau) d\tau \quad (3.23)$$

$$M(\tau) = \int_{-\infty}^{+\infty} I_{\text{FROG}}(\omega, \tau) d\omega \quad (3.24)$$

The delay marginal can be understood as the *auto-correlation* of the pulse intensity. Furthermore, the delay marginal's Fourier-transform is equal to the magnitude of the temporal intensity's Fourier-transform squared, thus losing all phase information in the process. The frequency marginal on the other hand is the *auto-convolution* of the pulse spectrum. Hence, the frequency marginal's Fourier-transform is the Fourier-transform of the spectral intensity $S(\omega)$ squared (not taking the magnitude).

$$\mathcal{F}\{M(\omega)\} = \mathcal{F}\{S(\omega)\}^2 = s(t)^2 \quad (3.25)$$

Solving for $s(t)$ gives a clear result up to the sign-ambiguity from the square-root operation

$$s(t)_{\pm} = \pm \sqrt{\mathcal{F}\{M(\omega)\}} \quad (3.26)$$

In principle, every point in time has two possible solutions, however, applying the Paley-Wiener Theorem, essentially using the fact that the pulse spectrum and its derivatives are continuous functions, reduces the possible solutions to two branches, namely $s_+(t)$ and $s_-(t)$. From here, $S(\omega)$ can be calculated via inverse Fourier-transform which allows to get the amplitude of the spectral field $E(\omega)$. This leaves only the spectral field $E(\omega)$ unknown which can be randomly initialized.

3.3 Design

3.3.1 General device layout

A pulse shaping device or a pulse compressor typically consists of a pair of prisms or gratings that introduce angular dispersion in order to separate the different frequencies in a given laser pulse. The spectral bandwidth, the desired spectral resolution as well as the systems angular dispersion dictates the overall size of the device, as a certain amount of propagation length after introduction of the angular dispersion is necessary in order to create suitable spatial dispersion. In this work, the propagation will be confined down to a single glass substrate, allowing for larger propagation length via mirrors on both sides of the substrate. The optical element that is supposed to introduce angular dispersion as well as the phase modulating medium will be replaced by a metasurface; compared to a standard

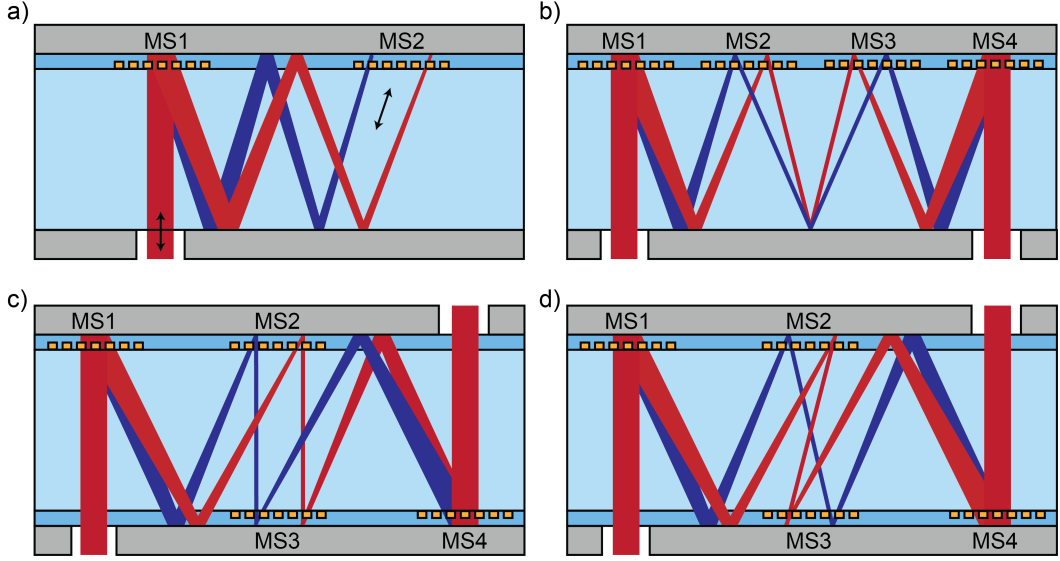


Figure 3.3: Initial ideas for the general layout of the pulse shaper. a) “reflective” design with two metasurfaces and three metasurface interactions in which the shaped pulses come out of the input aperture. b) Symmetric layout with 4 metasurfaces in total and one dedicated input and output aperture. c) Initial design idea using four metasurfaces on both sides of the glass substrate. This design has drawbacks when recombining the beams coming from MS3 onto MS4 as the diffraction angles over wavelength are not identical to the ones introduced on MS2. d) Alternate design of c) using a symmetry point in the middle of the glass-thickness. By swapping the beam positions, MS2 and MS3 can have identical gradient encoding to guide and recombine the different wavelength back onto MS4.

spectrometer, the spatial resolution can be higher since the pixel pitch in commercially available spectrometers is typically in the μm -range whereas a metasurface operating in the visible spectrum tends to have a unit cell periodicity in the order of a few hundred nanometers. Figure 3.3 illustrates some designs that have been initially considered.

The layouts show the beam paths for the shortest and longest wavelength used in the system and illustrate the beam path for a minimal requirement beam routing in order to creating an output beam. While only one of the 4 layouts was eventually used, I will briefly discuss the advantages and/or disadvantages of all the designs. First off, design a) introduces angular dispersion on MS1, distributing the wavelengths on MS2. By encoding a phase gradient with twice the magnitude into MS2, the beams are automatically rerouted backwards onto MS1 and finally - similar to assuming device operation under time inversion - the beams are collimated again and exit the device through the input aperture. This device features the lowest amount of metasurface-interactions provided that standard free-space propagation after initial angular dispersion is used to then apply locally changing, wavelength dependent phase information for the pulse-shaping functionality. The disadvantage with this layout is, that a beamsplitter is necessary to separate the shaped pulses from the incident laser source. More information is provided in the

experimental section. Design b) features 4 metasurface interactions and could distribute the application of wavelength dependent phase information to two metasurfaces (MS2 and MS3). Additionally, in order to achieve a completely symmetrical device and beam layout, a symmetry point is introduced in between MS2 and MS3. By performing this “beam sorting”, the functionality of MS3 and MS4 can be identical to the previous two metasurfaces. Ideally, all wavelengths intersect in one point, halfway between MS2 and MS3. As the generated spatial dispersion on MS2 is unique to the periodicity of MS1 and the propagation length, in general, a simple grating does not allow to perfectly invert the interaction point from MS2 to MS3, requiring something like a lens phase profile to achieve this function. The advantage with this design is, that all the generated signal can be used via the exit aperture since no beamsplitter is required. The design from figure 3.3 c) has no symmetry point and extends the previous design by placing metasurfaces on both sides of the glass substrate. In this design, it is not sufficient to encode a simple grating into MS3 in order to re-collimate the beam. The shorter wavelengths need to exhibit a larger diffraction angle than the long wavelengths, thus requiring a changing phase-gradient. In design d) this issue is tackled by creating another symmetry point in between MS2 and MS3 by introducing a lens-like phase profile on both metasurfaces. Since it is in general a good course of action to keep things as simple as possible, the pulse shaping devices adapted the metasurface layout of Figure 3.3 a) as it has a low amount of metasurface interactions and allows to create a symmetrically working device with little extra work required from the metasurfaces MS1 and MS2. While the collection of the generated signal is limited via a necessary beamsplitter, as long as the input power of the setup is not a bottleneck, something like a 30:70 beamsplitter can be used to efficiently collect the generated signal. Regarding design c) and d), while the alignment of two grating phase-profile metasurfaces (MS2 and MS3) is not too sensitive, as soon as wavelength dependent phase information for the pulse shaping is necessary, the alignment during fabrication will probably be an issue.

3.3.2 Geometrical phase and unit cell parameters

With the overall layout of the pulse shaping device explained, this section will focus on the method used to manipulate incident light and will also cover the required structural parameters of the unit cells to do so.

As the entire device will get its compactness from several interface reflections, I decided to use mirror-backed periodically arranged plasmonic antennas for the metasurfaces as these structures have proven to work well in reflective type applications [52]. Furthermore, they are easy to fabricate, typically requiring one deposition- and etching-step less compared to dielectric type metasurfaces. Since pulse shapers are ideally able to handle a large bandwidth, the method chosen for the phase encoding is the geometrical phase - also known as the Pancharatnam-Berry phase (PB-phase). It was first introduced by Shivaramakrishnan Pancharatnam in 1956 [53] and later explained extensively by Michael Berry in 1984 [54]. The geometrical phase uses structural anisotropy, introducing birefringence to change

the phase between orthogonal linear polarization states to alter an incident polarization. These structures can be described by e.g. two transmission coefficients corresponding to the structure's fast- and slow- axis. The easiest way to realize such birefringence would be by using rectangular shaped nano-antennas that exhibit different effective refractive indices along their short and long axis, respectively. As this approach is not necessarily tied to a resonance wavelength it is suitable in supporting broadband operation - similar to optical waveplates. Let's assume such rectangular structures rotated in their respective quadratic unit cell by an angle θ with respect to the x-axis. The transmission matrix $T(\theta)$ for this antenna can be expressed as

$$T(\theta) = \begin{pmatrix} \cos(\theta) & -\sin(\theta) \\ \sin(\theta) & \cos(\theta) \end{pmatrix} \cdot \begin{pmatrix} t_{xx} & 0 \\ 0 & t_{yy} \end{pmatrix} \cdot \begin{pmatrix} \cos(\theta) & \sin(\theta) \\ -\sin(\theta) & \cos(\theta) \end{pmatrix}. \quad (3.27)$$

t_{xx} and t_{yy} refer to the complex-valued transmission coefficients of the structure along both principle axis, assuming that they are oriented along the x- and y- axis of the unit cell. The matrices to the left and right corresponds to a rotation operation into the reference frame of such an antenna and back into the laboratory frame.

When a linear polarization state is incident onto this system, the transmitted polarization will depend on the orientation of the linear polarization with respect to the long and short axis of the nano-antenna. If the linear polarization is parallel to any semi-axis, it will acquire a phase shift from the corresponding transmission coefficient but will otherwise remain in that polarization state. However, for an arbitrary projection of the incident electric field onto both semi-axis of the antenna, generally an elliptical polarization state is generated. Conversely, if a circular polarization state interacts with this system, the projected electrical field will always be the same on both semi-axes. This means, the output polarization is always of the same kind, but the overall orientation angle of it can change. Assume that incident circular polarization is perfectly converted into linear polarization, then the vector of this polarization will depend on the orientation of the nano-antenna. However, if the transmitted polarization is supposed to yield the orthogonal circular polarization state, then its "orientation", which depends on the rotation angle θ is akin to an applied phase, as there is otherwise no apparent difference to a "rotated" circular polarization state.

So, for an incident circular polarization state with arbitrary helicity ($\sigma = 1/-1$) $|E_\sigma\rangle$, we can decompose the transmitted electric output field E_{out} into the co- $\langle E_\sigma|$ or cross-polarization $\langle E_{-\sigma}|$. For this example, co- and cross- polarization refer to the same and opposite helicity, respectively. This means, the transmitted co-polarization of an e.g. incident left circular polarization (LCP) is LCP, whereas the cross-polarization is right circular polarization (RCP) [55].

$$E_{\text{out}} = \langle E_\sigma| T |E_\sigma\rangle + \langle E_{-\sigma}| T |E_\sigma\rangle \quad (3.28)$$

$$E_{\text{out}} = \frac{t_{xx} + t_{yy}}{2} E_\sigma + e^{2i\sigma\theta} \frac{t_{xx} - t_{yy}}{2} E_{-\sigma} \quad (3.29)$$

We can see, that the resulting electric field has contributions of co- and cross-polarization

in dependence of the transmission coefficients. Also, the cross-polarization term carries a phase of $\phi = 2\sigma\theta$ that is related to the rotation angle of the rectangular antenna. The factor of 2 implies, that a 2π -phase shift can be realized by rotating the antenna 180° which - for a structure with C2-rotational symmetry - is equivalent to 0° rotation. Note that we thereby circumvent the issue, that a plasmonic resonance phase only allows for a $0 - \pi$ phase coverage. Since the co-polarization only carries a phase from the complex transmission coefficients but does not exhibit the geometrical phase term, the idea is to optimize the structural parameters in such a way, that the cross-polarization term of the outgoing field E_{out} is maximized. This can be achieved by satisfying $t_{xx} = -t_{yy}$, indicating a π -phase difference between the electric field components transmitted along both principle axis. Note that while the cross-polarization refers to a helicity-change in transmission type structures, for reflective type structures it refers to a maintained circular polarization. The derivation for a reflective system is mostly analogous, with the exception of the outgoing circular polarization, which is mirrored. With respect to equation 3.29, this effectively changes the sign in front of both t_{yy} terms as well as the sign of the helicity σ . Replacing the complex valued transmission coefficients with reflection coefficients r_{xx} and r_{yy} , we get

$$E_{\text{out}} = \frac{r_{xx}-r_{yy}}{2} E_{-\sigma} + e^{-2i\sigma\theta} \frac{r_{xx}+r_{yy}}{2} E_{\sigma} . \quad (3.30)$$

Note that for maximum reflected cross-polarization, the phase difference between the reflection coefficients has to be 0.

For these PB-phase metasurfaces, the geometrical properties of the antennas are typically not changed, only relying on the rotation angle θ of the antennas for the phase encoding. As a consequence, this phase encoding does not allow for sophisticated polarization or wavelength multiplexing. And it also does not allow for local amplitude modulation of a signal which would otherwise be performed by changing the antenna length. If the structural parameters are optimized and if the phase-change between the two orthogonal linear polarization states reaches π (or 0) for transmission (reflection) type unit cells, a full conversion to the cross-polarization is possible making the unit cell act as a half-wave plate. Another advantage of this phase encoding is, that the unwanted co-polarization can easily be filtered out in the experiment as both generated polarizations are orthogonal.

Because of the mentioned phase encoding principle, the unit cell layout considers cuboid shaped gold antennas (height H , length L , width W , periodicity P) on top of a BK7 glass substrate, covered by a spacer layer (thickness S) as well as a silver mirror of 100 nm thickness. The layout of the unit cell can be seen in figure 3.4 a). For the simulations, I used a MATLAB implemented version of an RCWA (rigorous coupled wave analysis) from Thomas Weiss [56] to simulate and optimize the unit cell's structural parameters depending on the cross-polarization conversion efficiency. Typically, birefringence of the system is maximized if the structure resembles a line whose length equals the periodicity of the unit cell but with a much smaller width - essentially forming a line grating. However, in this state, rotating the structures in their respective unit cell can cause a strong angle dependent coupling to neighboring antennas, turning their behavior from a local response

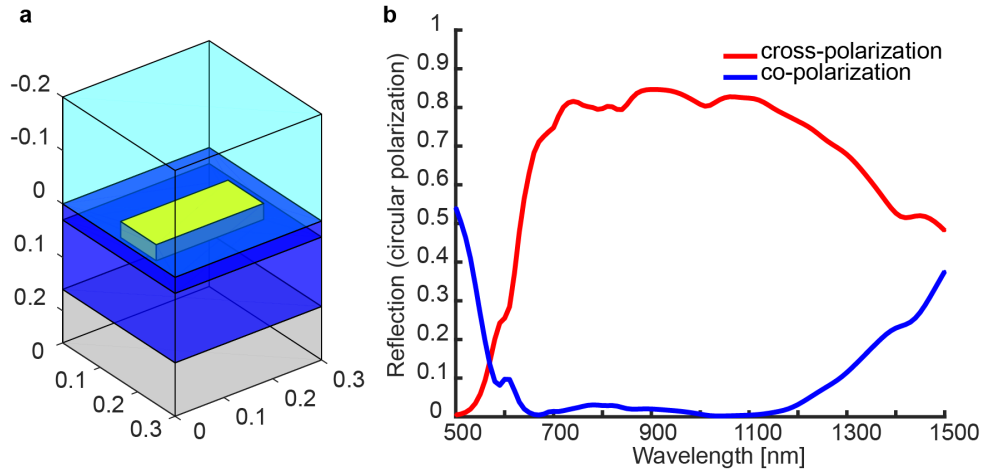


Figure 3.4: a) Optimized reflective type unit cell geometry. The light and dark blue colors resemble the glass substrate and the spacer layer. Yellow corresponds to the rectangular nanoantenna and the gray layer resembles the silver mirror. The numbers on the unit cell represent structural dimensions in μm . b) Corresponding spectral behavior of the structure. Both reflected circular polarization states - co- and cross-polarization - are plotted from 500 nm - 1500 nm. A targeted high cross-polarization reflection of over 80% can be seen between 700 nm - 1100 nm. [50]

to a nonlocal one. Therefore, these global maxima are avoided, as it might impact the idealized linear relation between rotation angle θ and applied phase.

The optimized parameters for the rectangular gold-antennas are a length of $L = 220$ nm, a width of $W = 95$ nm and a height of $H = 30$ nm. The unit cell periodicity is $P = 300$ nm and the spacer layer has a height of $S = 100$ nm. Figure 3.4 b) shows the conversion efficiency of the structure for optimized parameters over a broad spectral bandwidth. The structure shows a broadband, stable cross-polarization conversion efficiency of 80% between 700 - 1100 nm with very low reflected co-polarization. While the overall efficiency is good, the layout of the pulse-shaping device requires three metasurface interactions. Neglecting the losses from the mirror-reflections, this leads to an overall device efficiency of 50%.

3.3.3 Encoded phase information

This subsection will focus on the basic functionality, each metasurface has to provide in order to facilitate in- and out-coupling of laser pulses. First and foremost, the device must be able to guide light from MS1 to MS2 and couple it back out through the input aperture. To do this, we will draw an analogy to the classical grating and its corresponding grating equation

$$n_1 \sin(\theta_1) + n_2 \sin(\theta_2) = \frac{m\lambda}{\Lambda} . \quad (3.31)$$

In general, this equation describes a transmission type grating on the boundary of two different optical media (n_1 and n_2). $\theta_{1/2}$ correspond to the light propagation angle in medium 1 and 2, i.e. the angle of incidence (AOI) onto the grating and the diffraction angle. Δ corresponds to the grating periods/distance between adjacent slits and m is the respective diffraction order. When realizing a grating with a metasurface, we can replace the line-periodicity with a linearly increasing phase profile P_{blaze} along one spatial coordinate - akin to a blaze grating. By further simplifying that the AOI is 0° and that $m = 1$, we get the spatial phase $\phi_{\text{blaze}}(x)$ and the wavelength dependent diffraction angle $\theta_{\text{diff}}(\lambda)$ as

$$\phi_{\text{blaze}}(x) = \frac{2\pi}{P_{\text{blaze}}} \cdot x \quad (3.32)$$

$$\theta_{\text{diff}}(\lambda) = \text{asin} \left(\frac{\lambda}{n(\lambda) \cdot P_{\text{blaze}}} \right) . \quad (3.33)$$

Furthermore we have $P_{\text{blaze}} = N \cdot P$, with N being the number of meta-atoms per 2π -phase change and P as the period of an individual nano-antenna. In principle, any blaze-period P_{blaze} can be chosen as long as the period itself supports a diffraction order. For the device, we decided on a blaze-period of $2.4 \mu\text{m}$ which corresponds to 8 nano-antennas that cover a 2π phase increase. In terms of the geometrical phase this means that each antenna has a relative rotation of $180^\circ/8$ to its neighbors. The resulting diffraction angle at a wavelength of 760 nm is $\theta_{\text{diff}} = 12.1^\circ$. After the beam deflection, MS2 must be placed at an interface reflection point and has to incorporate another linear phase gradient to route the beam back towards MS1. As the AOI onto MS2 is now 12.1° instead of 0° and the diffraction angle is supposed to be anti-parallel to the AOI, a different blaze period is now necessary. Plugging these values into equation 3.31, we see that the left-hand side doubles, requiring halving of the blaze period on the right-hand side, i.e. twice the phase gradient. The resulting blaze period on MS2 is $P_{\text{blaze}} = 1.2 \mu\text{m}$ which corresponds to 4 nano-antennas per 2π -phase shift. Note that the linear phase gradient on MS2 now has a negative slope, scaling with $-x$ instead of x . After the interaction with MS2, the beam is directed back towards MS1 and diffracted normal to the surface allowing to couple light out of the input aperture.

In addition to the grating functionality, a lens phase profile on MS1 can be used to increase the spatial dispersion on MS2 as the beam size is reduced. The lens phase profile can be described via

$$\phi_{\text{lens}} = \frac{2\pi}{\lambda} (f - \sqrt{r^2 + f^2}) \quad (3.34)$$

r is the radial position on the metasurface and f is the focal length which will equal the propagation distance of the center wavelength from MS1 to MS2.

Furthermore, the device itself introduces dispersion which is constituted by the wavelength dependent paths in the device as well as the material dispersion. In order to apply a well defined phase information, this inherent dispersion has to be accounted for. To do this, the

Sellmeier equation for BK7 [57]

$$n(\lambda) = \sqrt{1 + \frac{B_1\lambda^2}{\lambda^2 - C_1} + \frac{B_2\lambda^2}{\lambda^2 - C_2} + \frac{B_3\lambda^2}{\lambda^2 - C_3}} \quad (3.35)$$

$$B_1 = 1.0396, B_2 = 0.2318, B_3 = 1.0105$$

$$C_1 = 0.0060, C_2 = 0.0200, C_3 = 103.56$$

is used to get the wavelength dependent refractive index $n(\lambda)$ - with B and C being the Sellmeier coefficients - and the grating equation 3.33 is used to calculate the diffraction angle. After diffraction from MS1, the beam is guided to the second metasurface. The center-center distance between MS1 and MS2 depends on the thickness d of the glass substrate, the diffraction angle $\theta_{\text{diff}}(\lambda)$ as well as the amount of mirror interface reflections r_i between MS1 and MS2. With these values defined, the location at which a specific wavelength hits MS2 can be expressed as

$$\frac{\lambda(x)}{n(\lambda)} = \sin \left(\tan \left(\frac{x}{r_i d} \right)^{-1} \right) P \quad (3.36)$$

Here, x can be an array of meta atom positions each separated by the periodicity P . Because of the wavelength dependent refractive index, this expression is not directly solvable for the vacuum wavelength. To approximate the corresponding wavelength-array λ for the nano antenna positions at coordinates x , a fixed-point iteration is used. This is done by initially setting the to be retrieved wavelengths equal to the center wavelength $\lambda_{\text{curr}} = \lambda_0$ and multiplying the left-hand side expression from equation 3.36 with the refractive index of the currently retrieved wavelengths $n(\lambda_{\text{curr}})$. This process is repeated for 10 iterations.

$$\lambda_{\text{curr}} = \frac{\lambda(x)}{n(\lambda)} \cdot n(\lambda_{\text{curr}}) \quad (3.37)$$

The resulting wavelengths λ_{curr} - referred to as λ in the following - will be used to calculate the propagation phase introduced by the device. The overall propagated geometrical distance $s(\lambda)$ over wavelength in the device can be calculated by:

$$s(\lambda) = 2d \left(\frac{r_i + 1}{\cos(\theta_{\text{diff}}(\lambda))} + 1 \right) \quad (3.38)$$

The propagation phase $\Phi_{\text{prop}}(\lambda)$ is

$$\Phi_{\text{prop}}(\lambda) = \frac{2\pi}{\lambda} \cdot s(\lambda) \quad (3.39)$$

In order for incident laser pulses to experience no pulse deformation through higher order dispersion terms, these contributions from the propagation phase have to be compensated on MS2. To extract the higher order terms, a linear regression is applied to Φ_{prop} and subtracted from it. Additionally, since this phase represents the accumulated propagation

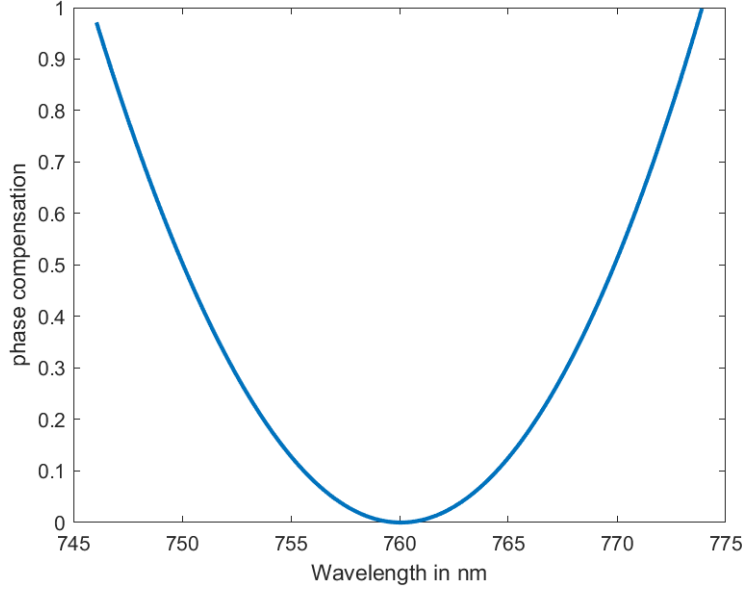


Figure 3.5: Phase information (given in rad) necessary to compensate the inherent device dispersion. The overall profile is mostly parabolic, indicating a dominating second order dispersion.

phase, in order to compensate for it the inverted phase $-\Phi_{\text{prop}}$ has to be encoded into MS2. With a substrate thickness of $d = 2$ mm, a center wavelength $\lambda_0 = 760$ nm, a bandwidth of $\Delta\lambda = 19$ nm, a blaze periods of $P_{\text{blaze, MS1}} = 2400$ nm and $r_i = 5$ interface reflections, the extracted dispersion that needs to be compensated is shown in figure 3.5. Because of the large amount of interface reflections, the propagation length of the center wavelength is $s(\lambda_0) \approx 2.85$ cm. From this the focal length of the lens phase profile on MS1 is calculated as $f(\lambda_0) = \frac{s(\lambda_0) - 2d}{2} \approx 1.225$ cm. With these parameters, the design of a device which allows the out coupling of a signal and compensation of the inherent device dispersion is possible. The already mentioned required phase profiles for MS1 and MS2 for such a device are illustrated in figure 3.6. The bottom right phase encoding is a placeholder for the actual pulse shaping functionality (as well as the dispersion compensation) that will be discussed in the measurement section. Note that while the focal length can be defined accurately for the center wavelength, this is generally not the case over the entire bandwidth. Recall, that the geometrical phase is applied via the nano-antenna orientation. By looking at equation 3.34 we can see that for a given lens phase profile $\phi_{\text{lens}}(\lambda_0)$ encoded into MS1, the phase for the entire bandwidth is set. With $\phi_{\text{lens}}(\lambda_0)$ fixed, if the wavevector on the right hand side increases for smaller wavelength, this has to be compensated by increasing the focal length f . This behavior can also be derived from the grating equation 3.33; by approximating a certain spot on the lens phase profile as a linear phase gradient, for a given 2π -phase period, the diffraction angle $\theta_{\text{diff}}(\lambda)$ decreases for smaller wavelengths. In this device, shorter wavelengths both propagate a shorter distance until they hit MS2 and they exhibit a larger focal length, causing a defocusing on MS2. Even though this effect

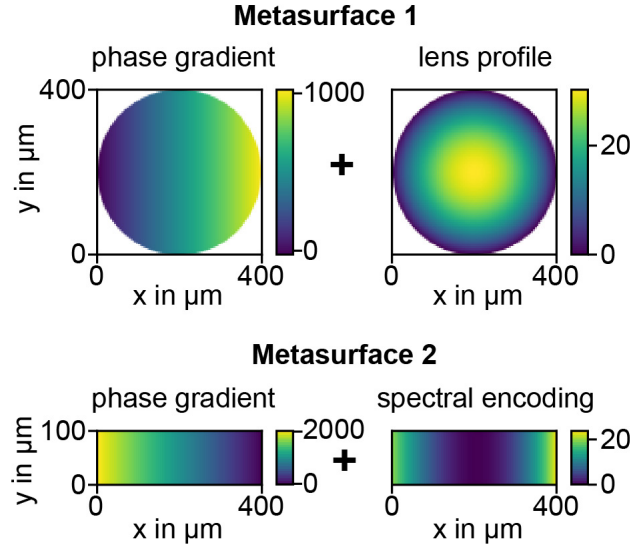


Figure 3.6: Phase information encoded in MS1 and MS2 given in rad. Both metasurfaces have a phase gradient to allow the basic routing of the beam. An additional lens phase profile on MS1 focuses the beam on MS2 and allows to re-collimate the beam. Additional spectral encoding such as the compensation of the inherent device dispersion are applied to MS2. [50]

does not allow for perfect focusing, it is still considered negligible for the bandwidth used in this work.

3.4 Fabrication

The pulse shaping device is fabricated on a single glass substrate and the workflow is illustrated in figure 3.7. I chose a 2 mm thick BK7-glass substrate (PGO), that has a thickness tolerance of $\pm 25 \mu\text{m}$. In addition to the larger than typical thickness of 1 mm - at least regarding the other samples presented in this work - a potential lateral offset of the diffracted beams is reduced since less interface reflections are required for the same amount of propagation distance, thus minimizing accumulated errors from deviating substrate thicknesses. The backside of the sample, meaning the side that hosts the input/output aperture of the device is fabricated first. The aperture is slit shaped with a width of $450 \mu\text{m}$ and will run across the entire substrate length of 10 mm in order to allow multiple sets of metasurfaces to be fabricated. Maximilian Protte kindly fabricated these apertures via laser lithography which takes seconds/minutes of patterning time as compared to electron beam lithography taking hours due to the large scale of the aperture. Additionally, there are no strict requirements on the resolution of the aperture, meaning that deviations of a micrometer will not affect the performance of the device. A negative tone photoresist is

used during the lithography process and after development a 100 nm thick silver layer is deposited using electron beam evaporation. This allows for a liftoff process done for 10 minutes in acetone at room temperature to finish off the aperture. As several fabrication steps are required for the frontside of the sample where the metasurfaces will be placed, a protective layer on the mirror is advised to prevent scratches or oxidation of the silver. 3 layers of IC1-200 spin-on-glass are spin coated at 3000 rpm for 30 s and are hard baked at 200 °C for 2 minutes each to achieve a roughly 500 nm thick protective layer.

In order to accurately place MS1 opposite to the aperture, a marker system is fabricated. A 150 nm thick PMMA layer is spin coated on the frontside of the sample (at 3000 rpm for 60 s) and hard baked at 180 °C for 2 minutes following a spin coating of 40 nm Elektra92 as a conductive layer (3000 rpm, 40 s) hard baked at 90 °C for 2 minutes. Not only the overall position of the aperture but possible tilt with respect to the sample edges need to be taken into account in order to place the metasurfaces opposite of the aperture. Therefore, several markers are patterned at an acceleration voltage of 50 kV and a dose of $450 \frac{\mu C}{cm^2}$ spaced by 100 μm near the top and bottom edge of the aperture. The Elektra92 is removed in deionized water (DI-water) for 1 minute and development is done in an MIBK based developer (AR-600-56) for 95 seconds. Development is stopped in Isopropanol for 30 seconds followed by an additional DI-water dip and nitrogen blowing to dry the sample. A 30 nm chromium layer is deposited using electron beam evaporation and the liftoff is done using acetone at 60 °C for 10 minutes followed by 30 seconds of low power ultrasonication. The aperture and the marker system is localized using a microscope, switching between imaging of the front and backside of the sample. The marker closest to the edge of the aperture is located and a scale included in the ocular of the microscope is used to estimate the distance between the closest marker and the edge of the aperture. To translate the scale from the ocular to a displacement in μm , the 100 μm distance between two markers is taken as a reference. These steps are followed for the two rows of markers (at the top and bottom edge of the sample) to derive an expression for the aperture position along the sample. With this information, PMMA and Elektra92 are spin coated again and MS1 and MS2 are patterned on the frontside. After development, 30 nm of gold are deposited via electron beam evaporation and the liftoff is done using acetone at 60 °C for 10 minutes followed by 30 seconds of low power ultrasonication. Figure 3.8 shows an SEM image of a metasurface after liftoff. The material for the spacer layer will also be IC1-200 spin-on-glass but in order to reach the desired 130 nm layer thickness, the spin-on-glass is diluted with the principle solvent Butan-1-ol at a 5:4 (IC1-200:Butan-1-ol) ratio and spin coated at 3000 rpm for 30 s. The desired thickness is confirmed via DektakXT profilometer measurements after scratching the film with a razor blade. Afterwards a 100 nm silver layer is deposited on top of the spacer layer using electron beam evaporation, finishing the reflective type metasurface stack. While not shown in figure 3.7, a 300 nm thick layer of maN-2403 resist is spin coated at 3000 rpm for 30 s and hard baked at 90 °C for 2 minutes. This layer is supposed to prevent oxidation of the silver mirror.

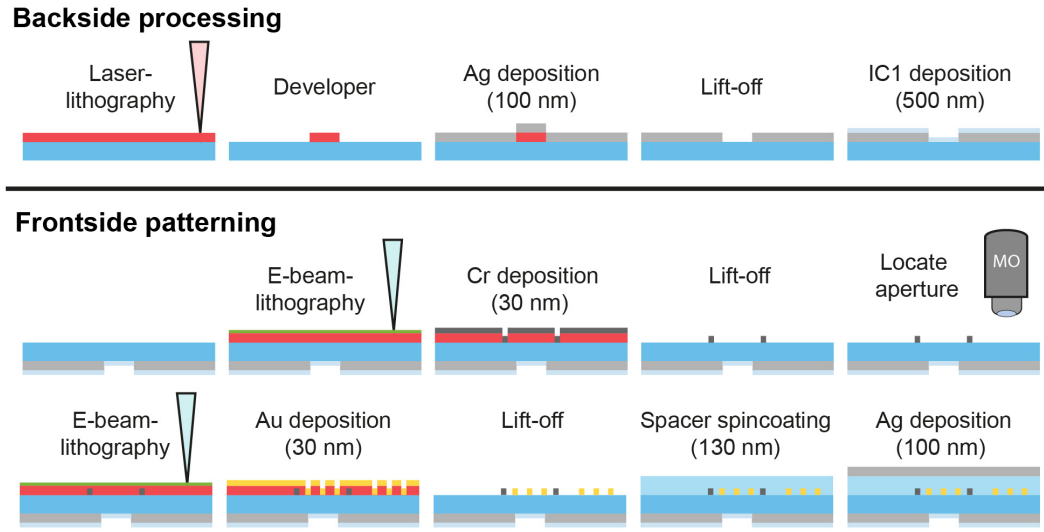


Figure 3.7: Workflow of pulse shaper sample fabrication. The workflow is divided into backside and frontside fabrication starting with the fabrication of the aperture via laser lithography, silver evaporation and a liftoff process. On the frontside a marker system is realized to properly align the metasurfaces with the aperture on the opposite side of the glass substrate. After patterning of the metasurfaces, a spacer layer is spin coated and finally the second silver mirror is deposited. [50]

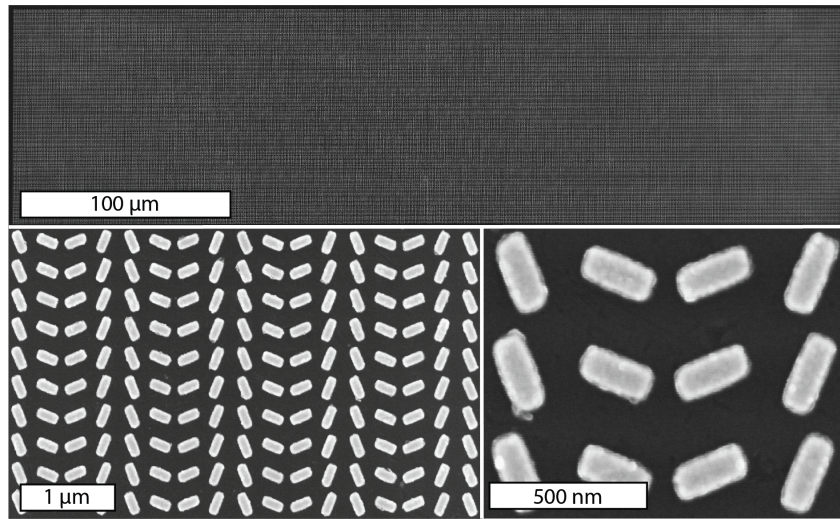


Figure 3.8: SEM micrograph of a MS2 variation in different magnifications. [50]

The described fabrication process covers the first successful sample fabrication while some steps have been optimized in later iterations. One major time save involved omitting the Cr-deposition for the marker system and only use developer on the PMMA resist instead.

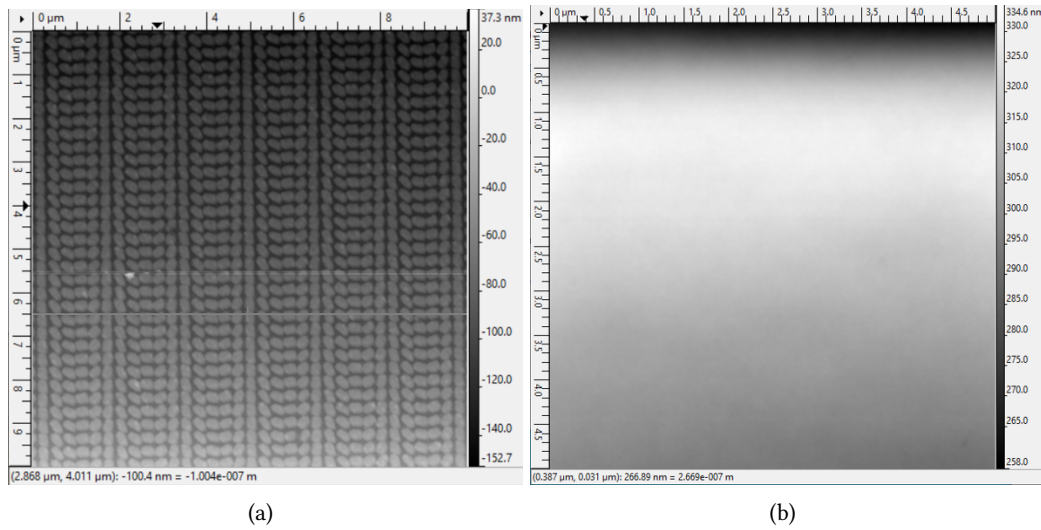


Figure 3.9: a) AFM measurement of a plasmonic metasurface covered in a 130 nm MgF₂ spacer layer deposited via electron beam evaporation. The MgF₂ spacer carries a clear footprint of the plasmonic nanoantennas and does not support a flat mirror application. b) AFM measurement of a plasmonic metasurface spin coated with a 150 nm PMMA spacer layer. The application of the spacer layer in liquid state leaves no traces of the embedded nanoantennas.

The developed markers are also nicely visible in the optical microscope allowing for the same localization quality as the Cr-markers. This saves an evaporation step and the only thing required between aperture localization and the patterning of the metasurfaces is the reapplication of Elektra92 since the PMMA was never removed. Finding the Cr-markers in the second lithography once they are buried in another PMMA layer is more of a headache anyway since the contrast in e-beam lithography is generally better if heavier metals are used. However, I did not want to deposit better contrast materials like gold for this step. Why did we opt to apply IC1-200 as the spacer layer? The choice of material itself does not really matter for this step but I should mention that certain spacer layer resists might deform if the sample is exposed to certain levels of heat, e.g. during baking of the resist that is supposed to protect the frontside silver mirror. In general, it is advisable to apply the spacer layer in a liquid state. Initial fabrication tests used electron beam evaporated MgF₂ and although the deposition height might be easier to control than during spin coating, the evaporation process left a clear footprint of the nanoantennas height profile which does not allow for a smooth mirror deposition (see figure 3.9). After switching to e.g. PMMA as a spacer layer, this height profile vanished and allowed to apply a flat mirror layer. In the end, the spacer material was changed to diluted IC1-200 since it is more resistant to heat. Note that if the protection layer of the frontside required a high temperature baking step, soft resists like PMMA can deform, ultimately destroying the smooth silver mirror.

3.5 Sample characterization and experimental setup

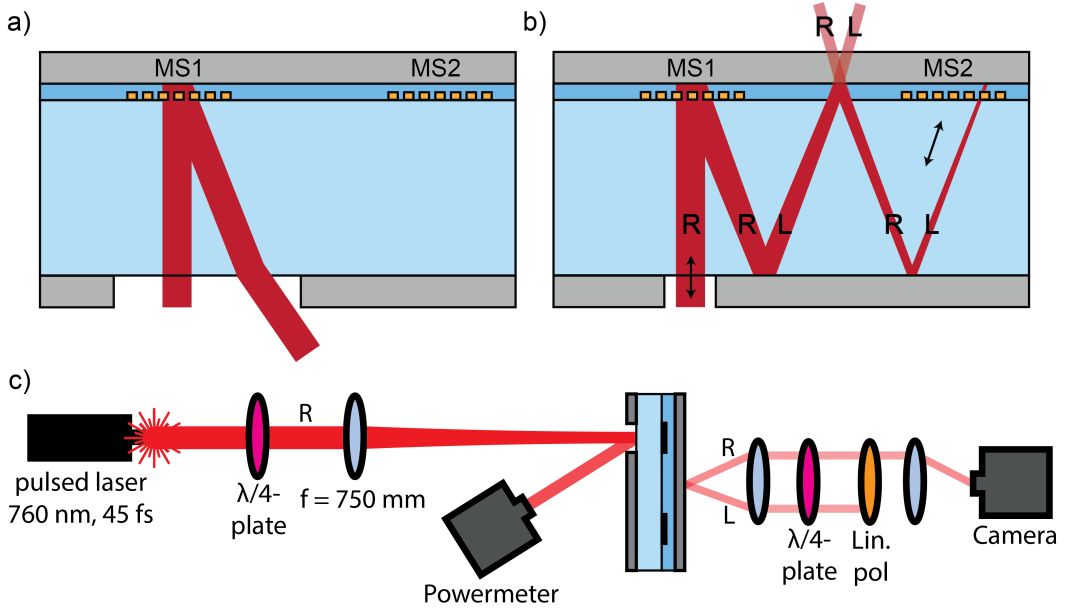


Figure 3.10: Illustration of the beam path relevant for determining the diffraction efficiency. a) Grating phase profile from MS1 with broader aperture to allow the diffraction order to couple to free space. b) Schematic beam path in a complete pulse shaping device, here illustrated with 3 instead of 5 interface reflections between MS1 and MS2. The helicity of the beam changes upon a mirror reflection whereas an interaction with a metasurface preserves the circular polarization state. The letters R and L refer to the circular polarization states in the propagation. By measuring the light which is transmitted through the metasurface-sided mirror with respect to the circular polarization, the ratio allows to gauge the diffraction efficiency of MS2 during normal device operation (excluding three silver mirror interface reflections). c) Experimental setup to determine the diffraction efficiency of MS1 and MS2. The powermeter is part of the diffraction efficiency measurement regarding MS1 and the imaging system behind the sample belongs to the measurement for MS2. A $\frac{\lambda}{4}$ -plate and a linear polarizer are used to filter a certain circular polarization out.

In order to measure the diffraction efficiency of the individual metasurfaces (see figure 3.10), the phase gradient from MS1 is fabricated as an individual structure (excluding the lens phase profile) and is also not positioned opposite to an aperture so that the generated diffraction order can exit the device unhindered. A $\frac{\lambda}{4}$ -plate is used to set the necessary circular polarization state and a lens with a focal length of 750 mm is used to focus the beam onto the metasurface. To get the diffraction efficiency, a powermeter is used to determine how much of the incident light is diffracted. For these measurements, the quality of the generated polarization and the two glass interface reflections on the sample were neglected. In contrast to MS1, MS2's efficiency is determined “in action” (see figure 3.10 b))

via a complete pulse shaping device in which the metasurface-side mirror is only 30 nm thick. An imaging system in combination with an analyzer behind the metasurfaces can be used, to collect the small amount of light that is transmitted through the silver mirror. By integrating over the camera image and comparing the images from the left and right circular polarization diffraction efficiency of MS2 (excluding three silver mirror interface reflections that were not considered) can be calculated. Individual measurements for both MS1 and MS2 showed efficiencies between 50-55% - definitely not in line with the expected 80% from the RCWA simulations. In order to figure out if a deviation in the targeted antenna sizes could cause the low efficiencies, several MS1 variants with different lengths and widths have been fabricated. Figure 3.11 shows the resulting diffraction efficiencies for incident right circular polarization. The measurements show that a generous parameter sweep around the targeted antenna dimensions of 220 x 95 nm still does not cover the expected 80% conversion efficiency from the simulations. Note that the simulations are referring to a single antenna with periodic boundary conditions whereas in the phase gradient from MS1, a total of 8 different orientation angles are present. And although diffraction efficiencies tend to decrease for large diffraction angles this alone should not amount to a decrease of 30% at $\approx 12^\circ$. While the cause could not be determined at the time, the measurements shown in figure 3.11 suggest to decrease the antenna width for future samples down to 90 nm and the design has not been further altered for the following samples.

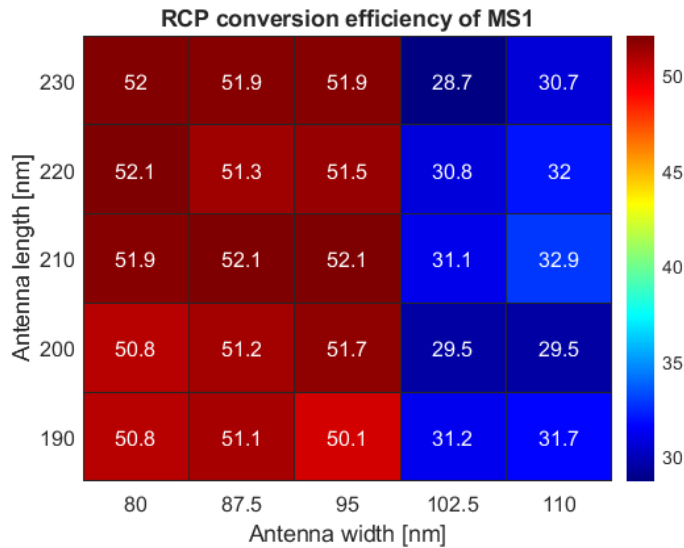


Figure 3.11: Measured diffraction efficiencies (cell values given in %) of phase gradients used in MS1 in dependence of the nanoantenna length and width. The initial design targets an antenna length of 220 nm and a width of 95 nm. The efficiency seems to drop significantly for slightly larger antenna widths.

Another concern was, whether the large amount of interface reflections introduces notice-

able polarization distortion caused by the reflection phase difference between s- and p-polarized light. Ideally, and mostly so for normal incidence, the reflection phase difference between these polarizations is π , causing RCP to convert to LCP after reflection. However, when the AOI is increased, the reflection coefficients for s- and p- polarization start to differ. From the BK7-Ag interface, using the AOI θ_1 in BK7 and the transmitted angle $\theta_2 = \frac{n_1}{n_2} \sin(\theta_{\text{inc}})$ in Ag from Snell's law, we can calculate the Fresnel coefficients via

$$r_s = \frac{n_1 \cos(\theta_1) - n_2 \cos(\theta_2)}{n_1 \cos(\theta_1) + n_2 \cos(\theta_2)} \quad (3.40)$$

$$r_p = \frac{n_2 \cos(\theta_1) - n_1 \cos(\theta_2)}{n_2 \cos(\theta_1) + n_1 \cos(\theta_2)} \quad (3.41)$$

with $n_1 = n_{\text{BK7}}$ and $n_2 = n_{\text{Ag}}$. Taking the phase from these complex valued coefficients, we can see that each reflection causes s- and p- polarization to de-phase by $\approx 0.83^\circ$ in addition to the relative π -phase shift. Assuming initial RCP and neglecting the polarization conversion to the opposite helicity for better clarity, we can illustrate the resulting polarization state after 10 interface reflections on a Poincaré sphere (see figure 3.12).

RCP after 10 reflections. AOI=12.1°, lambda=760 nm

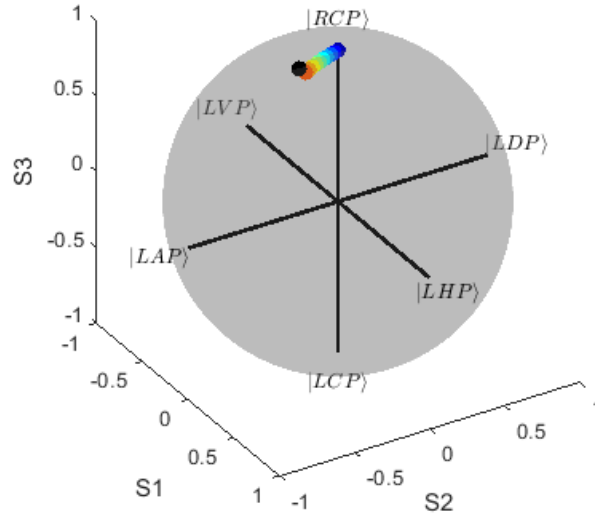


Figure 3.12: Poincaré sphere representation of a right circular polarization state at 760 nm after 10 BK7-Ag interface reflections under an AOI of 12.1°. The x-, y- and z-axes represent the Stokes parameters S1-S3 with circular polarization states at the poles and all linear polarization states along its equatorial plane. To improve visibility, the helicity conversion from RCP to LCP after each interface reflection is not included. The dot-markers represent the polarization state at each interface reflection starting with blue (initial polarization) and going towards red (10 interface reflections). For reference, the black dot corresponds to a single air-silver interface reflection at an AOI of 45°.

Increasing interface reflections are color coded with blue representing zero interface reflections and red corresponding to 10. We can see, that the polarization state is barely affected by consecutive interface reflections. After 10 reflections and neglecting intensity losses after a reflection, the remaining S3 parameter (corresponding to the degree of circular polarization) is 0.973, leaving the polarization mostly in the RCP state. This polarization distortion gets worse for larger AOI. The black marker shows the impact on RCP for a single air-silver interface reflection at an AOI of 45° . The change to the initial polarization state is roughly the same as the 10 BK7-Ag interface reflections, however the remaining intensity for the single air-silver interface is larger which is why the black dot looks a bit “out of line”. To put this into perspective: 6 air-silver interface reflections under an AOI of 45° are enough to convert a circular polarization state to a linear polarization state. This is why it is considered good practice to place quarter-wave plates close to where a certain polarization state is required, without excessive reflections in between the polarization state generation and the point of use. Note that the individual metasurfaces would anyway filter the partially converted polarization states as orthogonal circular polarization states each acquire an inverse geometrical phase. This leads to a spatial separation of opposite circular polarization states, especially when linear phase gradients are present. The reflectivity of the silver mirror was determined to be roughly 96% at 760 nm. Assuming a diffraction efficiency of 50% per metasurface and 10 interface reflections within the device until the signal exists through the aperture a total interaction efficiency of $\approx 8\%$ is expected.

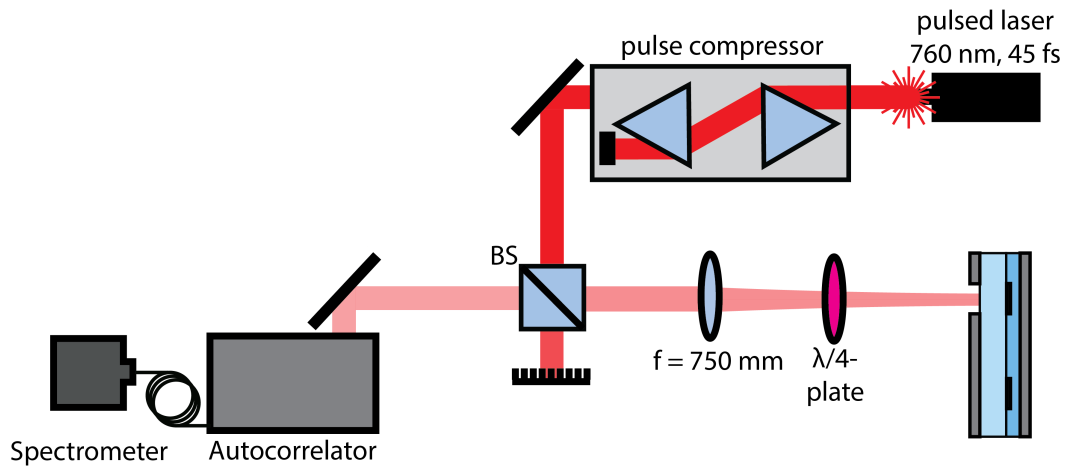


Figure 3.13: Experimental setup to characterize the applied phase of the modulated laser pulses. A pulse compressor allows initial control over the dispersion and the signal exiting the metasurface device is routed to an autocorrelator and a spectrometer for FROG measurements. An imaging setup behind the sample (not shown here) allows to ensure proper alignment of the sample by imaging the scattered light from the interface reflections.

The experimental setup used for dispersion characterization is illustrated in figure 3.13. A pulsed laser (Coherent Monaco laser and Opera-F optical parametric amplifier) at center

wavelength of 760 nm and a temporal width of 45 fs is used for the experiments. A pulse compressor controls the amount of second order dispersion in the optical setup while also weakly introducing higher order dispersion terms. The second prism inside the pulse compressor is motorized and its position is given by an arbitrary motor-step in the controlling software. By measuring the geometrical extend of the pulse compressor, considering the orientation of the prisms as well as their refractive index (SF10 glass), we can estimate the second and third order dispersion introduced per motor-step (see figure 3.14). At a wavelength of 760 nm, this estimation gives a GDD of 12.5 fs² and a TOD of 8.3 fs³ per prism position increment (motor-step). To put this into perspective, 8 motor-steps roughly amount to the second and third order dispersion of a 2 mm BK7-substrate at 760 nm.

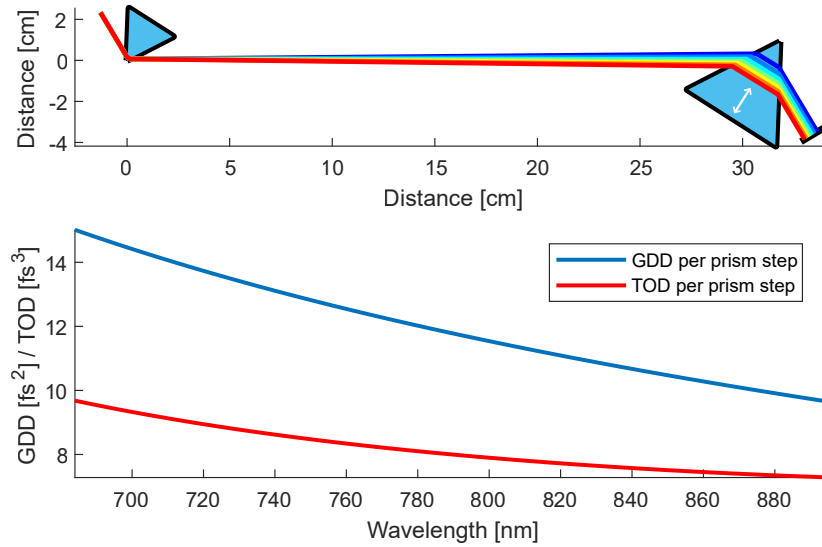


Figure 3.14: Top: Up to scale illustration of the used pulse compressor (side view). The shown beam paths cover a wavelength range from 680 nm (blue) to 900 nm (red). Bottom: Estimated second and third order dispersion per positive prism position increment (corresponding to an upwards movement of the second prism) plotted over wavelength.

A 70:30 (T:R) beamsplitter redirects the beam onto the sample while also increasing the collection efficiency of the generated signal. This is beneficial since the laser power is not a limitation in the setup while the efficiency of the sample is. Together with the 70% transmission from the beamsplitter, a total of 5% of the light incident onto the sample is expected to be rerouted to the autocorrelator. A lens with a focal length of 750 mm focuses the beam diameter down to $\approx 270 \mu\text{m}$. With a resulting Rayleigh length of $> 10 \text{ cm}$ this lens avoids severe beam divergence inside the device while also collimating the output signal from the sample. A $\frac{\lambda}{4}$ -plate is placed directly in front of the sample. Note that any output from the device can be separated into plain back reflection from the first metasurface (0th diffraction order that carries no geometrical phase information) and the signal which

came from an interaction-chain of MS1-MS2-MS1 via the respective 1st diffraction orders. Conveniently, light that is scattered into the 0th diffraction order does not preserve its helicity and can be compared to a simple mirror interface reflection whereas light scattering into the 1st diffraction order changes the helicity. As a consequence, the second pass through the waveplate separates these two signals into orthogonal linear polarization states. The relative fast-axis orientation of the waveplate is practically mirrored when the light is reflected off of the sample. Since the part of the light that interacted with MS1 is of the same helicity as the incident beam, a mirrored fast-axis of the waveplate undoes the polarization conversion from before thus casting the signal to the same linear polarization state that it was initially transformed from. This is a circumstance inherent to the chosen design as the reflective design of the pulse shaper is symmetric around the MS2 interaction and any change in helicity is applied twice. Otherwise the use of a polarizing beamsplitter would result in maximal signal collection. However, the separation into two orthogonal linear polarization states allows to easily filter the irrelevant signal. The experimental setup does not contain a linear polarizer at this point since the autocorrelator (GECO scanning autocorrelator from Light Conversion) works via SHG in which the nonlinear crystal (BBO) is sensitive to the incident linear polarization thus only converting the relevant signal. An illustration of the autocorrelation setup was shown in figure 3.1. A beamsplitter inside the autocorrelator generates two beam paths from which one introduces a time delay $\Delta\tau$, controlling the temporal overlap on the nonlinear crystal (BBO) and thus the amount of SH generated. By scanning a time delay range larger than the temporal width of the laser pulses, time delay dependent spectral information can be acquired via the FROG retrieval algorithm.

3.6 Pulse shaper measurements

The pulse shaper devices fabricated in this work feature four different phase profiles for MS2. Disregarding the necessary grating structure on MS2, these phase encodings are shown in figure 3.15 a). Firstly, we consider a device without any further phase encodings. We also design a metasurface, that is supposed to correct the inherent device dispersion introduced. These two samples are referred to as “uncompensated” and “compensated”. The phase profile responsible for compensating the device dispersion will also be integrated into the other MS2 designs. We also apply a large amount of TOD (30000 fs³) defined as

$$\phi(\omega) = \phi_{\text{comp}} + \frac{30000 \text{ fs}^3}{3!} \cdot (\omega - \omega_0)^3. \quad (3.42)$$

To put this into perspective, this amount of TOD is the equivalent of roughly 1 m of BK7 glass [46]. Lastly, we wanted to generate a double pulse with a temporal separation of $\tau_{DP} = 200$ fs. While ideally, this requires both the proper spectral phase and the modulation of the spectral amplitude, here we modulate the phase with a π -phase shift periodically in

the frequency domain via

$$\phi_{DP}(\omega) = \phi_{comp} + \text{sgn} \left(\sin \left((\omega - \omega_0) \cdot \frac{\tau_{DP}}{2} \right) \right) \quad (3.43)$$

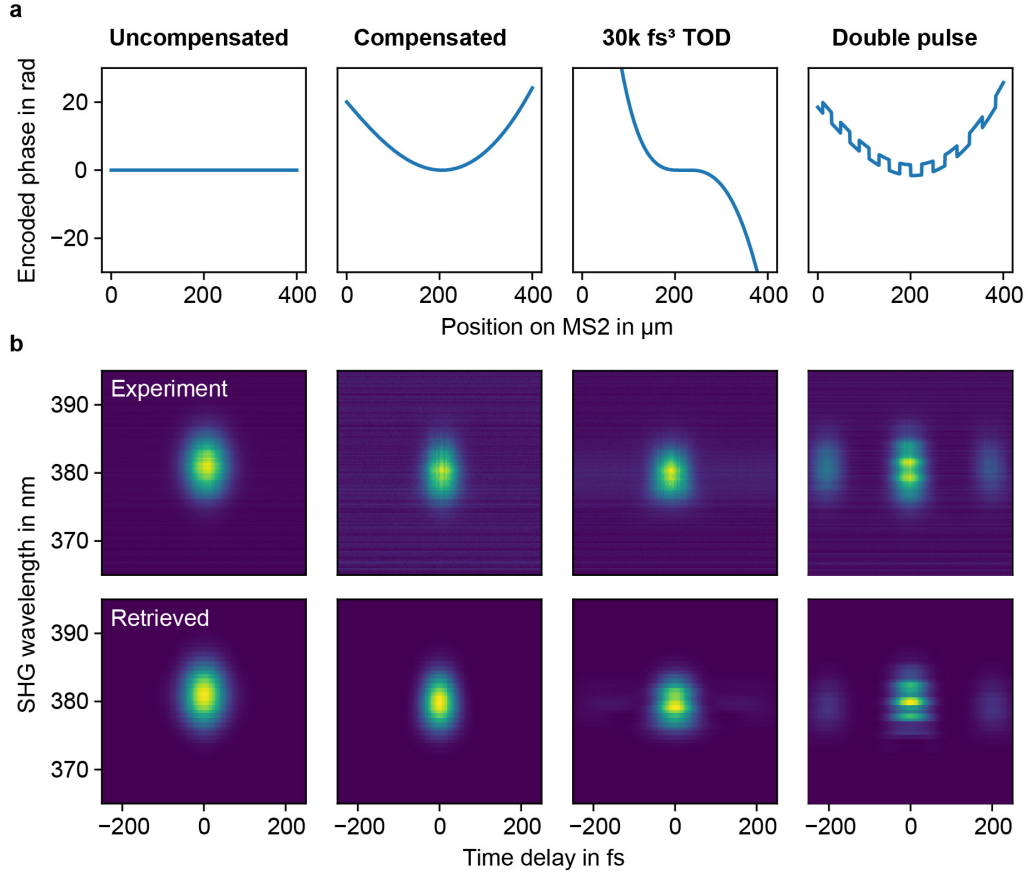


Figure 3.15: a) Phase profiles encoded into MS2, plotted against the position on MS2. Besides the “Uncompensated” design, all other samples contain the phase information shown in “Compensated”, which is meant to compensate the inherent dispersion otherwise introduced by the device. “30k fs³ TOD” introduces a TOD of 30000 fs³ and “Double Pulse” generates a double pulse with a temporal separation of 200 fs via a periodic π -phase modulation. b) Experimental SHG FROG traces (top row) and the retrieved traces (bottom row) after post-processing. [50]

The measurements of this work were recorded for a time delay range of 500 fs with a resolution of 1 fs. The wavelengths are recorded from 350 nm to 420 nm in steps of 0.21 nm. A reference measurement is acquired by reflecting off of one of the silver mirrors on the samples, leaving out the metasurfaces. In the experiments, we also changed the prism position of the pulse compressor to introduce different amounts of GDD while finding that a prism position of 480 results in the shortest pulses for the reference measurement.

FROG traces (experimental and retrieved) from the four designs mentioned are shown in figure 3.15 b). For the phase retrieval, post-processing of the traces is performed. The overall beam path length from the laser to the sample and into the autocorrelator is rather long, spanning several meters. Since the signal is also fiber-coupled to a spectrometer, laser power fluctuations as well as small beam steering due to air pressure fluctuations can severely impact the quality of the trace. To reduce the measurement artifacts from these effects, the traces are post-processed using a Savitzky-Golay smoothing filter. Furthermore, the boundary of the trace is considered as background and subtracted from the measurement. Also, the experimental trace is rescaled in order to improve the calculation speed of the performed Fast Fourier Transforms with a chosen grid-size of 251×251 data points.

The measurements shown in figure 3.15 b) were performed at a prism position of 480. The FROG trace of the “uncompensated” sample is wider in the temporal domain than the “compensated” sample, nicely showing the influence of the dispersion correction implemented in the latter. The “30k fs³ TOD” sample is expected to show an hourglass form in the measured FROG traces. This feature is not visible in the measurements which rather resemble a triangle pointing upwards, probably coming from the bottom half of the expected hourglass shape. An explanation for this might be a misalignment of the incident laser beam which does not properly hit the center of the second metasurface. With the saddle point of the TOD shifted away from the center wavelength, this might explain the “shift” of the expected pattern away from the hourglass shape. This assumption was not considered during the time of working on this project. Be it misalignment or a deviation in the expected substrate thickness, that caused a shift of the focused wavelengths along MS2, in the experiment, it would probably be easiest to change the center wavelength of the laser pulses and redo the FROG measurements. While I did not fabricate a new sample in order to verify this assumption, we can numerically calculate SHG FROG traces for a spectral shift of the TOD phase in comparison to a TOD phase centered around the center frequency of the laser pulses (figure 3.16).

The “Double Pulse” sample shows three characteristic peaks in the FROG trace with a strongly visible fringe pattern in the center peak. These fringes arise if multiple pulses interact with the spectrometer within an integration time-bin by interfering on the detecting electronic. The pitch of these fringes is dependent on the temporal separation of the detected pulse-train and can be deduced from the sine-term of equation 3.43. Taking the two intensity-minima at 378.8 nm and 381.2 nm and evaluating the argument of the sine-term while assuming $\tau_{DP} = 200$ fs and $\omega - \omega_0$ as the difference between the two mentioned wavelengths, we get $\approx \pi$, which relates the spectral intensity modulation to the 200 fs pulse separation. On a side note, the laser system used here has a repetition rate of 1 MHz, giving a temporal separation of 1 μ s between successive pulses. This means the frequency difference necessary to see a single period of this spectral oscillation is $5 \cdot 10^6$ times larger compared to the double pulse, explaining why this effect is typically not visible when observing the pulsed laser spectrum in “single pulse” operation, e.g. all the other traces. The temporal delay between the generated double pulses overall matches

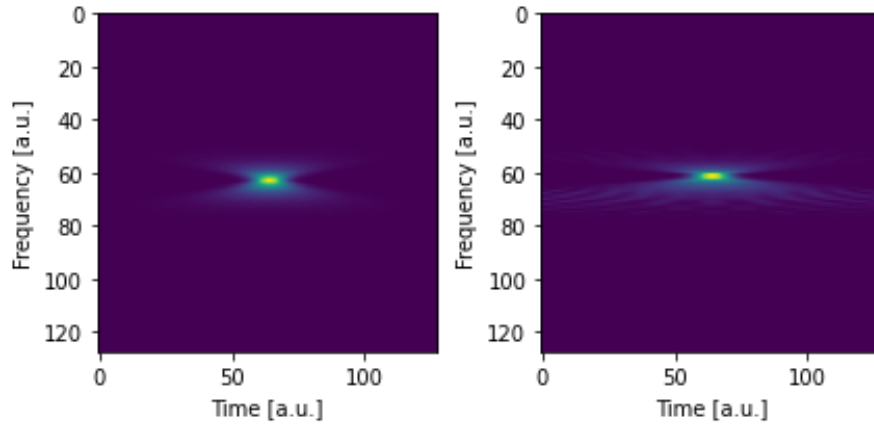


Figure 3.16: FROG trace with application of large TOD plotted over time and frequency with arbitrary units. left) FROG trace with TOD phase profile application symmetric around the spectral amplitude. right) FROG trace for TOD phase profile shifted to larger wavelengths/smaller frequencies. The symmetric hourglass vanishes instead showing a more triangular formation similar to the measured traces.

the intended 200 fs, which is visible from both the fringe-pattern as well as the faint side-maxima in the trace.

By changing the prism compressor position in our setup, thereby introducing different starting GDD values, we repeated the measurements shown in figure 3.15. Here, larger compressor positions correspond to larger GDD values. By applying the FROG retrieval algorithm (evaluation conducted by Philip Georgi) to these FROG traces and using a third-order polynomial fit on the retrieved spectral phase, we obtained the corresponding GDD and TOD shown in Figure 3.17. As expected, the reference measurement shows a linear dependency on the compressor position, resulting in near zero GDD for a position of 480. The reference curve covers roughly 2750 fs^2 across 200 motor-steps, resulting in 13.8 fs^2 per motor-step which is close to the prediction from figure 3.14 of 12.5 fs^2 per motor-step. The compensated phase profile shows similar GDD values indicating that a large amount of the device dispersion was compensated. In contrast, the “uncompensated” sample shows a large shift in the retrieved GDD. Near a value of zero GDD, both the “compensated” and “uncompensated” sample show a plateau which might come from a misinterpretation of the actual trace height and width. If the measured trace corresponds to a sub-time-bandwidth-limited laser pulse, the retrieval algorithm will pick the closest physically possible trace which is most likely a bandwidth-limited pulse with zero dispersion. This would introduce a plateau near the zero GDD point, causing the retrieved GDD to be systematically underestimated. Since this effect does not occur for the reference measurement, the measurement error might be linked to the much lower power coming out of the pulse shaping device. The retrieved TOD is shown in panel b). We expect a change of 8.3 fs^3 in the TOD per prism motor step which is negligible when compared to the expected TOD encoded into the metasurface, resulting in a nearly constant TOD for

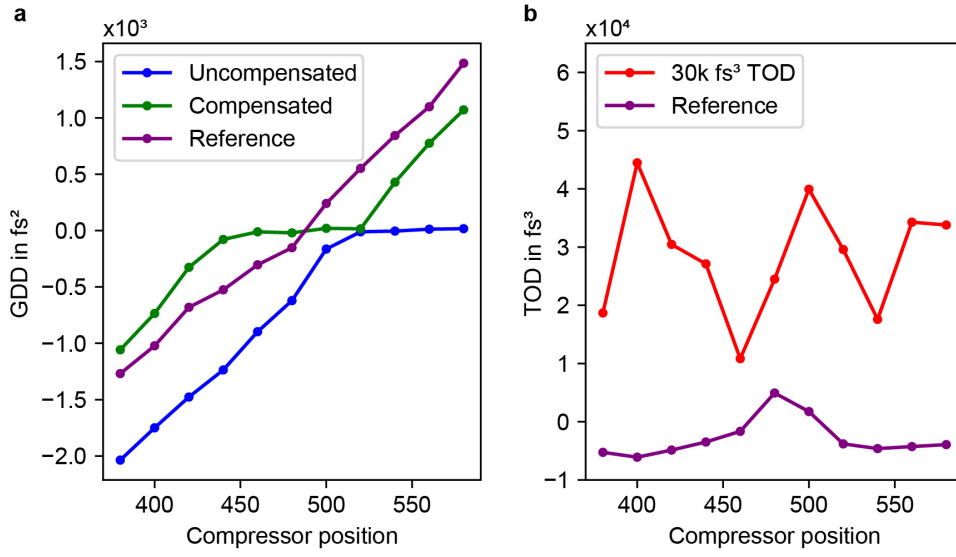


Figure 3.17: Retrieved GDD (a) and TOD (b) for various pulse compressor prism positions. The GDD of the reference measurement linearly increases with the prism position whereas “Compensated” and “Uncompensated” both show a plateau near their respective 0 GDD position with a linear slope for most other compressor positions. The retrieved TOD shows large variation for different compressor positions with a mean value close to the desired 30000 fs^3 . The reference measurement shows an unexpected peak near the compressor position where the temporal width of the laser pulse is expected to be the shortest. [50]

the reference measurement. Here, we see a peak around the 480 position which - similar to the retrieved GDD - is unexpected. The “ $30k \text{ fs}^3$ ” sample shows a large variation in the retrieved TOD which can not come from the pulse compressor itself. I am not sure, what caused this effect to arise but the alignment of the setup certainly plays a crucial role. Nevertheless, closer evaluation of the measured traces shows good agreement with intended functionality. By averaging over the difference between the “uncompensated” measurement and the reference measurement, we calculate a shift of $(-767 \pm 629) \text{ fs}^2$. This shift roughly corresponds to the inherent second order dispersion of our device of -784 fs^2 . The difference between the “compensated” measurement and the reference gives $(-17 \pm 768) \text{ fs}^2$. Meanwhile, the average TOD is $(31 \pm 22) \cdot 10^3 \text{ fs}^3$ which is close to the encoded value.

Lastly, we show the retrieved electric field amplitude of the generated double pulse (see figure 3.18). The measured temporal separation of 207 fs is in good agreement with the designed 200 fs. Furthermore, the electric field amplitude of the two pulses is noticeably imbalanced which is not intended by the design. Similar to the encoded TOD, this can also be attributed to a misalignment that causes the two phase-profiles that are separated by a π -phase shift, to be unequally illuminated thus favoring the generation of one of the two

pulses by introducing a wavelength shift along the phase-encoding direction on MS2. Since the spatial width of the individual phase sections in the center of MS2 is roughly $23\text{ }\mu\text{m}$ and with the entire bandwidth of the laser pulses only spanning roughly $100\text{ }\mu\text{m}$ on MS2, it is possible, that the center frequency shifted into the middle of either the “unaltered” phase profile or the sections that are raised by π , leading to said imbalance. To correct this, I could have changed the center wavelength by a couple nanometers which was not done at the time.

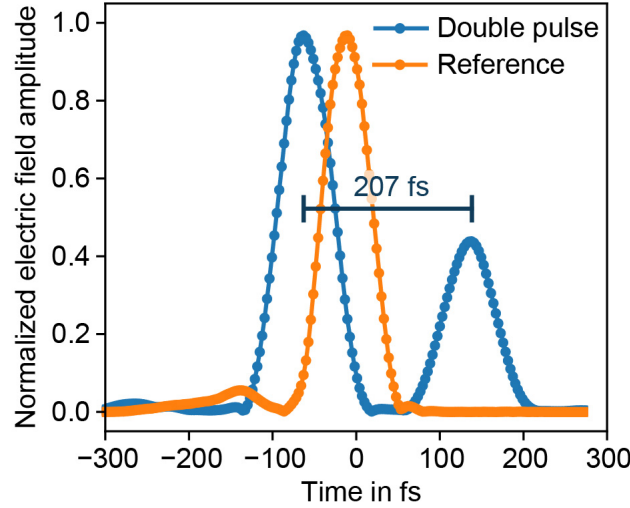


Figure 3.18: Retrieved temporal electric field amplitude from the reference pulse and the generated double pulse. The temporal separation of the double pulse matches the target of 200 fs but shows non-homogeneous field amplitude. [50]

3.7 Postliminary work

In cooperation with Dr. Vikas Remesh from Innsbruck university, I performed some follow-up work with these pulse-shapers and got the chance to understand some of the limitations and shortcomings of the previously presented devices.

His idea required a pulse-shaping device capable of applying a much larger second order dispersion ($3 \cdot 10^6\text{ fs}^2$) than we previously targeted. He wanted to use it to realize robust off-resonant excitation of a quantum dot (QD) biexciton state using chirped pulses, which he already showed in a more bulky setup [31, 58]. The advantage of the chirped excitation is, that a QD-ensemble with varying resonance frequencies can be collectively excited using a pulsed laser-source with a fixed center wavelength. Provided that the laser pulses are chirped enough, the QD excitation becomes invariant with respect to further laser power increases after the initial excitation power is reached.

An illustration of the intended setup is shown in figure 3.19, while the overall goal was to fully integrate the pulse shaper with the QD sample at a later stage. Similar to the

previous experimental setup, beam incidence onto and signal collection from the pulse shaper is done with a doubly-passed beamsplitter. Here, the pulse shaper is supposed to add 0.3 ps^2 to 150 fs pulses with a center wavelength of 770 nm (bandwidth of 5.8 nm). A linear polarizer allows filtering of the 0th diffraction order from the device. The QD sample, located in a cryostat, will also be used in a reflective scheme, requiring another double-pass of a beamsplitter. Since the QD sample will be excited with circularly polarized light, another QWP is used. The excited biexciton state (excitation wavelength of 780 nm) will decay by emitting two photons - one from the decay of the biexciton to the exciton state, another from the exciton-ground state transition. In a coincidence measurement, using single photon detectors, the excitation behavior of the system will be analyzed. Ideally, after reaching enough laser power to excite the QD, its excitation behavior will become invariant to an increase in laser power.

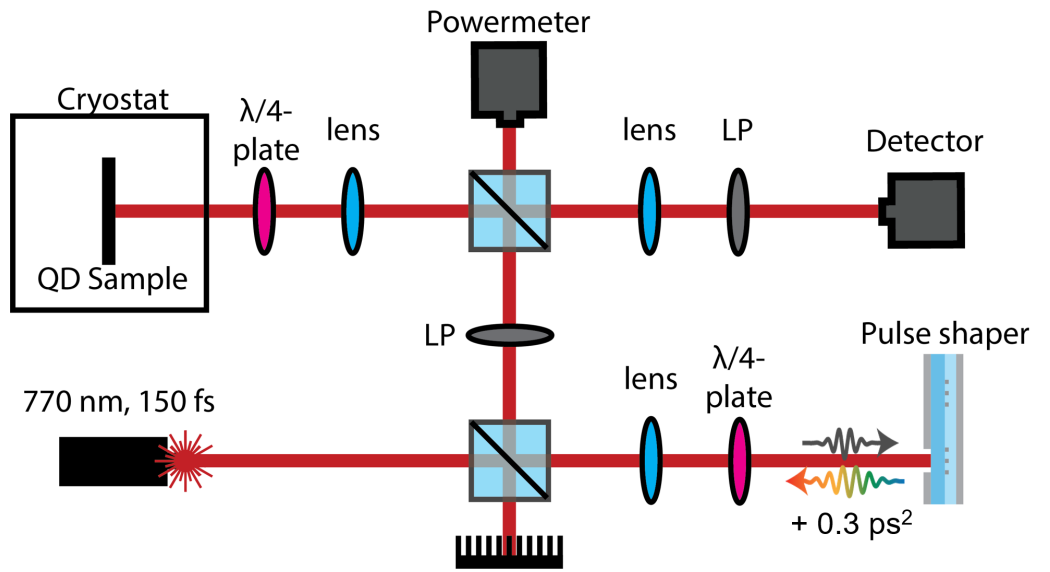


Figure 3.19: Experimental setup for chirped pulse excitation of QDs via the pulse shaping metasurface. The laser pulses with a center wavelength of 770 nm and a temporal width of 150 fs are routed to the pulse shaping metasurface and the generated signal carries an additional second-order dispersion of 0.3 ps^2 . By passing the QWP in front of the sample a second time, the polarization of the shaped pulse is transformed to a linear polarization state. Afterwards, the shaped pulses are routed to the QD sample in a cryostat, passing a QWP to generate circular polarization. The emitted photons from the QDs are detected in a single photon detection coincidence measurement setup (here abbreviated with a detector symbol).

The required spectral phase for this application as well as the calculated inherent device dispersion is shown in figure 3.20. The top panel shows the assumed gaussian intensity distribution of the used laser pulses over wavelength. The $\frac{1}{e^2}$ bandwidth is roughly 10 nm (FWHM of 5.8 nm) and the bottom panel shows the necessary MS2 phase information for

the required GDD as well as the compensation of the device dispersion (neglecting the phase gradient of MS2). The data points of the plotted phase profiles correspond to the later phase sampling of the metasurface, considering a lateral extend twice as large as the previous pulse shaper, featuring 11 interface reflections between MS1 and MS2, totaling 22 interface reflections and 3 MS interactions. Besides that, the design scheme of MS1 and MS2 is still analogous to the previous design, using 8 unit cells for the grating structure on MS1 and 4 unit cells for the grating on MS2. The lateral extend of MS2 is however reduced to 100 μm , which still fully covers the required bandwidth. At 765 nm and 775 nm, a 2π phase increase is still covered by 10 antennas indicating good phase sampling.

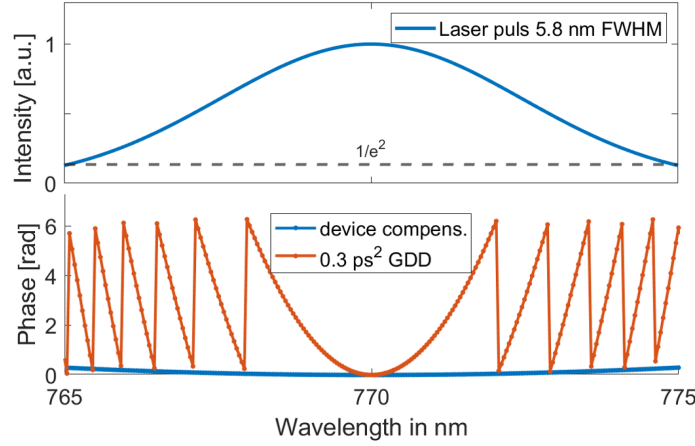


Figure 3.20: Top) Gaussian intensity distribution of the laser pulses that are used in the pulse-shaping experiments. The $\frac{1}{e^2}$ bandwidth of the laser pulses is roughly 10 nm. Bottom) Required phase compensation from the inherent device dispersion and phase information to apply a GDD of 0.3 ps² plotted over wavelength.

As a heads up: Because the project idea did not work out, I will not go into further detail regarding this excitation scheme. Instead, I will share new insides about the pulse shaping devices that were acquired along the way.

One of the main issues to tackle for this setup definitely was the rather low device efficiency. Considering the increased number of interface reflections, the low laser induced damage threshold, the requirement to increase the power later on to verify the power-invariant QD excitation, as well as the additional beamsplitter in the setup, figuring out why the samples have such a low diffraction efficiency was an important problem to solve. In order to figure out, at what point the previous sample's performance differs from the simulations, I measured the transmission spectra for incident x- and y- polarization of a strictly periodic nanoantenna arrangement - in accordance with the performed simulations. The gold antenna parameters fabricated/simulated here are identical to the ones used in the previously presented pulse shaper. Figure 3.21 shows the measured and simulated transmission spectra for the bare metasurface (left panel) as well as the metasurface with the additional spacer layer (right panel).

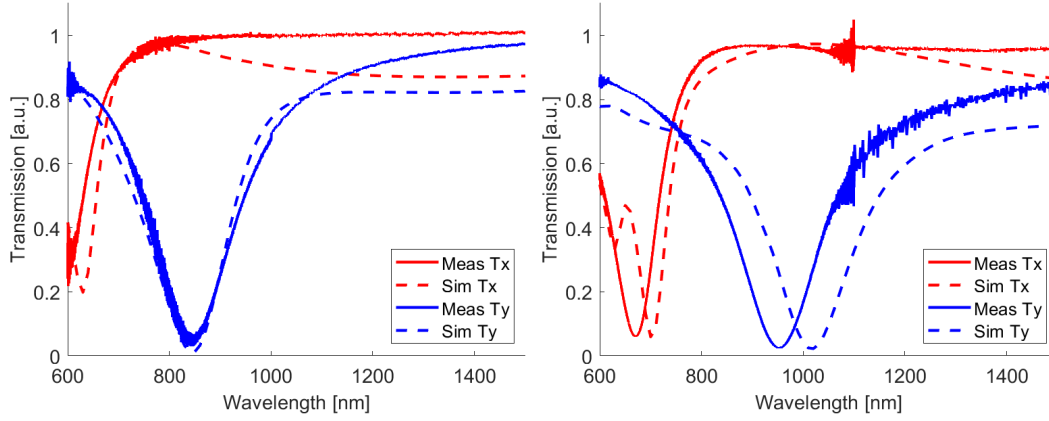


Figure 3.21: Left) Transmission measurement of a Au-metasurface before application of the spacer layer. The plot shows both the simulated and measured transmission of incident x- and y-polarized light, which are in good agreement especially around the target wavelength of 760 nm. Right) Transmission measurement of the Au-metasurface after application of the spacer layer and required baking of 180 °C for 2 minutes. The measured and simulated transmission of both x- and y-polarized light show a noticeable difference with blue-shifted resonances compared to the simulations.

We can see that the simulated optical properties of the bare metasurface - especially the transmission dip around 840 nm - are nicely captured by the simulations. Note that the y-polarization in the experiment is aligned with the long axis of the rectangular antennas. After depositing the spacer layer and performing the baking at 180 °C for 2 minutes, we can see that the measured resonance for y-polarization is blue-shifted roughly 100 nm from the simulations. This indicates that the optical properties of the nano-antennas change when they are heated too much which consequently diminishes the predictability of the pulse-shaper device efficiency. Instead of anticipating/estimating this resonance shift, I changed the spacer material to a negative tone e-beam resist (maN-2401) which is baked at only 90 °C. Additionally, I switched the nano-antenna material from gold to silver as it tends to give larger conversion efficiencies in the simulations. Considering the larger refractive index of maN-2401 (1.621 at 770 nm), RCWA simulations yield $L = 185$ nm, $W = 85$ nm, $H = 30$ nm, $P = 300$ nm and a spacer layer thickness of $S = 100$ nm as good parameters, resulting in 94% conversion efficiency.

Figure 3.22 a) shows the measured transmission spectra for a bare silver-antenna metasurface without geometrical phase modulation (all antennas are oriented equally) before and after heating at 90 °C (without application of a spacer layer). We can see, that the temperature used does not noticeably affect the transmission of the structure. Additionally, from the mentioned geometrical parameters, the grating structures from MS1 and MS2 are fabricated and their diffraction efficiency is measured (see panel b)). The red curve corresponds to the MS1 phase grating with 8 unit cells per 2π -phase increase, whereas the blue curve gives the diffraction efficiency of the MS2 grating with 4 unit cells per 2π -phase increase. We can see that the measured diffraction efficiency is around 75% over

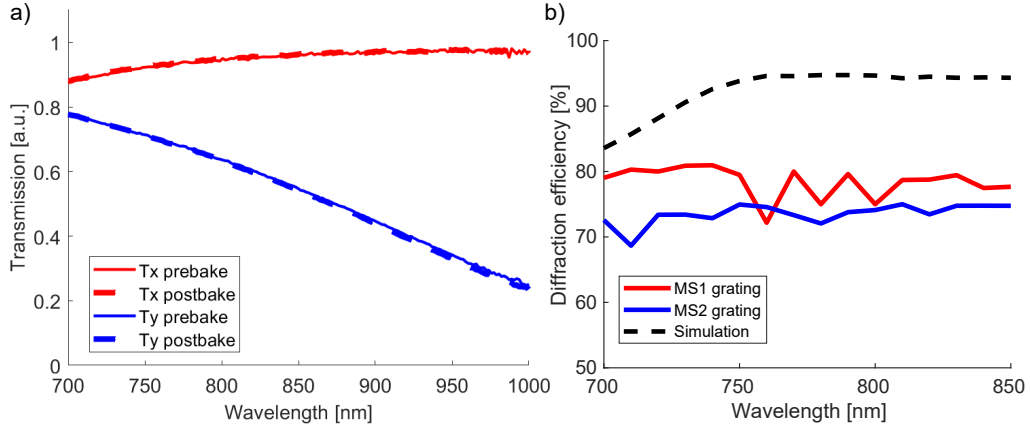


Figure 3.22: a) Polarization dependent transmission measurement of an Ag-metasurface before (pre) and after (post) baking at 90 °C for 2 minutes without application of a spacer layer. The temperature of 90 °C represents the recommended hard bake temperature of the to-be-applied spacer material (negative EBL resist maN-2401) and the measurements show, that the spectral behavior is not influenced by the heating. b) Measured and simulated diffraction efficiency of the silver-antenna based gratings from MS1 and MS2 (including spacer and mirror layer).

the wavelength range from 700 nm - 850 nm. The slightly lower diffraction efficiency of the MS2 grating is attributed to the larger diffraction angle. While still being lower than the simulations, the results are more in line with other literature with similar unit cell layouts [52]. With the current improvement, the overall efficiency of the device would increase by more than 3.4 times. To put this into perspective, if the sample were to reach the simulated 94% diffraction efficiency, the overall efficiency would increase by 6.6 times.

In addition to addressing the efficiency issue, I wanted to estimate the actual BK7-thickness compared to the assumed 2 mm substrate. As the device in question features more interface reflection than the previous pulse-shapers, a thickness deviation causes more lateral shift of the beam onto MS2. The overall thickness d of the substrate with roughly 2 mm is in an awkward spot where thin-film interference/ellipsometry measurements are out of the question and more direct measurement devices are too imprecise. While there are probably more efficient ways to determine the actual substrate thickness, here I used a metasurface in combination with a mirror on opposite sides of a BK7-substrate (see figure 3.23 a)). The idea is to illuminate the metasurface with laser pulses at a center-wavelength of 770 nm that partially reflect off of said metasurface. The transmitted part resembles a ghosting pulse that is reflected off of the backside mirror on the substrate and then transmitted through the metasurface creating a time delay Δt between the first and second reflected pulse equal to $\Delta t = \frac{2nd}{c}$. I will not go into too much detail regarding the metasurface design as the performance was also not characterized in the later experiment, only mentioning that the reflectivity is ideally chosen in such a way, that the directly reflected pulse is of roughly equal intensity as the doubly-transmitted pulse. The two pulses are guided into

the GECO autocorrelator where the zero delay (overlap of a pulse with itself, here located at -1565 fs) is taken as reference to the detected ghosting pulse. In order to maximize the generated SH, it is beneficial to have both reflected pulses at roughly equal intensity, while deviations in the targeted optical behavior of the metasurface decrease the visibility a bit. By doing sweep scans around different stage positions, the ghosting pulse was ultimately found with a nice symmetric intensity distribution at a delay position of -21785 fs (see Figure 3.23 b)). Using the difference to the zero-delay position as well as the refractive index of BK7 at 770 nm $n(770 \text{ nm}) = 1.5114$, the ghosting pulse has a total time delay of 20220 fs which results in a thickness $d = 2005.4 \text{ }\mu\text{m}$. Since the time delay considers twice the substrate thickness d and is affected by the refractive index n , a deviation of 1 fs in the ghosting pulse localization results in a thickness-deviation of only 100 nm. Pairing this with the very obvious scan-asymmetry caused by a mere 30 fs shift in the stage position shown in figure 3.23 b), the overall error is estimated to be ± 5 fs, resulting in a thickness of $d = 2005.4 \pm 0.5 \text{ }\mu\text{m}$. Since MS1 diffracts the center wavelength at an angle of 12.25° , the lateral shift Δs for every two interface reflections is $\Delta s \approx 1.15 \text{ }\mu\text{m}$, leading to a total lateral shift of $6.9 \text{ }\mu\text{m}$. This deviation is not expected to severely compromise the device functionality, however it can be one of the reasons, why e.g. the TOD FROG trace showed a more triangular shape rather than an hourglass shape (see figure 3.16) which is attributed to a misalignment of the phase information to the incident spectrum. For this reason, the newly fabricated devices considered this offset by slightly adjusting the MS2 position.

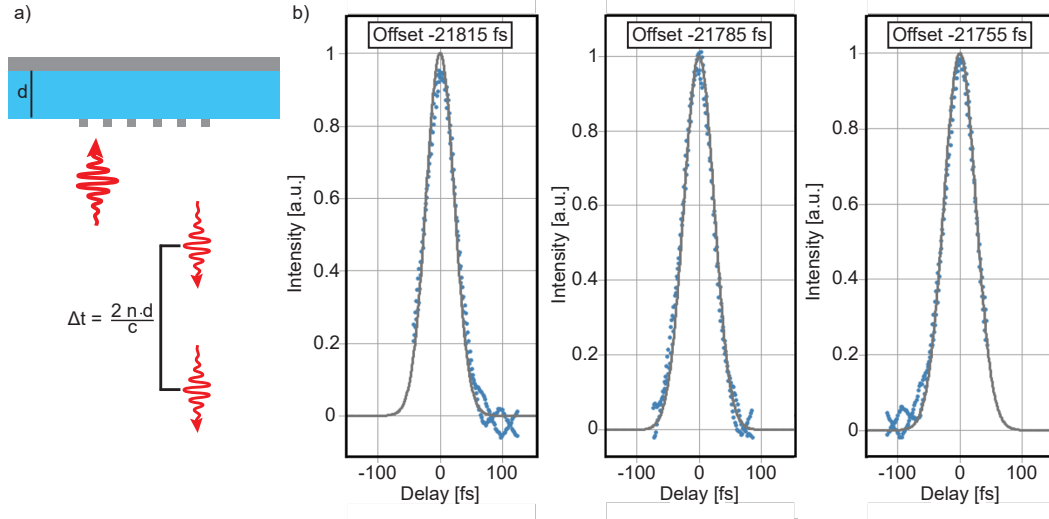


Figure 3.23: Indirect thickness measurement scheme of the BK7-substrate thickness d via autocorrelation. a) A partially transmissive/reflective film on one side of the BK7-substrate and a mirror on the other allows an incoming laser-pulse to create two reflections at both glass interfaces. The two pulses are sent into the GECO autocorrelator. b) Autocorrelation measurements of the direct back reflection from the metasurface with the ghosting pulse. The autocorrelator sweeps the delay stage around a certain delay position. The three panels represent measurements around three slightly different delay positions in 30 fs steps. An offset of -21785 fs shows a symmetric sampling around the found SHG peak intensity.

With these improvements, I fabricated the pulse shaping device on the glass substrate that was also used for the thickness measurement. During experimental characterization, a powermeter was placed behind the first LP in figure 3.19. Since the signal that did not fully interact with the device is filtered out by the LP, the measured power can be used to gauge the efficiency of the device by comparing it to the power incident onto the pulse shaper. The losses introduced from the lens and the QWP as well as the exact performance of the 50:50 beamsplitter were not investigated. In order to operate the device below the damage threshold, the incident power is limited to 30 mW. However, the overall power detected by the powermeter only resembles 0.6% of the input power while the estimate was $\approx 40\%$ considering a metasurface diffraction efficiency of $\approx 75\%$. The measured output efficiency was too low to be of use in the experiments, as the required power was around 0.7 mW [58].

In order to understand this poor performance, the device has been modeled in a MATLAB implemented 2D beam propagation method (BPM). The input beam radius is set to 135 μm and the spatial resolution is adjusted to the metasurface periodicity, allowing for a 1:1 sampling of the metasurface phase profiles. To increase visibility of the propagation process, the device is “unfolded”, resembling a transmission type design. In the BPM, both the metasurfaces and the mirror reflections were considered to be lossless. Figure 3.24

a) shows the beam propagation through an example device, while only considering the grating functionality on MS2 with no additional phase information. The y-axis corresponds to the lateral propagation/extension of the device whereas the x-axis is the propagation in z-direction of the unfolded device. The colormap shows the electric field magnitude in arbitrary units. For visibility, the wavelengths simulated in panel a) all have equal intensity and are chosen as 725 nm, 760 nm and 795 nm, nicely illustrating the incoupling into the device (0 mm z-position) until reaching MS1 after $d = 2$ mm propagation in z-direction. The three wavelengths spatially separate and are focused on MS2 after which they are recombined and interact with MS1 again, resulting in a collimated beam. Panel b) shows the propagation of the currently studied device from MS2 to MS1 with only 12.5% of the targeted GDD. Multiple wavelengths between 763 nm - 777 nm are calculated and overlapped, in accordance with the spectral composition of the experimentally used laser pulses. By integrating over the signal that reaches the output aperture and comparing this to the signal incident on the input aperture, it can be seen, that for the full dispersion, roughly 1.6% of the light entering the device is also coupled out again. Factoring in the metasurface diffraction efficiencies of 75%, this gives 0.7% outcoupling - close to the measured 0.6%. This low efficiency can be understood by the steep parabolic phase profile on MS2, which acts as a lens creating a virtual focus very close to MS2 and introducing large amounts of beam divergence ultimately spreading the light to such a degree that only a fraction interacts with MS1 again. Figure 3.24 c) shows the cross section of the BPM at the second MS1 interaction. The black lines mark the boundaries of MS1, showing that most of the light is coupled out of the device if no dispersion is applied. However, when gradually increasing the GDD, significant losses are introduced. This was an oversight in the design and in hindsight it was a coincidence, that the GDD values chosen in the previously realized pulse shapers were small enough to not exhibit this effect to such a degree. Simulating the propagation of the “compensated” and “30k TOD” design from the previous chapters predicts an outcoupling of 94.5% and 91.5%, respectively (excluding the diffraction efficiencies) which is decently high.

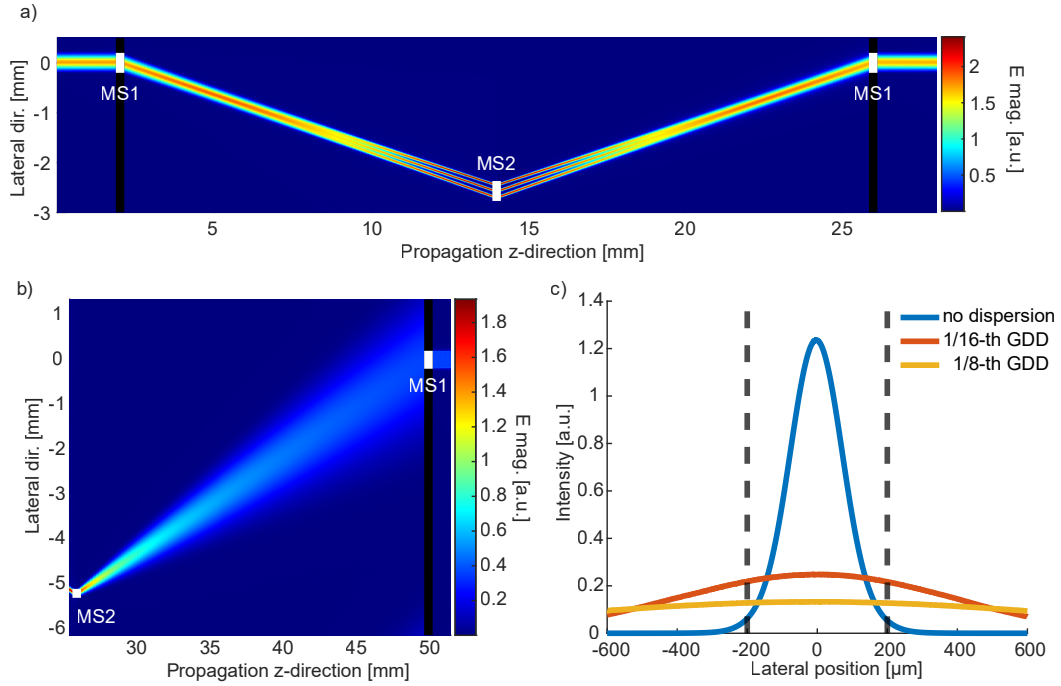


Figure 3.24: a) Beam propagation of an unfolded pulse shaping device (cross section). The phase information on MS2 only includes the grating functionality. The simulation is done for the wavelengths of 725 nm, 760 nm and 795 nm. b) Beam propagation of an unfolded pulse shaping device (cross section). The figure shows the propagation from MS2 to MS1 for a wavelength range between 763 nm - 777 nm. MS2 is encoded with only 1/8th of the actually used GDD in the fabricated device (initial goal were 0.3 ps^2), already showing strong beam distortion. The two white lines mark the position and lateral extent of MS1 and MS2. b) Cross section around the second MS1 interaction, illustrating the beam distortion in dependence of the encoded GDD on MS2. The black vertical lines mark the left and right boundary of MS1.

For this particular pulse shaper geometry, the encoded phase does not seem to be compatible with a high efficiency output. Note that significantly increasing the diameter of MS1, thus increasing the collection capability of the system does not solve the issue. This change would have to go hand in hand with an increase of the input/output aperture size and create issues in the minimal necessary diffraction angle. This way, light that enters the device gets diffracted on MS1 and directly exits again through the input aperture. However, increasing the collection optic diameter is possible in other pulse shaper layouts. Considering the presented layout, one solution to this problem would be to change the size of the entire pulse shaper. Assume, the amount of interface reflections in the device is halved, this would also scale the necessary size of MS2 down by a factor of two and compress the to be encoded phase information. While the overall parabolic shape of the GDD phase profile stays the same, and with the phase equation of a lens 3.34 in mind, this means that for half the spatial extension, the same phase change has to be realized. Along with possible

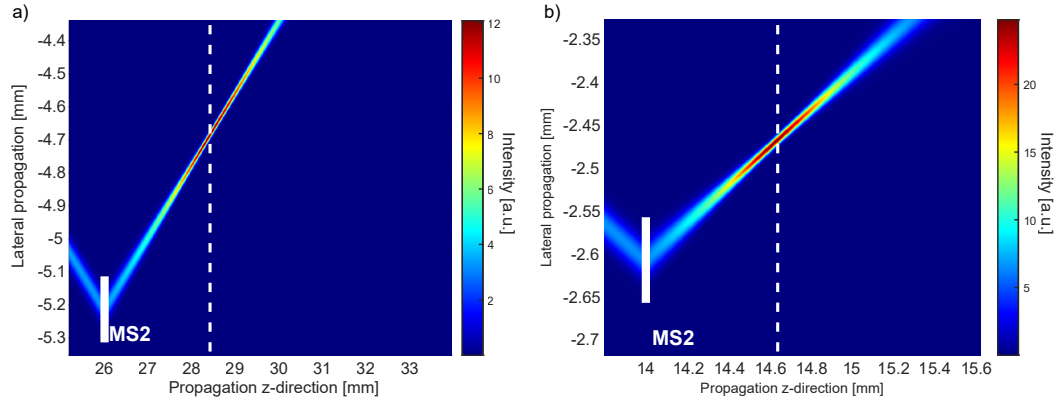


Figure 3.25: BPM of two different pulse shaper device extensions for fixed GDD. The simulations are intended to visualize the behavior of the perceived focal length in comparison to device extension. For visibility, only 5% of the required GDD is simulated while inverting the phase sign to create a real focus between MS2 and MS1, which otherwise is virtual and situated in front of MS2. a) BPM of the fabricated pulse shaper layout. The roughly evaluated focal length along z-direction is 2.4 mm. b) BPM of a smaller pulse shaper which considers only half of the interface reflections. As a consequence, the extension of MS2 is halved to roughly 100 μm . The focal length in z-direction corresponds to roughly 600 μm .

phase sampling issues for steep phase profiles, this can cause the apparent focal length to decrease by a factor of roughly 4. Conversely, increasing the size of the pulse shaper by a factor of 2 increases the focal length by a factor of 4. At some point the ratio between focal length to device propagation length is increased to a tolerable amount, reducing the effects of beam distortion significantly. Note that the comparison between an actual focal length from a lens phase profile to a strictly parabolic phase profile of the second order dispersion is only superficial, does however decently capture the observed effect.

Figure 3.25 illustrates this focal length difference for two device configurations, where the device extensions differ by a factor of 2. To increase visibility, the applied GDD phase profile is only 5% of the targeted 0.3 ps^2 and the phase sign is inverted, in order to create a real focus between MS2 and MS1. Using the original device dimensions, for the mentioned phase profile, a focal length of roughly 2.4 mm in propagation direction is observed whereas a device with half of the lateral extension (realized by halving the number of interface reflections) shows a focal length of roughly 600 μm . For the full phase encoding in the current pulse shaping layout, the focal length from MS2 would be around 130 μm . In order to salvage the functionality of the device, the observed focal length would have to be in the same order of magnitude as the distance between MS2 and MS1. This would mean, the device has to be extended by a factor of over 200 times leading to a pulse shaper with a propagation length in the 5 m range. For the purpose of this collaboration, the required dispersion is not realistic to implement with the current sample layout and was thus not further pursued.

3.8 Conclusion

In this chapter, I presented a compact metasurface-based pulse shaping device, that allows for successive metasurface interactions on a single glass substrate by introduction of mirrors with an interaction volume of less than 2 mm^3 . Incident laser-pulses with a center wavelength of 760 nm are spectrally decomposed via the introduction of angular dispersion and a wavelength dependent phase is encoded through the device's second metasurface. In order to evaluate the performance of the device, we conduct SHG FROG measurements and evaluate the device functionality via a FROG retrieval algorithm (implemented by Philip Georgi).

By comparing the retrieved GDD of the "Compensated" and "Uncompensated" sample as well as a reference measurement, we find that the "Uncompensated" sample shows a GDD difference to the reference of $(-767 \pm 629) \text{ fs}^2$ close to the expected -784 fs^2 . The "Compensated" sample results in GDD difference of $(-17 \pm 768) \text{ fs}^2$ to the reference which is also close to the expected value of 0 fs^2 . However, we find that the retrieved GDD curves for the two samples show a plateau around small GDD values which we attribute to measurement errors that result in a misinterpretation of the FROG traces width and height. For compressor positions further away from this plateau, the GDD over compressor position seems to have the expected linear slope.

We also applied 30.000 fs^3 of TOD which were measured to be $(31 \pm 22) \cdot 10^3 \text{ fs}^3$. This measurement series showed a large variation in the retrieved TOD over compressor position which does not originate from the pulse compressor itself. The reference measurement also shows a peak in the TOD around the prism position where the pulse width is minimized. This effect could not be explained by the author. We also found that the corresponding traces do partially show the expected hourglass shape however a misalignment resulting in a lateral shift of the laser spectrum on MS2 probably caused a de-symmetrization of this trace.

Lastly, we show a more complex pulse shaping operation by generating a double pulse with a temporal separation of 200 fs (measured to be 207 fs) to great success. The electric field amplitude of the two generated pulses shows a clear imbalance however - similar to the TOD sample - this can also be explained by a small lateral shift along MS2 which causes uneven illumination of the two present phase profiles.

Overall, we found that the encoded phase mostly showed the expected behavior however some issues remain with the measurement setup and the subsequent phase retrieval. This might be coupled to the lower signal-to-noise ratio because of the low device efficiency which becomes even more pronounced because of the nonlinear optical process involved in the autocorrelation measurement. The measured FROG traces are sensitive to misalignment of the samples as a beam shift on the second metasurface leads to spectral shift in the to-be-encoded phase information. Also, the plasmonic nano-antennas result in a relatively low laser-induced damage threshold which could be circumvented by fabricating devices with dielectric nanoantennas. Our design is easy to adapt and the spectral resolution can be increased by allowing for larger propagation length between MS1 and MS2, while this comes at the cost of increased interface reflection losses. Another idea is to allow for

dedicated input and output ports that do not require a beamsplitter in the experimental setup and also allow for spatial pulse shaping by placing metasurfaces near the output port (e.g. Vortex-beam generators).

The results mentioned up to this point were published in “Nano Letters” in 2023 [50].

Further research in due to an outreach of Dr. Remesh Vikas allowed me to better understand the limitation of the pulse shaping devices. I found that the baking at 200 °C necessary for the spacer layer causes the optical properties of the gold nanoantennas to change ultimately reducing their diffraction efficiency down to roughly 50%. By changing the spacer layer to the negative-type e-beam resist maN-2401, we can reduce the temperature requirement during baking down to 90 °C which seems to be low enough to not alter the optical properties of the antennas. The newly fabricated devices also feature silver antennas instead of gold antennas and show a greatly increased diffraction efficiency of up to 80%.

With the larger GDD requirement of 0.3 ps², the device performance significantly decreased and I found that beam distortion severely reduces the amount of light that can be coupled out of the device. After simulating the previously fabricated pulse shapers, I confirmed, that the previously applied GDD and TOD was small enough to not affect the overall device performance too much.

The presented device would benefit from an implementation based on dielectric nano-antennas (shown e.g. by Soma et al. [43] in their optical mode converter) which would significantly increase the laser induced damage threshold. Additionally, fabricating a device which has a distinct input and output aperture would make integration of such a device more convenient. An ultimate limitation of the presented device is the beam distortion which is introduced when applying large amounts of dispersion. This problem requires larger optical components (i.e. larger metasurfaces) to properly collect the modulated pulses or the extension of the overall pulse shaper. Overall, I think that these kind of integrated devices will become more and more popular, especially when considering optical networking where multiple metasurface layers could be used in a neural network-approach.

Nonlinear geometrical phase metasurfaces

4

4.1 Introduction

With an ever increasing requirement for miniaturization as well as data transfer rates the need for compact, flexible and efficient routing of optical signals becomes more and more important. In 2025, the global mobile data traffic recorded by the International Telecommunication Union (ITU) exceeds 1500 Exabytes (157 Exabytes in Europe), with an average annual increase of 19% since 2021. Putting this into perspective, this equates to an average of 17.9 GB of monthly mobile traffic per person in high-income countries [59]. In order to keep up with growing demand, free space optical terahertz radiation is considered for the next generation of cellular network technology, i.e. 6G. THz communication, spanning from 0.1 THz - 10 THz, potentially allows for large bandwidth high data transfer rates, far surpassing current 5G technology [60]. However, as of now, the atmospheric absorption of this radiation due to water molecules in the air heavily limits the application, imposing exponentially power losses as communication distance increases [61]. In its crudest implementation, one simply transmits the signals into the entire surrounding space while simply increasing the signal strength in order overcome absorption losses or signal-blocking obstacles. The obvious downsides are high power consumption considering the inefficient approach as well as interference effects. A more sophisticated approach only directs signals to where the actual receivers are, making sure to avoid obstacles such as weather imposed absorption zones by clever beam steering to achieve meaningful communication distances [62]. The capability of such systems can be extended further by use of nonlinear optical processes, allowing convenient conversion from different targeted transmission bands within the THz regime. If the beam steering and transmission channel conversion can be combined, it completely circumvents the requirement to demodulate signals and to re-encode them in a new transmission channel, thereby reducing the footprint and energy cost.

One potent platform that can cover such functionalities to great success are metasurfaces. With the introduction of the generalized Snell's law of reflection and refraction introduced by Yu et al. [63], the field of metasurfaces providing abrupt local phase changes opened up. By designing linear phase gradients from such structures, the capability to steer beams has quickly covered anything from visible to microwave frequencies [55]. Additionally, high quality factor metasurfaces already allow for high harmonics generation in the optical and NIR regime [64]. In 2025, Sanjari and Aflatouni presented a nonlinear active metasurface that down-converts a pre-modulated 193 THz (1550 nm) input wave into the contained 28

GHz wave at a 2 Gb/s data rate, bridging the gap between fiber-based and free space optical transmission with a single active device [7]. Down-conversion is done via the intensity dependent photocurrent in the photodiodes, allowing extraction of the applied mm-wave modulation and beam steering is enabled by on-chip post-processing of the generated signal.

THz communication nicely illustrates the potential of multi-functional beam manipulation. Further miniaturization allows a transfer of these concepts into the NIR and visible regime, allowing LiDAR systems or augmented reality devices to profit from it. And one well established way to realize said beam manipulation is the geometrical phase. By introducing birefringent nano-structures - designed as miniature half-wave plates - and using circular polarized light, the local phase change is dictated by the structure's in-plane rotation. This concept works broadband and allows for easy design of metasurfaces as only one nano-structure design is required to cover a full $0 - 2\pi$ phase range. In 2021, Xie et al. analyzed the linear geometrical phase terms arising from various rotationally symmetric nano-structures placed in square and hexagonal lattices [65]. Additionally, prior research by Li et al. in 2015 described the structures without coupling effects and established selection rules on allowed geometrical phase terms depending on different generated harmonics and the rotational symmetry of a meta-atoms [66].

The following work is an extension of Li et al.'s and Xie et al.'s research. By considering higher harmonic order selection rules through a wave-mixing picture and combining it with the lattice dependence, this chapter expands upon a popular method to impart local phase changes. The derived wave-mixing picture can be combined with findings concerning the linear geometrical phase in square or hexagonal lattices in a modular manner, creating additional phase factors. The validity of the model is confirmed by fabrication and characterization of reflective, hybrid, gradient metasurfaces that mainly generate third harmonic.

4.2 Theory

Since this chapter will combine the geometrical phase with nonlinear generation as well as different lattice symmetries, we will have a look into different "layers" of the geometrical phase. First, I want to cover how the linear and nonlinear geometrical phase for individual structures with n -fold rotational symmetry were previously described, thereby referring to Li et al.'s work [66, 67]. After that, we will have a look into how square and hexagonal lattice layouts can impact the applied geometrical phase in the linear regime, which was nicely shown by Xie et al. [65]. And lastly, we will have a look at a more rigorous explanation on what nonlinear phase factors can arise from induced nonlinear polarization [68]. The latter was derived within the confines of this work (theoretical description done by Bingyi Liu) and can be interpreted as an extension of the nonlinear geometrical phase described in [66]. Additionally, in simulations and experiments, it will be combined with the concept of [65] by realizing square and hexagonal lattice configurations.

4.2.1 Single nano-structure nonlinear geometrical phase generation

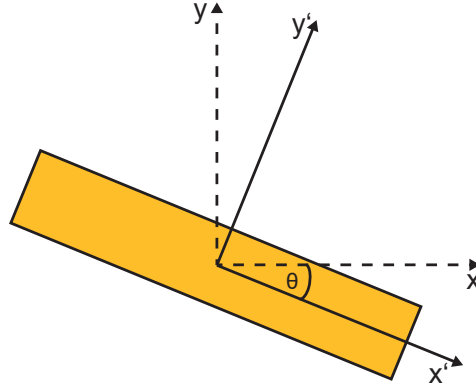


Figure 4.1: C2 nano-structure rotated by an angle θ in a laboratory reference frame (x, y) and a local reference frame (x', y') . [recreated from Li et al. [66]]

The geometrical phase arises from an interaction between a rotated, n -fold rotational symmetric structure and a circularly polarized electric field. The derivation of the expected and allowed geometrical phase contributions described in the following is taken from Li et al. [66]. They consider a nano-structure rotated by an angle θ inside a laboratory (x, y) / local (x', y') reference frame (see figure 4.1) excited by an electric field with helicity σ . Here, σ is 1 (-1) for LCP (RCP). We can describe the electric field inside the local/rotated coordinate system via

$$E'^{\sigma} = E^{\sigma} e^{i\theta\sigma}. \quad (4.1)$$

E' refers to the electric field in the (x', y') -coordinate system. By considering h electric field contributions for a harmonic generation of order h , the nonlinear polarization is given by

$$\mathbf{P}'^{(h)} = \chi_0^{(h)} (E^{\sigma} e^{i\theta\sigma})^h. \quad (4.2)$$

$\chi_0^{(h)}$ is the susceptibility tensor of the structure in its (unrotated) local coordinate system. The induced nonlinear polarization in the local coordinate system can be decomposed into two orthogonal circular polarization states $\pm\sigma$

$$\mathbf{P}'^{(h)} = \mathbf{P}'^{\sigma, (h)} + \mathbf{P}'^{-\sigma, (h)} \quad (4.3)$$

with each of these terms carrying the phase $e^{ih\sigma\theta}$. For transmission type structures, the σ ($-\sigma$) contribution is called co- (cross-) polarization and the transformation back into the laboratory reference frame is done by multiplying with $e^{-i\sigma\theta}$ ($e^{i\sigma\theta}$) [66]. Moving the

generated phase terms to the susceptibility tensor, we get:

$$\chi_{\theta,\sigma,\sigma}^{(h)} = \chi_{0,\sigma,\sigma}^{(h)} e^{i(h-1)\sigma\theta} \quad (4.4)$$

$$\chi_{\theta,-\sigma,\sigma}^{(h)} = \chi_{0,-\sigma,\sigma}^{(h)} e^{i(h+1)\sigma\theta} \quad (4.5)$$

χ_θ refers to the susceptibility including rotation operations by an angle θ into the local reference frame and back into the laboratory frame. This expression indicates, that co- and cross-polarization acquire different geometrical phase terms and, as of now, contain no considerations regarding different rotational symmetric structures. However, we can include a symmetry relation from Neumann's principle to derive selection rules for higher harmonics of order h with respect to the n -fold rotational symmetry of the nano-antenna [69]. Note that the optical properties of the n -fold rotational symmetric structure have to be identical at rotation angle of $2\pi/n$ and 0° . We can hence infer, that the argument of the exponential function in 4.5 must be an integer multiple of 2π for a rotation of $\theta = 2\pi/n$:

$$(h \mp 1) \sigma \frac{2\pi}{n} = j \cdot 2\pi. \quad (4.6)$$

$-(+)$ refers to co- (cross-) polarization. Neglecting σ and rewriting this expression, we get polarization dependent selection rules for nonlinear generation

$$h = j \cdot n \pm 1. \quad (4.7)$$

j is an integer > 0 and $+(-)$ corresponds to a solution for co- (cross-) polarization. Taking $h = 3$ and $n = 4$ as an example, this expression is solved at $j = 1$ ($3 = 1 \cdot 4 - 1$) giving a solution to cross-polarization but does not yield a solution for the “+1” part. Hence, a C4 structure only generates third harmonic in opposite helicity but not for co-polarization. As for the phase factor, this is given by equation 4.5. A couple of evaluated solutions from the last two expressions are collected in table 4.1. Empty entries mean that no solution to equation 4.7 exist.

Table 4.1: Linear geometrical phase for C1-C4 rotational symmetric structures. The row with “+” refer to co-polarization and “-” to the cross-polarization phase terms. [66]

Harmonic order	polarization	C1	C2	C3	C4
h = 1	co				
	cross	$2\theta\sigma$	$2\theta\sigma$		
h = 2	co	$1\theta\sigma$			
	cross	$3\theta\sigma$		$3\theta\sigma$	
h = 3	co	$2\theta\sigma$	$2\theta\sigma$		
	cross	$4\theta\sigma$	$4\theta\sigma$		$4\theta\sigma$
h = 4	co	$3\theta\sigma$		$3\theta\sigma$	
	cross	$5\theta\sigma$			

Taking the linear regime as an example, we can see that structures with rotational symmetries of $n \geq 3$ do not apply a θ -dependent phase and are therefore considered isotropic. However, higher harmonics can exhibit nonlinear geometrical phase terms for $n \geq 3$, implying that the different harmonics are decoupled and do not necessarily require an existing linear geometrical phase for a certain n .

4.2.2 Square and hexagonal lattice symmetries

This subsection will go into more detail on the relation between the rotational symmetry of an individual meta-atom and the resulting lattice symmetry for which optical properties repeat in the linear regime. As we can see from table 4.1, for $h = 1$ only the cross-polarization channel allows for a θ -dependent geometrical phase. The following description, taken from Xie et al. [65], thus only focuses on generated cross-polarization.

While a structure can be rotated up to 360° geometrically, for a C2-structure, optical properties already repeat after a rotation of 180° . Similarly, the optical properties of higher order rotational symmetric structures repeat after $\frac{360^\circ}{n}$. Xie et al. therefore differentiate between a purely geometrical rotation angle θ and the resulting effective rotation angle θ_{eff} . Collectively rotating a 2D-array of meta-atoms by an angle θ in their respective unit cell results in an effective rotation of the system's optical axis by an integer multiple $\theta_{\text{eff}} = l \cdot \theta$ which translates, with a factor of 2 because of the cross-polarization (see equation 4.5), to the linear geometrical phase $\Phi_{\text{geom}} = 2l\theta$. Xie et al. illustrate the magnitude at which the linear geometrical phase for various rotational symmetric structures in a square and hexagonal lattice changes when the nano-antennas are rotated. Using this approach, contrary to the selection rules from the previous subsection given by equation 4.7, the authors also derive geometrical phase factors for rotational symmetries $n \geq 3$.

Their simulated structures consist of thin gold films placed on a SiO_2 substrate. Air-holes in the gold film, e.g. equilateral triangle-shaped holes, make up the rotational symmetric structures. They simulated and phenomenologically illustrated the respective phase factors l by observing at which rotation angle the entire lattice appears as a rotated version of itself, but aligned to a different lattice axis. In addition to that, they experimentally verify the determined phase factors for C2, C3 and C5 structures in a square lattice configuration by generating vortex beams from the encoded geometrical phase in a metasurface. They define γ as the minimal meta-atom rotation angle necessary to rotate the optical axis of the entire lattice by a certain angle α . Slightly rewriting their expression gives:

$$\gamma(\alpha, n) = \alpha - \frac{360^\circ}{n} \left\lceil \frac{\alpha}{360^\circ} \cdot n \right\rceil. \quad (4.8)$$

$\lceil \cdot \rceil$ is the nearest integer function and n is the rotational symmetry of the meta-atom. Note that α is chosen in such a way, that another lattice-axis is addressed. For a square lattice, that means $\alpha = \pm 90^\circ$, whereas for the hexagonal lattice, rotations around $\pm 60^\circ$ and $\pm 120^\circ$ are both valid. The minimal rotation angle γ_{hex} necessary in order to rotate the optical axis

from one hexagonal lattice axis to another is thus given by

$$|\gamma_{\text{hex}}| = \min(|\gamma(60^\circ, n)|, |\gamma(120^\circ, n)|) . \quad (4.9)$$

For a square lattice, equation 4.8 gives $\gamma_{\text{sq}} = 0^\circ$ at $\text{mod}(n, 4) = 0$. In these cases, the structure optically behaves the same along every lattice axis and is therefore isotropic. Similarly, for a hexagonal lattice, isotropic behavior is expected for $\text{mod}(n, 3) = 0$. For all other structures, the scaling factor l can be determined by dividing the angle α by the antenna rotation angle γ . The geometrical phase $\Phi_{\text{geom, sq}}$ of n -fold symmetric structures in a square lattice depending on the geometric rotational angle θ therefore is

$$\Phi_{\text{geom, sq}} = 2\theta \cdot \frac{\pm 90^\circ}{\gamma} . \quad (4.10)$$

Note that the factor of 2 comes from the transformation into the rotated coordinate system and back, or in other words: If the systems optical axis was rotated by 180° , the corresponding geometrical phase is 360° . In general, numerical simulations are necessary in order to determine the sign of the geometrical phase [65].

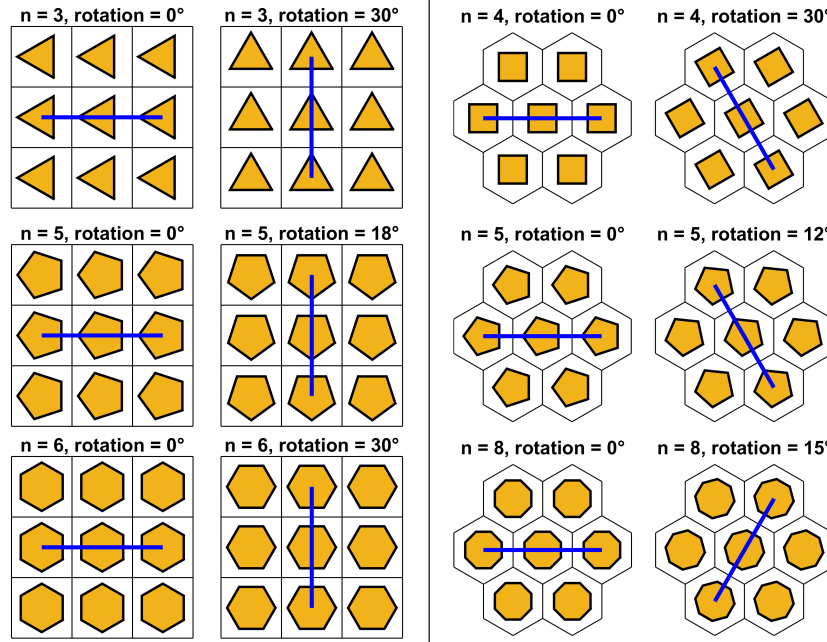


Figure 4.2: Illustration of lattice-similarity of $n = [3, 5, 6]$ (square lattice, left) and $n = [4, 5, 8]$ (hexagonal lattice, right) rotational symmetric structures for different ccw rotation angles $\theta = [0^\circ, \gamma_{\text{sq/hex}}]$. The blue line corresponds to an optical axis of the entire lattice, which is rotated by $\pm 90^\circ$ (square lattice) or $\pm 60^\circ / \pm 120^\circ$ (hexagonal lattice) for the right panel of each structure with respect to the left panel. Note that despite the ccw rotation of the individual structures, it can appear as if the entire 2D-array was rotated cw. [recreated from Xie et al. [65]]

The re-occurrence of lattice-similarity in square and hexagonal lattices for different n -fold rotational symmetric structures is illustrated in figure 4.2. Taking equation 4.8, the ccw rotation angles $\theta = [0^\circ, \gamma]$ are shown for $n = [3, 5, 6]$ for the square lattice and $n = [4, 5, 8]$ for the hexagonal lattice. The blue line in the lattices represent an optical axis and its rotation resulting from the angle γ . For the e.g. $n = 3$ structures in the square lattice configuration, the optical axis is rotated by $\pm 90^\circ$ while the triangles are only rotated by 30° , giving $l = 3$ and $\Phi_{\text{geom, sq}} = 2\theta_{\text{eff}} = 2l\theta$. While it is clear for the square lattice that the optical axis rotation of the lattice is $\pm 90^\circ$, in the case of the hexagonal lattice this is more ambiguous. Take the $n = 4$ rotational symmetry at 0° and 30° as an example; the optical axis rotation can either correspond to a 120° or -60° rotation which makes a difference in the generated geometrical phase factor's sign and magnitude. The optical axis rotations for e.g. both the $n = 4$ and $n = 5$ structure in the hexagonal lattice were each identified as a 120° rotation by Xie et al., whereas all other presented structures in the hexagonal lattice showed a $|\alpha| = 60^\circ$ rotation.

Taking all evaluations from Xie et al., the geometrical phase factors $2l$ for C1-C10 structures in square and hexagonal lattices are summarized in table 4.2.

Table 4.2: Linear geometrical phase factors for C1-C10 rotational symmetric structures in square and hexagonal lattices. Note that all phase factors correspond to generated cross-polarization. [65]

Lattice	C1	C2	C3	C4	C5	C6	C7	C8	C9	C10
Square	$2\theta\sigma$	$2\theta\sigma$	$6\theta\sigma$		$-10\theta\sigma$	$-6\theta\sigma$	$-14\theta\sigma$		$18\theta\sigma$	$10\theta\sigma$
Hexagonal	$2\theta\sigma$	$2\theta\sigma$		$8\theta\sigma$	$20\theta\sigma$		$14\theta\sigma$	$8\theta\sigma$		$-10\theta\sigma$

With respect to the square lattice, they identified the following trend for the geometrical phase:

$$\Phi_{\text{geom, sq}} = \begin{cases} \pm 2n\theta & \text{if } n \text{ is odd,} \\ \pm n\theta & \text{if } n \text{ is even,} \\ 0 & \text{if } \text{mod}(n, 4) = 0. \end{cases} \quad (4.11)$$

While not stated in their work, it almost seems as if the sign of the phase changes whenever a rotational symmetry is passed which does not carry a geometrical phase. In contrast to that pattern, the hexagonal lattice mostly shows the same absolute values as the square lattice but differs for the C5 structure. Here, no strictly alternating sign is observed when multiples of $n = 3$ are passed.

Let's recall that the identified rotation of the optical axis for the C4 and C5 structure in the hexagonal lattice by Xie et al. was 120° which was confirmed by their simulations. While just an assumption: If that behavior were to only appear because of a specific parameter combination and would otherwise yield -60° instead, the sign and magnitude of these two entries would change. This way, the alternating sign pattern would appear again and

the absolute values of the $2l$ -phase factors would follow the same alternating pattern as equation 4.11 but evaluating as 0 on $\text{mod}(n, 3) = 0$, instead. With these results, the observed angle γ would in all presented cases be $\pm 60^\circ$, taking the nearest/smallest possible rotation angle.

4.2.3 Pancharatnam-Berry-Phase

A more general description of the geometrical phase was demonstrated within the confines of this work by Bingyi Liu. The following description provides rotational symmetry-based selection rules for linear geometrical phase terms in a forward- and backward-propagating basis representation. It further addresses nonlinear wave mixing processes inside the nano-structures and is applicable to transmission and reflection type structures. I will outline the most important steps of the formulation but more details can be found in our corresponding publication in [68].

The overall formulation considers two sets of basis vectors representing the forward and backward circular polarization representation (FCPR and BCPR) while also transforming an e.g. incident electric field vector E into the nano-antenna's rotated frame of reference. The unit vectors refer to fixed cartesian unit vectors (e_x, e_y, e_z) and are expressed as:

$$e_L^\pm = \frac{e_x \pm ie_y}{\sqrt{2}}, \quad e_R^\pm = \frac{e_x \mp ie_y}{\sqrt{2}}, \quad e_z^\pm = \pm e_z. \quad (4.12)$$

Here, + and - refer to the FCPR and BCPR, respectively. Note that the definition of LCP and RCP changes owing to the change in propagation direction. A representation of an electric field vector E in the rotated and unrotated reference frames is given by:

$$E = \sum_{\alpha} {}^{\pm} E^{\alpha} e_{\alpha}^{\pm} = \sum_{\alpha'} {}^{\pm} E^{\alpha'} e_{\alpha'}^{\pm}, \quad (4.13)$$

α represents the circular polarization representation (CPR), i.e. 1, -1 and 0 for L, R and z, respectively. The primed CPR corresponds to an already rotated frame of reference in accordance with the nano-antenna orientation. If the CPR is superscripted it implies the contravariance of the respective vector whereas a subscripted CPR corresponds to covariant vectors. The primed reference-frame corresponds to a rotation by an angle θ_{eff} that represents the meta-atom's effective principle optical axis rotation (not just the geometric rotation of the structure) and is described by:

$$\begin{bmatrix} e_{L'}^{\pm} \\ e_{R'}^{\pm} \\ e_{z'}^{\pm} \end{bmatrix} = \begin{bmatrix} e^{\pm i\theta_{\text{eff}}} & 0 & 0 \\ 0 & e^{\mp i\theta_{\text{eff}}} & 0 \\ 0 & 0 & 1 \end{bmatrix} \cdot \begin{bmatrix} e_L^{\pm} \\ e_R^{\pm} \\ e_z^{\pm} \end{bmatrix}, \quad \begin{bmatrix} {}^{\pm} E^{L'} \\ {}^{\pm} E^{R'} \\ {}^{\pm} E^{z'} \end{bmatrix} = \begin{bmatrix} e^{\mp i\theta_{\text{eff}}} & 0 & 0 \\ 0 & e^{\pm i\theta_{\text{eff}}} & 0 \\ 0 & 0 & 1 \end{bmatrix} \cdot \begin{bmatrix} {}^{\pm} E^L \\ {}^{\pm} E^R \\ {}^{\pm} E^z \end{bmatrix}. \quad (4.14)$$

Note that the rotation around the z-axis for co- and contravariant vectors is performed in

opposite directions. This gives the general relation:

$$\mathbf{e}_{\alpha'}^{\pm} = \sum_{\alpha} {}^{\pm}R_{\alpha'}^{\alpha} \mathbf{e}_{\alpha}^{\pm} = e^{\pm i\alpha\theta_{\text{eff}}} \delta_{\alpha'\alpha} \mathbf{e}_{\alpha}^{\pm} \quad (4.15)$$

$${}^{\pm}E^{\alpha'} = \sum_{\alpha} {}^{\pm}R_{\alpha}^{\alpha'} {}^{\pm}E^{\alpha} = e^{\mp i\alpha\theta_{\text{eff}}} \delta_{\alpha'\alpha} {}^{\pm}E^{\alpha} \quad (4.16)$$

With R as the corresponding rotation matrix from equation 4.14 and δ as Kronecker delta owing to the fact that the off-diagonal elements in the rotation matrix are zero. We furthermore have the two relations

$${}^{\pm}R_{\alpha'}^{\alpha} = \left[{}^{\pm}R_{\alpha}^{\alpha'} \right]^{\dagger} \quad (4.17)$$

$${}^{\pm}R_{\alpha}^{\alpha'} = \left[{}^{\mp}R_{\alpha'}^{\alpha} \right]^{\dagger} \quad (4.18)$$

with the dagger-superscript as the conjugate transpose of the matrix. We can now describe the polarization P in the rotated frame of reference via

$${}^{\pm}P^{\alpha'} = \sum_{\alpha} {}^{\pm}R_{\alpha}^{\alpha'} {}^{\pm}P^{\alpha} = \epsilon_0 \sum_{\alpha,\beta} {}^{\pm}R_{\alpha}^{\alpha'} \chi_{\alpha\beta}^{(1)} {}^{\pm}E^{\beta} \quad (4.19)$$

By using equation 4.13 and relation 4.17, we can rewrite ${}^{\pm}E^{\beta}$ as

$${}^{\pm}E^{\beta} = \sum_{\beta'} \left[{}^{\mp}R_{\beta}^{\beta'} \right]^{\dagger} {}^{\pm}E^{\beta'} \quad (4.20)$$

Inserting this expression into equation 4.19, we get

$${}^{\pm}P^{\alpha'} = \epsilon_0 \sum_{\beta'} \chi_{\alpha'\beta'}^{(1)} {}^{\pm}E^{\beta'} \quad (4.21)$$

with

$$\chi_{\alpha'\beta'}^{(1)} = \sum_{\alpha,\beta} {}^{\pm}R_{\alpha}^{\alpha'} \chi_{\alpha\beta}^{(1)} \left[{}^{\mp}R_{\beta}^{\beta'} \right]^{\dagger}. \quad (4.22)$$

By incorporating the rotation matrices and evaluating $\chi^{(1)}$ for different circular polarization projections, we get the linear geometrical phase terms. For example, LCP incidence and RCP reflection and LCP incidence with LCP reflection (now expressed in the BCPR), give

$$\chi_{R'L'}^{(1)} = {}^{-}R_R^{R'} \chi_{RL}^{(1)} \left[{}^{+}R_L^{L'} \right]^{\dagger} = \chi_{RL}^{(1)} \quad (4.23)$$

$$\chi_{L'L'}^{(1)} = {}^{-}R_L^{L'} \chi_{LL}^{(1)} \left[{}^{+}R_L^{L'} \right]^{\dagger} = \chi_{LL}^{(1)} e^{2i\theta_{\text{eff}}}. \quad (4.24)$$

Here we can see, that a polarization conversion from LCP to RCP does indeed correspond to co-polarization that does not carry the geometrical phase term. θ_{eff} contains information on the rotational symmetry of the meta-atom and the lattice, as discussed in the previous

subsection.

In a similar fashion, we can give an expression for the nonlinear polarization corresponding to THG.

$$\pm P^{\alpha'}(3\omega) = \epsilon_0 \sum_{\beta', \gamma', \delta'} \chi_{\alpha' \beta' \gamma' \delta'}^{(3)} \pm E^{\beta'}(\omega) \pm E^{\gamma'}(\omega) \pm E^{\delta'}(\omega) \quad (4.25)$$

and

$$\chi_{\alpha' \beta' \gamma' \delta'}^{(3)} = \sum_{\alpha, \beta, \gamma, \delta} \pm R_{\alpha}^{\alpha'} \chi_{\alpha \beta \gamma \delta}^{(3)} \left[\mp R_{\beta}^{\beta'} \right]^{\dagger} \left[\mp R_{\gamma}^{\gamma'} \right]^{\dagger} \left[\mp R_{\delta}^{\delta'} \right]^{\dagger}. \quad (4.26)$$

Once established, one can see, that the derivations above follow a simple pattern. Thus, to summarize these derivations, we can generalize the geometrical phase for h^{th} -harmonic generation for a transmissive/reflective-type metasurface in dependence of the effective optical axis rotation θ_{eff} as:

$$\Phi(h, m, \sigma, \theta_{\text{eff}}) = \pm[2m(\pm\sigma) - (h + 1)] \cdot \sigma \theta_{\text{eff}}. \quad (4.27)$$

$+$ ($-$) represents solutions to the geometrical phase for a transmissive (reflective) type metasurface. m refers to the number of fundamental wave contributions of helicity $\pm\sigma$ (depending on whether the structure is transmissive or reflective), excluding the generated output helicity. This means, that m ranges from zero to h contributions $m = [0, \dots, h]$. σ is the generated helicity and equals $+1$ for left- and -1 for right-circular polarization. E.g. when looking for allowed geometrical phase factors of a reflective type metasurface that generates right circular polarization at the 1st-/2nd- and 3rd-harmonic, we get the expressions:

$$\begin{aligned} \Phi(h = 1, m, \sigma = -1, \theta_{\text{eff}}) &= [-2, 0] \cdot \theta_{\text{eff}} \\ \Phi(h = 2, m, \sigma = -1, \theta_{\text{eff}}) &= [-3, -1, 1] \cdot \theta_{\text{eff}} \\ \Phi(h = 3, m, \sigma = -1, \theta_{\text{eff}}) &= [-4, -2, 0, 2] \cdot \theta_{\text{eff}} \end{aligned}$$

I want to emphasize again that this yields a pool of possible solutions, however θ_{eff} can still be 0 because of the lattice symmetry. The derived expression can be seen as a modular extension to Xie et al.'s work, in the way that it provides allowed phase factors for the nonlinear geometrical phase that θ_{eff} can be multiplied with.

With the current description, the meaning of co- and cross-polarization becomes less clear because we consider varying linear electric field contributions of helicity $+\sigma/-\sigma$ *inside* the meta-atom. The participating electric fields inside the structure might not necessarily exhibit the same helicity as the incident fundamental beam. To avoid confusing, co- and cross-polarization will refer to the relation between incident and generated helicity; in a reflective type setting, generation of the opposite (the same) helicity is considered co- (cross-) polarization.

4.3 Design

To test the formulated theory, this work covers square lattice configurations of C2, C3 and C4 structures as well as C2 and C4 structures in hexagonal lattices. The structural parameters are optimized using a finite element method (FEM) solver in COMSOL Multiphysics' wave optics module (performed by Bingyi Liu). A schematic of a few respective unit cells for a square and a hexagonal lattice is shown in figure 4.3.

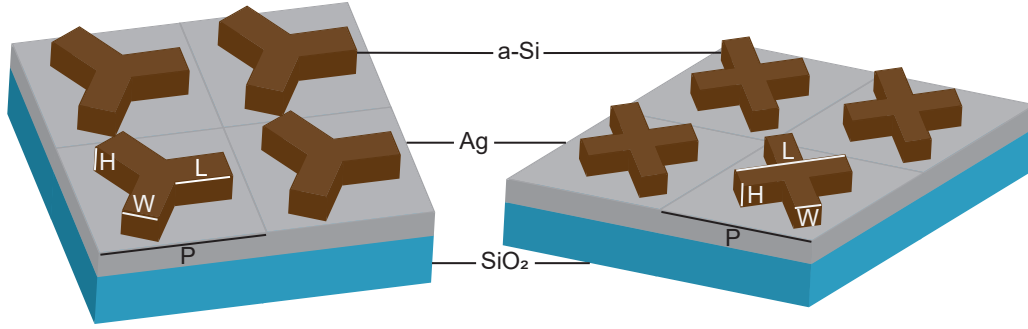


Figure 4.3: Unit cell design for COMSOL simulations. The nano-antennas consist of amorphous silicon and are designed by multiple rotated cuboids with length L , width W and height H . The antennas are placed on a layer system of 200 nm Ag and 200 nm silica glass substrate. Structural periodicity is realized with periodic boundary conditions and the THG intensity/phase with respect to the individual antennas rotation angle are analyzed/optimized.

The unit cell consists of rotationally symmetric amorphous silicon based structures, exposed to air and placed on a 200 nm thick Ag layer followed by 200 nm of SiO_2 substrate. The different rotational symmetries of the antennas resemble multiple combined rectangular nano-rods with length L , width W and height H with equidistant rotation angles where the C2 and C4 structures consist of 1 and 2 nano-rods and the C3 structures of 3 nano-rods, respectively. Periodic boundary conditions are applied to the square and hexagonal lattice borders of the unit cell and the fundamental wavelength is set to 1250 nm at an electric field amplitude of $E = 10^8 \frac{\text{V}}{\text{m}}$ which corresponds to an intensity of $I = 1.33 \frac{\text{GW}}{\text{cm}^2}$. Additionally, sharp corners of the structures are rounded off using the fillet geometry function in order to avoid diverging field amplitudes. After propagating left circular polarized light normally into the simulation domain, the resulting electric field distribution in the meta-atoms in combination with the third-order nonlinear susceptibility of amorphous silicon (assumed to be a constant scalar with $\chi_{a-Si}^{(3)} = 2.45 \times 10^{-19} \frac{\text{m}^2}{\text{V}^2}$ [70]) are used to derive the third-order nonlinear polarization $\mathbf{P}^{(3)}(\mathbf{r}, 3\omega) = \epsilon_0 \chi_{a-Si}^{(3)} \mathbf{E}^3$. In a second simulation step, the efficiency of the radiated TH is determined on the input port and the phase-behavior in dependence of the meta-atom's rotation angle is evaluated. Table 4.3 shows a list of the parameters chosen for fabrication. Note that the meta-atom height is fixed at 105 nm.

Table 4.3: Structural parameters of structures analyzed in this work. The height of the nano-antennas is fixed at 105 nm.

	square lat.			hexagonal lat.	
	C2	C3	C4	C2	C4
L [nm]	300	132	330	330	330
W [nm]	200	130	130	130	130
P [nm]	400	390	400	430	430
λ [nm]	1250	1200	1250	1250	1200

The simulated phase response for co- and cross-polarization of the different structures is shown in figure 4.4. Evaluating the phase increase over rotation angle gives the integer scaling factor and sign of the nonlinear geometrical phase which is shown in the legend of the respective panel. Note that the C3 structure shows a very low but periodic phase coverage in the cross-polarization channel, indicating a $\pm 12\theta$ dependence. For the C4 structure in a square lattice the co-polarized THG seems to be suppressed and no phase modulation is visible. Since no reasonable phase response for the co-polarized TH signal of these C4 structures is visible, figure 4.5 shows the electric field amplitude near the meta-atom and contrasts the radiated plane wave of the cross-polarization to the suppressed co-polarized response. Additionally, the phase response of the C2 structures placed in a hexagonal lattice (simulated by me) fall short of covering the entire 2π -phase range and show less linear behavior than the simulated data from Bingyi Liu. This might stem from unoptimized mesh-settings but roughly shows the same phase behavior that was predicted by Bingyi Liu for these structures [68]. Note that non-linear scaling of the geometrical phase over rotation angle can come from a variation of meta-atom distances and thus different coupling between the unit cells [71]. This effect goes hand in hand with a variation of TH conversion efficiency over rotation angle (see the cross-polarized signal in figure 4.5).

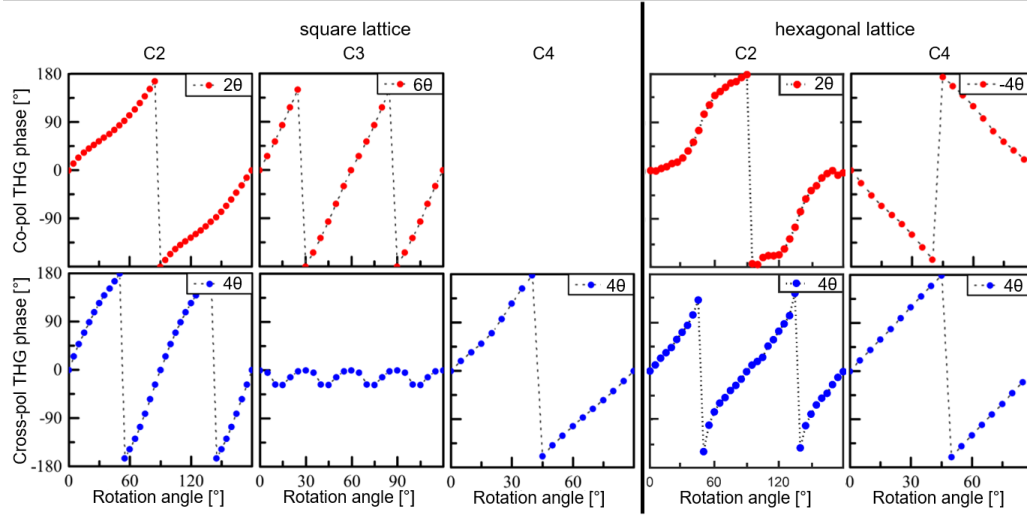


Figure 4.4: Simulated THG phase response based on the structural parameters listed in table 4.3 plotted over rotation angle θ . The top and bottom row refer to co- and cross-polarization, respectively. The phase response for C2 structures in a hexagonal lattice is recreated by me whereas the other plots were generated by Bingyi Liu. Note that C3 structures in a square lattice do not cover the full 2π -range. Also, co-polarized TH signal from C4 structures in a square lattice (not shown) are suppressed, yielding very low TH response and no phase modulation. [adapted from [68]]

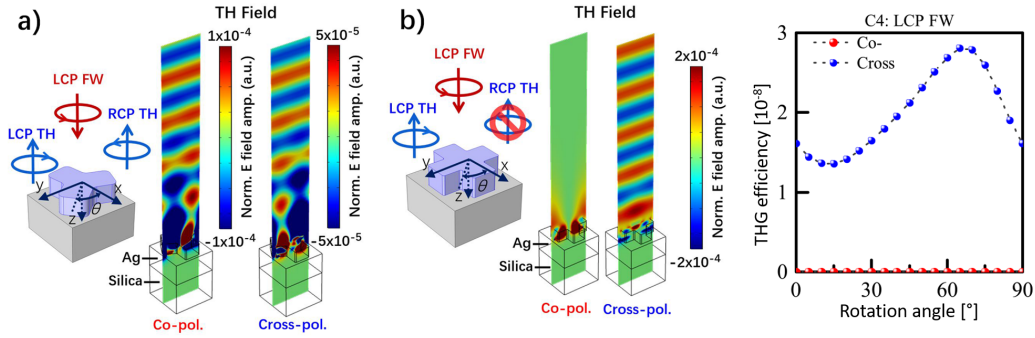


Figure 4.5: Normalized electric field amplitude of generated THG from an excited C3 a) and C4 b) structure placed in a square lattice. The emitted signal from the C3 structure results in a plane wave emission for both the co- and cross-polarization. On the other hand the co-polarization of the C4 structure is suppressed. The plotted TH efficiency over meta-atom rotation angle also shows negligible generation of co-polarized signal leading to no reasonable phase evaluation. [adapted from [68]]

By additionally simulating the linear phase response of these structures, we can determine the relation between effective and geometric rotation angle, analogous to table 4.2 from the previous subsection. The linear and third harmonic geometrical phases are collected in

table 4.4, including the undeveloped $\pm 12\theta\sigma$ of the C3 structure's THG cross-polarization channel.

Table 4.4: Linear and nonlinear geometrical phase factors for C1-C4 reflection-type structures in square and hexagonal lattices with structural parameters from 4.3.

Harmonic order	Lattice	polarization	C1	C2	C3	C4
h = 1	Square	co				
		cross	$2\theta\sigma$	$2\theta\sigma$	$6\theta\sigma$	
	Hexagonal	co				
		cross	$2\theta\sigma$	$2\theta\sigma$		$-4\theta\sigma$
h = 3	Square	co	$2\theta\sigma$	$2\theta\sigma$	$6\theta\sigma$	
		cross	$4\theta\sigma$	$4\theta\sigma$	$\pm 12\theta\sigma$	$4\theta\sigma$
	Hexagonal	co	$2\theta\sigma$	$2\theta\sigma$		$-4\theta\sigma$
		cross	$4\theta\sigma$	$4\theta\sigma$		$4\theta\sigma$

For the linear geometrical phase, the determined values mostly agree with those from table 4.2. However, the cross-polarization phase factor of the C4 structure in hexagonal lattice configuration contradicts the value reported by Xie et al. While they observe a factor of $8\theta\sigma$ in their research, we get $-4\theta\sigma$ indicating that this factor might depend on structural parameters.

Combining this effective rotation with the model derived for the multi-wave mixing process, we can find all the observed nonlinear geometrical phase factors in the set of solutions allowed from equation 4.27. For example, the co- and cross-polarized THG phase response from the C4 structure in the hexagonal lattice can be explained with the following electric field contributions:

$$\chi_{R'L'L'L'}^{(3)} = \chi_{RLLL}^{(3)} \exp(2i \cdot (-2\theta)) \quad (4.28)$$

$$\chi_{L'R'R'R'}^{(3)} = \chi_{LRRR}^{(3)} \exp(-2i \cdot (-2\theta)) \quad (4.29)$$

Importantly, different solutions from equation 4.27 can be accessed by varying the structural parameters that impacts the nonlinear wave-mixing in the structures. In order to enable the 2π -phase coverage of the C3 structure's cross-polarized THG, the height could be changed from 105 nm to 775 nm. Figure 4.6 illustrates this impact and also shows that while not only enabling said THG signal, it can also switch between different solutions of equation 4.27 which is shown for a C5 pentagonal-structure placed inside a square lattice with a height of 105 nm and 500 nm. The C5 structure's THG phase response is simulated at a fundamental wavelength of 1200 nm, a periodicity of 380 nm and a center-to-vertex distance of 180 nm. For both heights, we find a linear geometrical phase proportional to $\theta_{\text{eff}} = 5\theta$. With this relation, the nonlinear geometrical phase response of the 105 nm and

500 nm high structures, respectively, is interpreted as a wave-mixing process of:

$$\chi_{R'L'L'L'}^{(3)} = \chi_{RLLL}^{(3)} \exp(2i \cdot (5\theta)) \quad (4.30)$$

$$\chi_{R'L'R'R'}^{(3)} = \chi_{RLRR}^{(3)} \exp(-2i \cdot (5\theta)) . \quad (4.31)$$

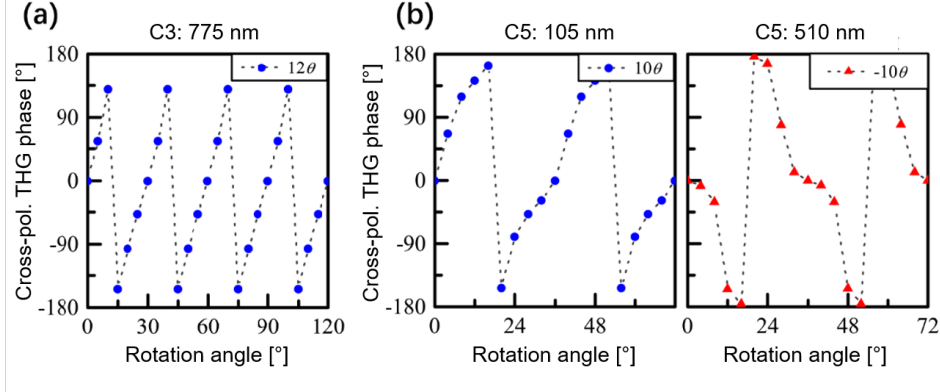


Figure 4.6: (a) Cross-polarized nonlinear geometrical phase over meta-atom rotation angle for an adapted C3 structure in a square lattice with a height of 775 nm instead of 105 nm. (b) Comparison of cross-polarized nonlinear geometrical phase of pentagon-shaped C5 structures in a square lattice at a height of 105 nm and 500 nm. The fundamental wavelength, center-to-vertex distance, and periodicity are set to 1200 nm, 180 nm and 380 nm, respectively. While not as well defined as other presented phase profiles, the data shows that structural parameters impact the dominant electric field contributions in the THG process leading to different phase-rotation angle relations. [adapted from [68]]

Let's evaluate these results with the previously established selection rule from equation 4.7. While its derivation is not tied to a particular lattice geometry, it predicts the presence of both co- and cross polarization of C2 structures as well as the cross-polarization of the C4 structure. Let's assume we can simplify the C4 structure in a square lattice as two individual C2 structures with 90° rotation between each other. By using the phase relation of C2 antennas from figure 4.4, an argument can be made that 90° rotation between the C2 antennas yields $4 \cdot 90^\circ = 0^\circ$ phase difference between the cross-polarized THG. However, 90° rotation results in 180° phase difference between the two co-polarized THG signals leading to destructive interference in the far-field. According to the selection rule, third harmonic from C3 structures is not allowed. Excited third order polarization does not couple to free space and dissipates as Joule heat in the vicinity of the meta-atom. Staying with the simplification of individual C2 antennas, a relative phase difference of 120° (co-polarization) and 240° (cross-polarization) between neighboring arms would lead to destructive interference for the THG in both cases. However, in contrast to the C4 structure, since the C3 symmetry does not match the square lattice symmetry, a non-uniform coupling between the three individual C2 antennas to neighboring unit cells can occur,

impacting their phase relation, enhancing optical anisotropy and enabling the radiation of THG to free-space. Furthermore, in our case, the coupling is strongly supported by the plasmonic layer underneath the silicon structures. Presumably, co-polarized THG signal from C4 structures might be enabled by slightly altering the lattice symmetry to rectangular unit cells. In the case of the hexagonal lattice, this symmetry mismatch between lattice and structure allows the emission of co-polarized THG.

In addition to offering a variety of accessible phase relations - provided that appropriate structural adjustments are performed - the simulated efficiency of these reflective type metasurfaces are also rather high mostly being in the range of $\eta_{\text{THG}} = \frac{P_{\text{THG}}}{P_{\text{FW}}} = 10^{-8}$ which is up to 3 orders of magnitude larger than similarly optimized transmission-type structures placed on an SiO_2 substrate [71]. Table 4.5 contains the simulated nonlinear conversion efficiencies for structures listed in figure 4.4.

Table 4.5: Calculated, averaged third order conversion efficiencies for co- and cross-polarization $\eta_{\text{THG, Co}}$ and $\eta_{\text{THG, Cross}}$.

	square lat.			hexagonal lat.	
	C2	C3	C4	C2	C4
$\eta_{\text{THG, Co}} [10^{-8}]$	6.5	0.35	-	/	0.48
$\eta_{\text{THG, Cross}} [10^{-8}]$	2	-	2	/	5.2

In order to experimentally verify the predicted behavior, it is necessary to differentiate between the different solutions that equation 4.27 allows for. A convenient method involves the realization of a linear phase gradient by rotating each meta-atom by an increasing rotation angle, depending on e.g. the x-coordinate on the to-be-fabricated metasurface. This way, the phase response is accessible by the resulting k-vector of the respective diffraction order, allowing comparison to the theoretical model. I want to emphasize one important distinction between the simulations and the phase gradient designs; by varying the rotation angle of the structures the coupling strength to neighboring antennas might also change. This could in principle lead to additional wave-mixing contributions from equation 4.27. This effect was not considered in our simulations, however in Xie et al.'s work they showed gradient type structures which exhibited the expected *linear* geometrical phase behavior as simulated for their strictly periodic designs [65].

4.4 Fabrication

A schematic of the fabrication can be seen in figure 4.7. First, a 3 nm chromium layer, followed by 100 nm silver and another 3 nm chromium layer are deposited via electron beam evaporation. The chromium layers are supposed to increase the adhesion from glass

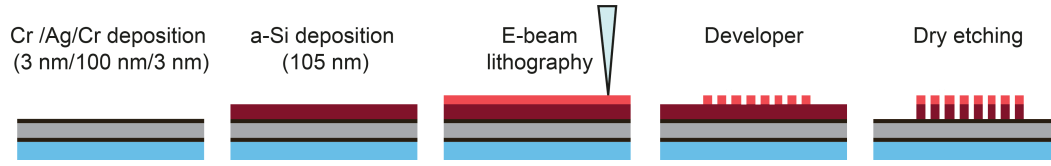


Figure 4.7: Fabrication workflow for the reflective-type nonlinear geometrical phase metasurfaces. A thin Cr layer below and on top of the Ag-layer is used as an adhesion layer. After deposition of 105 nm a-Si, a negative type ebeam-resist (maN-2401) is used during the electron ebeam lithography and serves as an etching mask to transfer the sample pattern into the a-Si layer.

to silver and silver to amorphous silicon, respectively. Afterwards, a 105 nm a-Si layer is deposited via PECVD. A negative resist (maN-2401) is spin coated onto the a-Si layer (30s, 3000 rpm, baking at 90° C for 2 minutes) and patterned using electron beam lithography (dose of $300 \frac{\mu C}{cm^2}$ at 50 kV acceleration voltage). The patterned files are dose corrected via a proximity effect correction (PEC) made in the Software Beamer to improve the shape of the structures. After developing the exposed maN-2401 in maD-525 for 20 seconds and rinsing in water, the remaining resist is used as a dry etching mask. The RIE etching process uses SF_6 (17 sccm) and C_4F_8 (35 sccm) as gases at a pressure of 10.5 mTorr with an RF- and ICP-power of 42 W and 900 W resulting in a DC-bias of 127 V. Several short etch steps are performed in order to avoid etching into the Ag film as the silver might deteriorate from sputtering processes or in the presence of sulfur-containing gases. Figure 4.8 shows SEM micrographs of some of the finished designs that were characterized in the following chapter. Additional information on the super lattice periodicity is also provided.

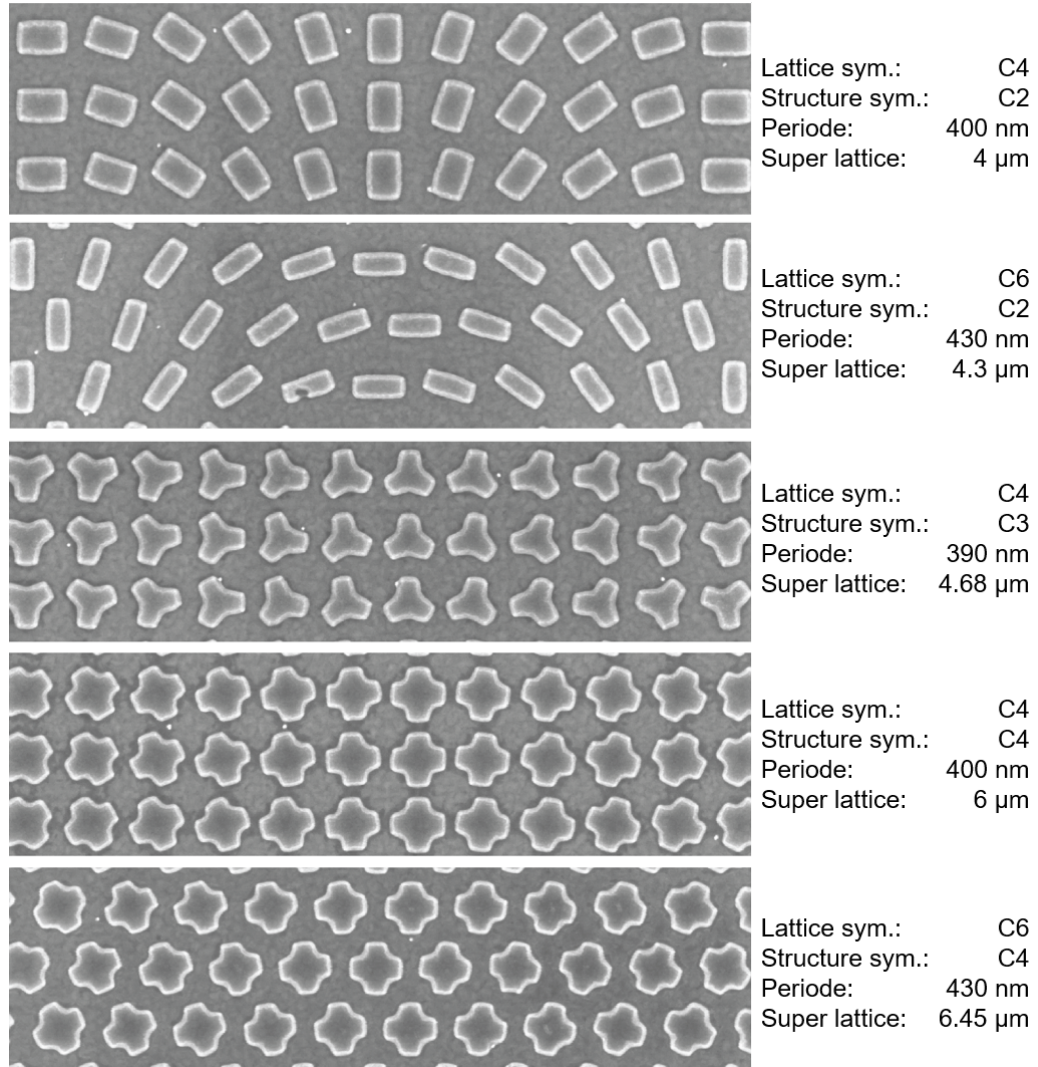


Figure 4.8: SEM micrographs from the different characterized metasurfaces of this work, covering several combinations of C4 and C6 lattices symmetries with C2, C3 and C4 nano-antenna symmetries.

4.5 Measurements and Results

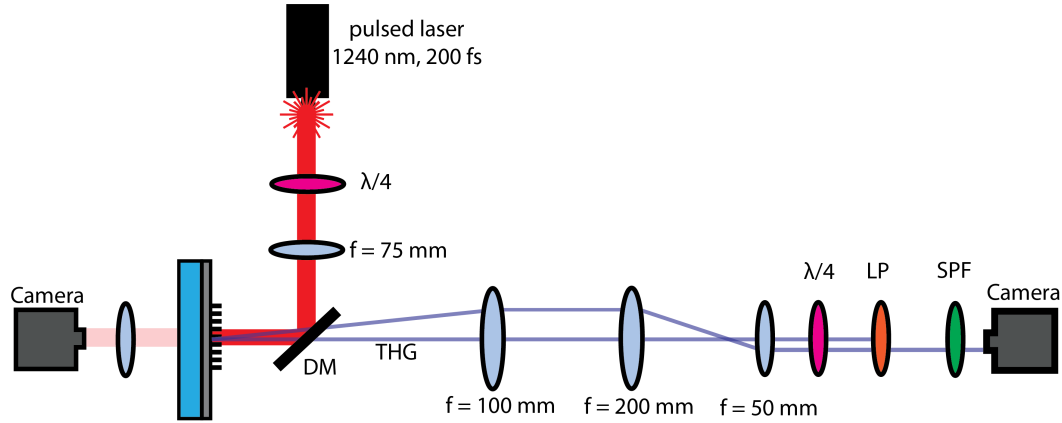


Figure 4.9: Top view of the optical setup. A dichroic mirror (DM) is used to allow illumination of the sample with the fundamental wavelength (varied from 1180-1300 nm) while transmitting the generated third harmonic. A collimation lens is used to allow imaging in the Fourier space (separating the diffraction orders) and an additional telescopic setup is necessary, to reduce the spatial separation of these diffraction orders to allow simultaneous imaging. Note that a smaller focal length of the collimating lens ($f = 100$ mm) is not possible because of limited space). The analyzer allows to filter different circular polarization states. [68]

The first iteration of the optical setup is shown in Figure 4.9. Since THG benefits from high peak powers, the setup includes a pulsed laser source with a repetition rate of 80 MHz (Coherent Chameleon Ultra II). The fundamental wavelength for the measurements is varied from 1180 - 1300 nm (the design wavelength of the samples are 1200/1250 nm). In this regime, the laser pulses have a temporal width of 200 fs (corresponding bandwidth of 8 nm). Since the samples rely on the geometrical phase to realize the diffraction gratings, a QWP is used to set the incident circular polarization state. The laser beam is focused onto the sample via a 75 mm focal length lens to achieve small spot sizes. Note that using the QWP twice, similar to the pulse shaping setup, is not advisable, as the operation wavelength changes from 1200 nm to 400 nm and does not guarantee clean polarization conversion outside of the indicated operation range (1100 - 2000 nm). A short pass dichroic mirror (DMSP950) is used, to guide the beam onto the sample. This has the benefit, that any generated TH can pass the dichroic mirror, allowing for easy collection of the signal without the use of a beamsplitter. Similar to the pulse shaping setup, an imaging system behind the silver mirror of the sample can be used, to ensure proper focusing onto the individual samples. The sample is mounted on an xyz-stage with an additional rotational mount allowing to set the pitch rotation. A lens with a focal length of 100 mm is used to collimate the beams and a telescopic setup with a magnification factor of 0.25 is used to minimize the spacing between the diffraction orders, allowing simultaneous imaging on a CCD camera (Andor Zyla). Optionally, another QWP (350 - 850 nm) and a linear polarizer are used to differentiate between the co- and cross- polarized components of the generated

signal. In order to filter out the remaining fundamental wavelength, a short pass filter (SPF) with a cutoff wavelength of 800 nm is used. Due to the imaging of the Fourier space, the diffraction angles from the samples have a linear distance relation to the non-diffracted beam position on the camera chip. To calibrate the setup, one could either calculate the scaling factor between the spatial coordinate and the diffraction angle by taking the used focal lengths into account, which might be rather inaccurate, especially since three lenses are used to collimate the signal. Or - since the metasurface is placed on a silver mirror - one can alter the sample's pitch angle using the rotational mount and evaluate the beam deflection on the CCD chip. With the relation between the incident and reflected angle $\theta_{in} = \theta_{out}$, a mirror rotation of 1° results in a beam deflection of 2° . These measurements are performed for a rotational mount setting from -3° to 3° in steps of 1° , leading to a beam deflection from -6° to 6° . By averaging over the resulting distance to the reflection under normal incidence (0° pitch), the scaling factor between camera pixel position and deflection angle is acquired, allowing comparison of the measured and expected diffraction angles.

By dividing the super lattice periodicity P_{SL} of each design with their respective nonlinear geometrical phase factor Φ_{nonlin} taken from figure 4.4, we can calculate the expected diffraction angles $\theta_{\text{co/cross}}$ via

$$\theta_{\text{co/cross}} = \text{asin} \left(\frac{\lambda_{\text{THG}} \Phi_{\text{nonlin, co/cross}}}{P_{SL}} \right). \quad (4.32)$$

Table 4.6 contains the super lattice periodicity of each design as well as the calculated/expected diffraction angles $\theta_{\text{co/cross}}$ at the respective TH design wavelength.

Table 4.6: Expected diffraction angles for co- and cross-polarization ($\theta_{\text{co/cross}}$) for all fabricated structures based off of their super lattice periodicity P_{SL} . Additionally fabricated P_{SL} and their diffraction angles are given in brackets.

	square lat.			hexagonal lat.	
	C2	C3	C4	C2	C4
$P_{SL} [\mu\text{m}]$	4 (8)	4.68 (9.36)	6	4.3 (8.6)	6.45
$\theta_{\text{co}} [^\circ]$	5.98 (2.99)	4.90 (2.45)	-	5.56 (2.78)	-3.7
$\theta_{\text{cross}} [^\circ]$	12.02 (5.98)	- (-)	3.98	11.17 (5.56)	3.7

The numerical aperture of the 2 inch diameter collection lens (focal length 100 mm) in the setup is $\text{NA} \approx 0.24$, equation to an angle of roughly 14° . This puts the largest expected diffraction angle of 12° close to the edge of that lens. In order to reduce potential imaging errors for large diffraction orders, the pitch of the sample is adjusted so that the undiffracted beam is de-centered from the collecting lens (not shown in figure 4.9). As a consequence, only one half of the k-space is imaged - with the exception of the C4 structure in the square lattice, which is measured with no pitch adjustments. Figure 4.10 shows the measured diffraction orders from the structures referenced in the previous table (excluding the C4

structure in the hexagonal lattice) at a fundamental wavelength of 1200 nm. In addition, a logarithmic plot of the detected signal - after summing over the individual rows of the images - is shown to improve the visibility of weaker diffraction orders.

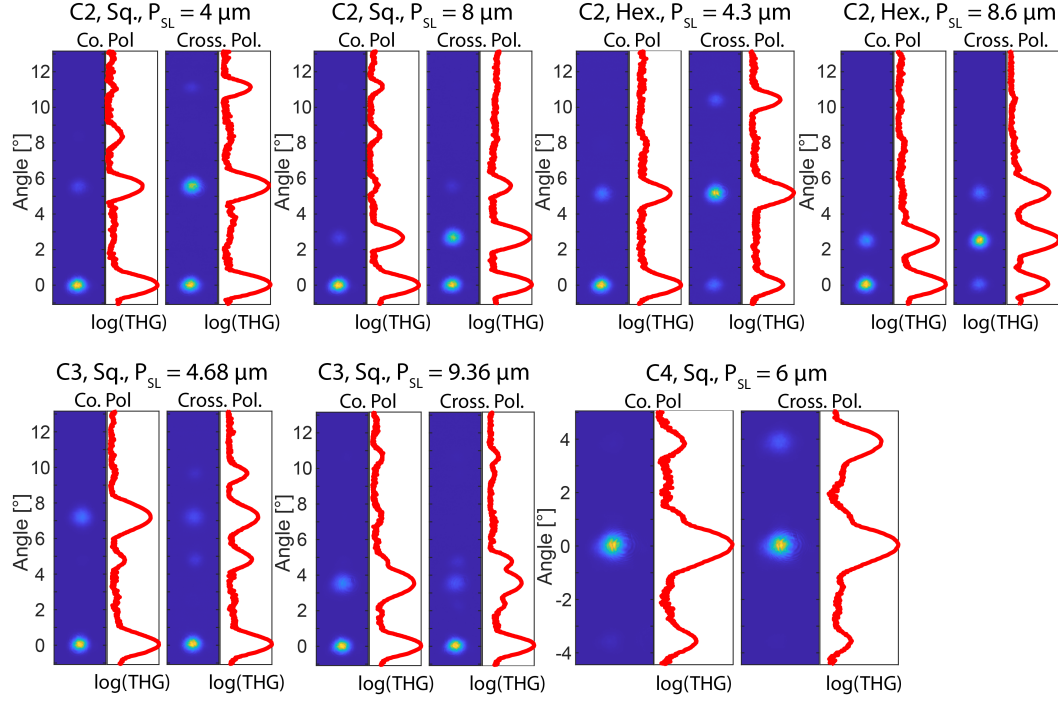


Figure 4.10: Measured diffraction orders from the structures referenced in table 4.6 (except for C4 structures in hex. lattice). The logarithmic plot improves the visibility of weaker diffraction orders. These measurements were conducted with a fundamental wavelength of 1200 nm.

For the C2 structures in square lattice with $P_{SL} = 4 \mu\text{m}$, the strongest diffraction orders can be found at 5.5° and 11.1° . Taking the deviation of the used fundamental wavelength into account (1200 nm instead of 1250 nm), we get expected diffraction angles of 5.74° and 11.54° instead, which is close to the measured values. These two values seem to indicate a relative deviation of $\approx 4\%$. This small deviation might come from an inaccurate calibration of the k-space or slight misalignment of the three lenses in between the sample and the camera. We can also see an unexpected diffraction order at around 8.3° - mainly in the detected co-polarization, visible in the logarithmic plot. Interestingly, this diffraction order is not an integer multiple of the “first”/smallest diffraction angle. Correcting the 8.3° by the $\approx 4\%$ deviation and considering $P_{SL} = 4 \mu\text{m}$ we can use equation 4.32:

$$\lambda = \sin(8.6^\circ) \cdot 4 \mu\text{m} = 598 \text{ nm} \quad (4.33)$$

indicating, that the measurements also include SHG. Aside from this, we see two diffraction orders for the cross-polarization instead of only the contribution at 11.1° . The co-polarized signal also shows two contributions; one at 5.5° which is expected and - besides the SHG

diffraction order - a small peak at 11.1° . A similar behavior can also be found for the C2 structures in square lattice with $P_{SL} = 8 \mu\text{m}$, as well as for both C2 designs in the hexagonal lattice.

The C3 structures with $P_{SL} = 4.68 \mu\text{m}$ show strong diffraction orders at 7.1° which is unexpected and also found to match an SHG diffraction angle. Two additional diffraction orders can be identified for the cross-polarized channel at 4.8° and 9.7° , whereas only the first was predicted by our model to appear in co-polarization. The larger diffraction angle could correspond to a $\Phi_{\text{nonlin}} = 12\theta$ -contribution, predicted in cross-polarization but for different structural C3-parameters as shown in figure 4.6. Smaller contributions at the same two angles can also be seen in the co-polarization measurement. The C3 structures with larger P_{SL} seem to behave in a similar manner.

Finally, the diffraction orders of the C4 structure in the square lattice are barely visible in the co-polarization channel, here shown for both halves of the k-space, at 3.8° and -3.6° . The asymmetric positioning of the diffraction orders may point to slight alignment errors in the setup. Ideally, the co-polarized THG is completely suppressed but shows a strong contribution at 3.8° in the cross-polarized channel. The minute contribution of co-polarization diffraction orders might be an effect of non-symmetric coupling between neighboring unit cells caused by the fabricated phase gradient, decoupling the structural symmetry from the lattice symmetry. Since the -3.6° -diffraction order is stronger this might be an indication, that a $\Phi_{\text{nonlin}} = -4\theta$ -contribution, similar to the C4 structure in the hexagonal lattice (see figure 4.4) is present.

To summarize, the systematic error of around 4% is found in all measurements, however, after correction the measured angles are close to the anticipated values, validating the simulated phase responses shown in figure 4.4. Furthermore, the C3 structures show an unexpected diffraction order at twice the co-polarization diffraction angle, which - in the simulations - was predicted for different structural parameters.

After learning of the SHG contributions, future measurements include dedicated THG and SHG measurements with proper color filters. For THG measurements, both an FESH0650 and FESH0500 short-pass filter are used. While the FESH0500 alone already has excellent suppression of the SHG wavelength, blocking of the fundamental wavelength is insufficient which is why the FESH0650 is also used. To measure the SHG, an FELH0550 long-pass filter in combination with the FESH0650 short-pass filter is placed in front of the camera. Figure 4.11 shows measurements for the C3 structures (THG and SHG) and for the C4 structures (hex. lattice, THG) while varying the fundamental wavelength from 1180 nm to 1300 nm in 10 nm steps. In these measurements, out of the fabricated C3 structures only $P_{SL} = 9.36 \mu\text{m}$ is analyzed and the adjusted pitch from the sample-holder is reset, in order to show both halves of the k-space. Additionally, the strong contribution from the undiffracted beam is blocked. Note that THG wavelengths below 393 nm (equating to 1180 nm fundamental wavelength) are heavily suppressed because of the optical properties of the FESH0650 short-pass filter. Additionally, transmission of the fundamental wavelength increases past 1300 nm for both color filters, influencing the measurements. Similarly, the transmission of SHG is heavily suppressed for wavelengths larger than 650 nm (1300 nm fundamental wavelength).

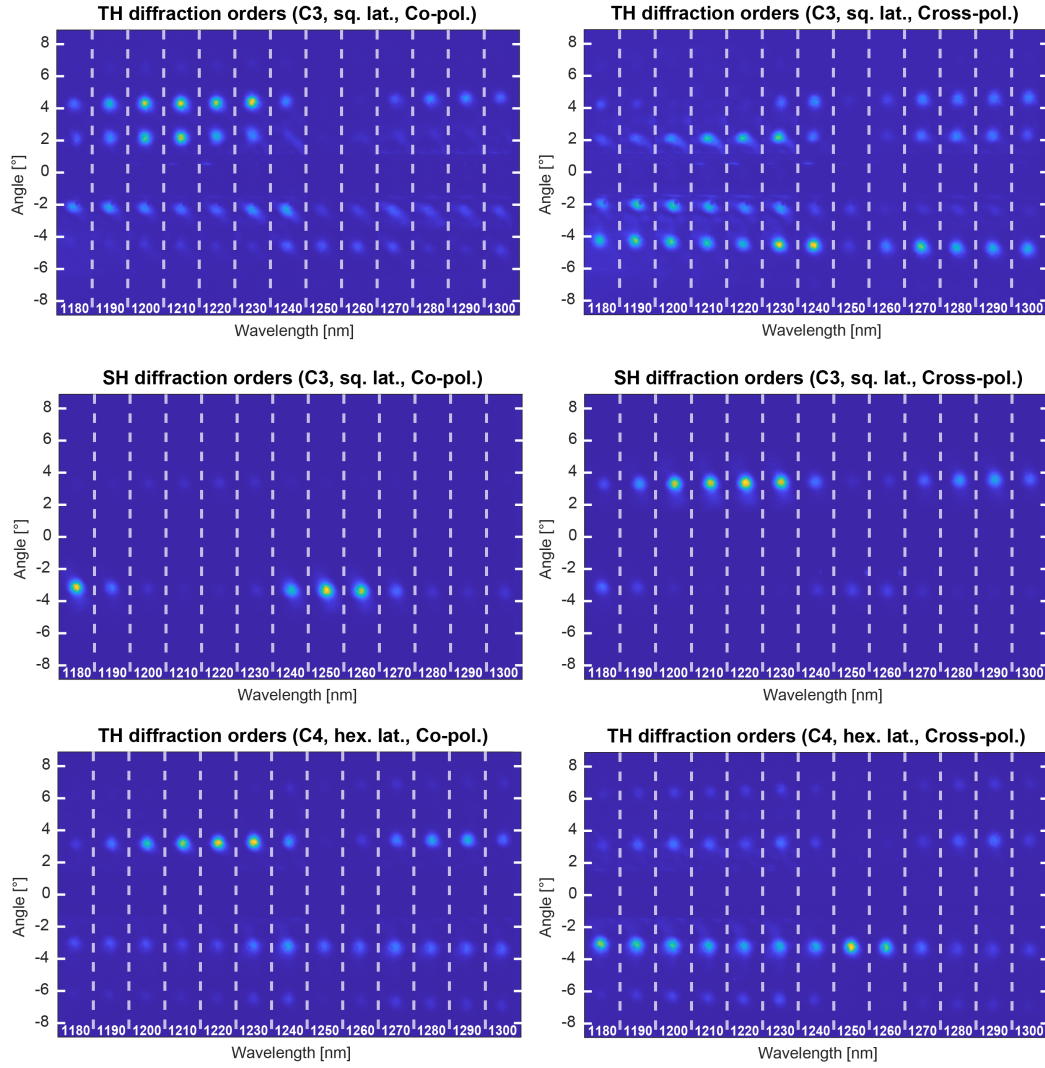


Figure 4.11: Diffraction orders of C3 structures (SHG and THG) and C4 structures (THG) in co-polarization (left) and cross-polarization (right) for fundamental wavelengths varying from 1180 nm to 1300 nm. The strong undiffracted beam is blocked to increase visibility of the diffraction orders. [68]

Now with the symmetric k-space visible, we can see that along the fundamental wavelength sweep, strong diffraction order contributions can be found in both halves of the k-space, whereas for a purely circular polarization incident on the samples THG diffraction orders in only one half are expected. Additionally, the measurements exhibit something akin to a beating effect for the individual diffraction orders, showing vanishing diffraction orders/reappearance at angles with opposite signs for some wavelengths - especially visible for the SHG measurements. After testing the polarization of the fundamental wave after reflecting off of the dichroic mirror (DM), a wavelength depended distortion of the

polarization state leading to strong ellipticity/linear polarization for some wavelengths could be observed. The polarization state of the fundamental beam was measured by placing a LP and a power meter behind the DM. The LP was rotated from 0° to 360° in steps of 5° . Figure 4.12 shows such a measurement for a wavelength range from 1200 - 1300 nm in steps of 20 nm. From the measurement, it can be seen, that most wavelengths end up nearly linearly polarized with the exception of 1260 nm showing elliptical polarization. This behavior explains the beating effect and the switching of the diffraction orders from one half of the k-space to the other. At around 1250 - 1260 nm, where the polarization state seems to be closed to circular polarization, the diffraction orders mostly vanish for positive diffraction angles.

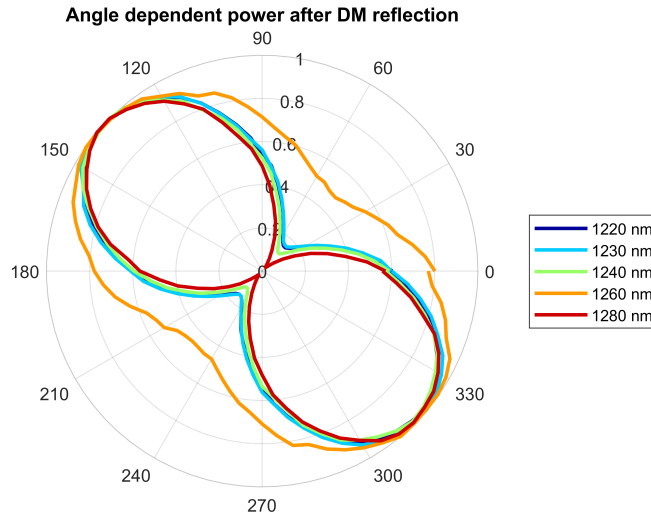


Figure 4.12: Polarization characterization of the fundamental beam from 1200 nm - 1300 nm in 20 nm steps. The polarization is tested after the beam passes the LP, the QWP and the DM. By introducing another LP and changing its orientation angle (azimuthal angle given in degree), we expect a constant power (here normalized to 1) as a function over rotation angle for circular polarized light. However, the results show a nearly linear polarized state for most wavelengths implying a strong wavelength dependent reflection phase with an elliptical polarization at 1260 nm.

Since this measurement does not provide full information on the wavelength dependent polarization state, the Stokes parameters were also determined. To do this, the beam first passes a QWP to generate circular polarization, reflects off the dichroic mirror and passes a second QWP as well as a linear polarizer. The orientation angle of the fast axis from the second QWP is varied in 22.5° -steps - relative to the LP axis - covering a total of 16 settings from $0^\circ - 337.5^\circ$. The transmitted power is measured with a spectrometer to increase the wavelength resolution since the center wavelength from the previous measurement is an average of a larger bandwidth. The measured powers from this rotating QWP measurement can be used to determine the Stokes parameters [72]. Additionally, a reference measurement

is done by replacing the DM with a normal mirror. The measurement is repeated for a laser-wavelength range from 1200 nm - 1290 nm in steps of 10 nm. From the spectrometer, this results in a Stokes measurement from 1200 nm - 1300 nm in steps of 1.6 nm. The results of both setups are plotted in figure 4.13. The Stokes parameters S_1 , S_2 and S_3 were normalized by S_0 , whereas the degree of polarization was between 0.9 - 0.95 for all measurements.

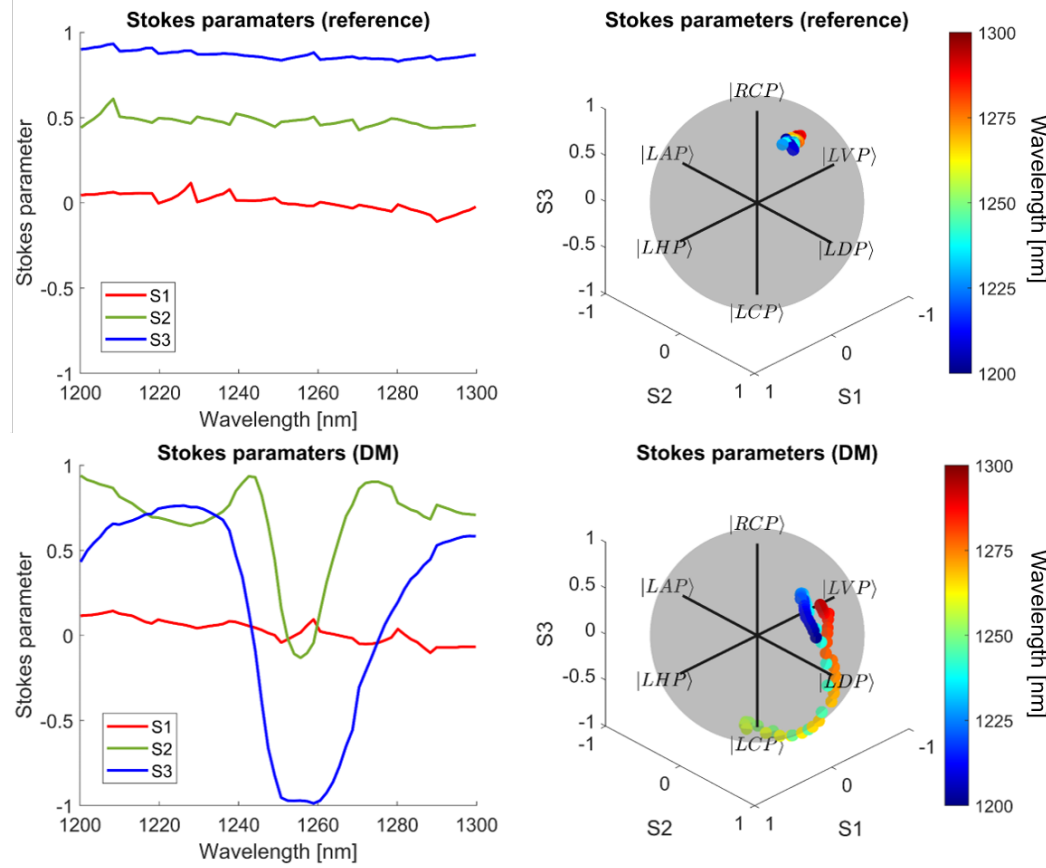


Figure 4.13: Stokes parameters over wavelength (left) and plotted on a Poincaré sphere (right) from 1200 nm - 1300 nm. (top) reference measurement of the initially set right circular polarization state after reflection off of a silver mirror. (bottom) influence of the dichroic mirror on the polarization state. The polarization seems to shift in between the initial RCP (+ offset) and its diametrically opposite point. Instead of continuing over the LAP states after reaching this point, the polarization state seems to reverse, passing a pure linear polarized state on the equator twice.

The Stokes parameters from the reference measurement seem to be mostly constant over wavelength leading to a polarization state close to the north pole on the Poincaré sphere. The offset from a pure RCP state can be attributed to the silver mirror reflection under an angle of 45° (compare figure 3.12 from the pulse shaping chapter). Interestingly, the influence from the DM seems to lead to a shifting of the polarization state from the almost

circular polarization to its diametrically opposite point. The change in polarization has similarities with the influence of birefringent materials in that a continuous change in the polarization state over a narrow bandwidth can be achieved.

After replacing the dichroic mirror, I noticed that other beamsplitters also introduce a similar effect. This polarization distortion explains the beating effect visible in the previous measurements, leading to the shifting of the diffraction orders from one half of the k -space to the other. This distortion is probably introduced by different reflection phases for s- and p- polarized light. Normally, the relative phase between s- and p- polarized light after a reflection changes by nearly 180° which is why a mirror illuminated parallel to the surface normal changes an RCP state to LCP; the spatial orientation of the electric field vectors stays the same but the propagation direction is changed from $\mathbf{k}$ to $-\mathbf{k}$. However, any difference in the relative phase after an interface reflection past the typical 180° gradually changes the polarization state. Due to symmetry reasons - there is no real distinction between s- and p- polarized light when illuminating a mirror normal to the surface - this effect tends to be minimized for small AOI and is of no concern, if the incident polarization for any AOI is only s- or p- polarized to begin with. Hence, to ensure clean circular polarization states, placing the QWP as close to the sample as possible (meaning as few optical components as possible in between the QWP and the sample) is desirable. In contrast to the experimental setup from section 3, placing the QWP directly in front of the sample doesn't work, as the wavelength range of the component does not encompass the THG.

In order to avoid using a beamsplitter altogether, I eventually use a silver-mirror at a small AOI of 8° (to minimize the polarization distortion) close to its edge. The lens used to focus on the metasurfaces is used slightly off-axis to generate a beam-offset between the incident fundamental wave and the collimated third harmonic, allowing to bypass said mirror and guide the generated nonlinear signal to the camera. Because of this off-axis use, the sample is illuminated under an angle of roughly 2° . Figure 4.14 illustrates the new optical setup.

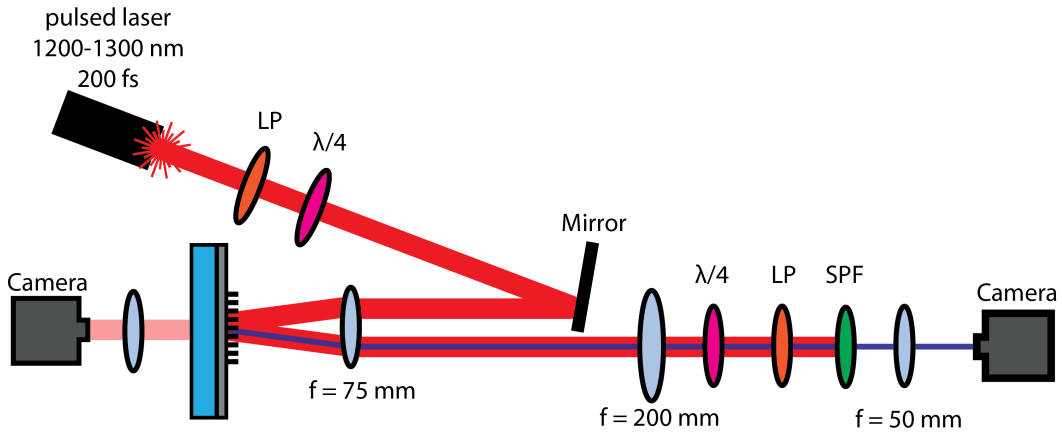


Figure 4.14: Top view of the new optical setup. The small AOI of 8° on the mirror helps to minimize distortion of the circular polarization and the off-axis use of the focusing lens allows the third harmonic to bypass the mirror [68]

Similar to figure 4.12, the polarization state of the fundamental wavelength is measured after interaction with the added silver-mirror for various linear polarizer settings (see figure 4.15). One reference measurement at 1240 nm is taken directly before the fundamental beam reflected off of the silver-mirror, showing that the polarization is unaffected by the new experimental setup. Additionally, one of the generated TH diffraction orders is tested in a similar way by evaluating the camera images for various linear polarizer settings (the QWP intended for the THG wavelength is removed in this measurement).

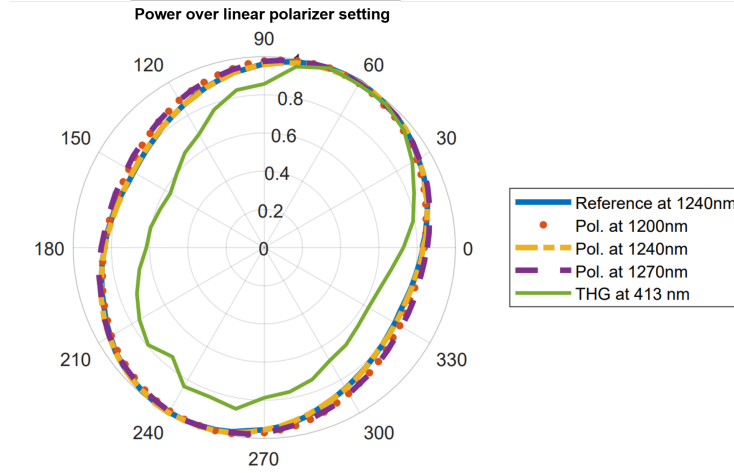


Figure 4.15: Polarization state of the fundamental and the third harmonic wavelength. The third harmonic is evaluated from one of the measured TH diffraction orders on the camera using only a linear polarizer instead of an analyzer. The fundamental wavelength is characterized after its reflection off the silver-mirror. The reference measurement is taken directly after the used QWP and before interacting with the mirror. [68]

The measurements do not indicate a wavelength dependent change of the polarization anymore, however, similar to the reference measurement from figure 4.13, we can see that the fundamental beams polarization state is not perfectly circular. The ratio between the long and short semi-axis of the FW's elliptical polarization state is 1.26 but nearly constant over wavelength. The TH shows a similar behavior albeit with a larger dependence on the set linear polarization. I used the new setup to repeat the measurements shown in figures 4.10 and 4.11 (see figures 4.16 and 4.17). Since the collimating lens of the generated nonlinear signals is now 1 inch in diameter (instead of 2) with a focal length of 75 mm (instead of 100 mm), the numerical aperture of the system is reduced to $NA \approx 0.16$. For the new measurements, only the structures with the larger super lattice periodicity - resulting in smaller diffraction angles - are measured, capturing diffraction angles from -7° to 7° .

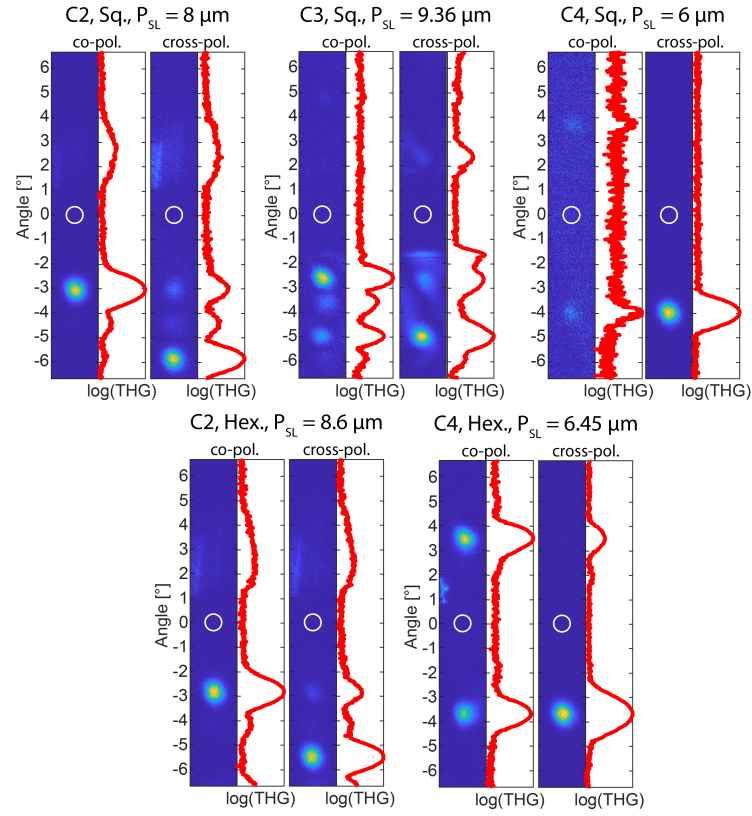


Figure 4.16: Measured diffraction orders from the structures referenced in table 4.6. The logarithmic plot improves the visibility of weaker diffraction orders. These measurements were conducted with a fundamental wavelength of 1200 nm and with blocked undiffracted beams.[adapted from [68]]

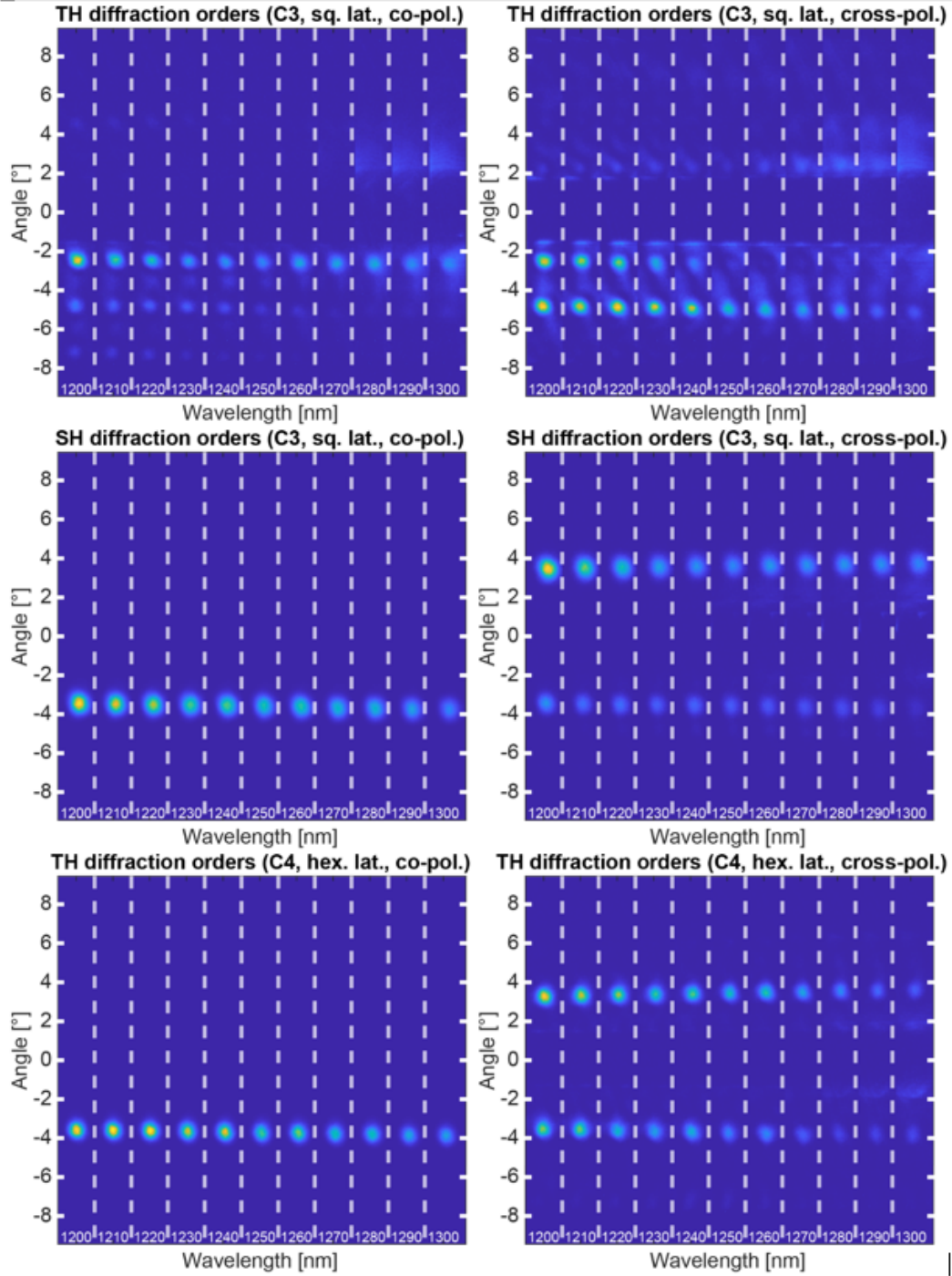


Figure 4.17: Diffraction orders of C3 structures (SHG and THG) and C4 structures (THG) in co-polarization (left) and cross-polarization (right) for fundamental wavelengths varying from 1180 nm to 1300 nm. The strong undiffracted beam is blocked to increase visibility of the diffraction orders.[adapted from [68]]

From the measurements in figure 4.16 we can see, that the co- and cross-polarization diffraction orders show better resemblance to the expected values. Both C2 structures show a good distinction between their polarization channels with only a minor contribution at $\approx 3^\circ$ in the cross-polarization channel where a peak in the co-polarization is expected. The C4 structure in the square lattice shows nice suppression of the co-polarization channel and is much more pronounced, as soon as the structure-lattice symmetry mismatch is introduced via antenna-antenna coupling (C4 in hex. lattice, co-polarization). Note that the expected diffraction order in co-polarization is opposite to the pronounced cross-polarization diffraction order and not supposed to be in both halves of the k-space. In comparison to the square lattice configuration, the additional coupling introduced in the hexagonal lattice increases the co-polarized signal strength by 2 orders of magnitude. Lastly, the C3 structures show the expected co-polarized diffraction order with a smaller contribution at twice that angle. Additionally, as mentioned before, a presumable 12θ -contribution (attributed to C3 structures with totally different structural parameters) seems to be present for the cross-polarization.

From figure 4.17 we can also see, that the new setup fixed the previously present beating effect, now showing consistent behavior over different fundamental wavelengths.

Lastly, I want to compare the simulated THG conversion efficiency to the measurements. In order provide the conversion efficiency $\eta_{\text{THG}} = \frac{P_{\text{THG}}}{P_{\text{FW}}}$, closer examination of the laser system and the utilized optical components is necessary. Here, I will focus on the respective co- and cross- THG conversion efficiency of the C4 structures in the hexagonal lattice and additionally on the co- and cross- SHG conversion efficiency of the C3 structures from figure 4.17.

The pulsed laser system (Coherent Chameleon Ultra II) has a repetition rate of 80 MHz and a temporal width of around 200 fs in this wavelength range (8 nm bandwidth). The mean power was set to 100 mW/50 mW for each wavelength during the THG/SHG measurements (measured after the silver-mirror interaction). This means, that the only optical component introducing losses to the fundamental wavelength is the focusing lens (AC254-C-075-ML). On the other hand, the THG passes three lenses (AC254-C-075-ML, AC508-A-200-ML and AC254-A-050-ML from Thorlabs), two short-pass filters (FESH0500, FESH0650 from Thorlabs), a quarter-wave plate (AQWP05M-580 from Thorlabs) and a linear polarizer (colorpol VIS 500 BC3 CW01 from Codixx) until finally being detected by the camera (Zyla5.5 from Andor). For the SHG measurements, an FELH0550 instead of the FESH0500 is considered. Since no information on the linear polarizer and the first lens (C-coating, specified use from 1050 nm - 1700 nm) for the wavelength range around 400 nm or 600 nm exists, their transmission/losses are measured for center wavelengths ranging from 405 nm to 440 nm as well as 600 nm to 650 nm in steps of 5 nm with a bandwidth of 10 nm using a Fianium WhiteLase SC-400 in combination with a monochromator. The combined transmission for the THG/SHG signal from all these components over wavelength can be seen in figure 4.18 (top-left).

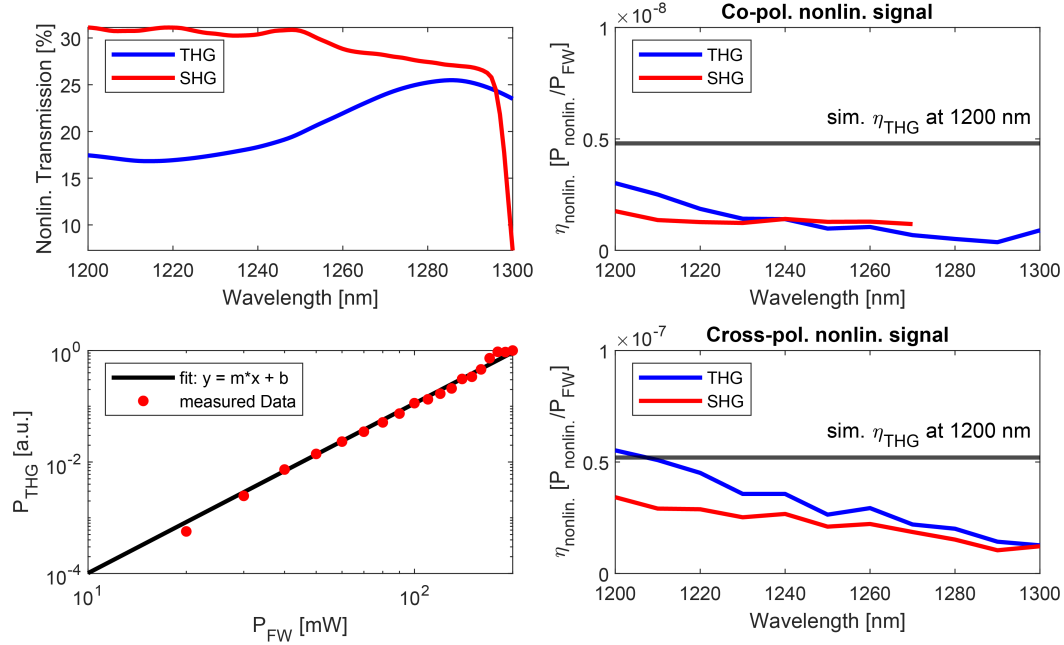


Figure 4.18: (top-left) Total transmission of the generated nonlinear signal plotted over its corresponding fundamental wavelength. (bottom-left) double-logarithmic scale of incident fundamental wave power and normalized generated TH power. The linear fit has a slope of 3.044 (R-square value of 0.993) showing the expected cubic nature of the third harmonic. (top-right) co-polarized and (bottom-right) cross-polarized THG/SHG conversion efficiency signal from C4 structures in a hexagonal lattice/C3 structures in square lattice.

The background corrected camera images are integrated row-wise and a gaussian fit is applied to each diffraction order that is clearly visible in the images. Afterwards, each diffraction order from the row-wise integrated images is integrated within a $2\text{-}\sigma$ environment and the result is divided by the total transmission shown in figure 4.18 to determine the total counts C_{tot} . Scaling this down to one individual pulse by the the product of the laser's repetition rate R_{rate} and the integration time t_{int} (set to 10 s/3 s during THG/SHG measurements) as well as multiplying by the THG/SHG photon energy, we get the THG/SHG energy per pulse $E_{\text{nonlin., pulse}}$.

$$E_{\text{nonlin., pulse}} = \frac{C_{\text{tot}}}{t_{\text{int}} R_{\text{rate}}} \cdot \frac{h c}{\lambda_{\text{nonlin}}} \quad (4.34)$$

The nonlinear peak power $P_{\text{nonlin., peak}}$ is determined by dividing $E_{\text{nonlin., pulse}}$ by the temporal width of the pulse (here also assumed as $\tau = 200$ fs). Similarly, we can calculate the fundamental wave peak power by correcting the measured power P_{FW} by the optical losses $\text{loss}(\lambda)$ and dividing the resulting power by the product of R_{rate} and the temporal width

τ .

$$P_{\text{FW, peak}} = \frac{P_{\text{FW}}}{\text{loss}(\lambda)} \cdot \frac{1}{R_{\text{rate}} \tau} \quad (4.35)$$

Before evaluating the conversion efficiency, in order to assure comparability to the simulations, the power ratio needs to be scaled in accordance with the intensity set in the simulations ($I_{\text{FW}} = 1.33 \frac{\text{GW}}{\text{cm}^2}$). To evaluate the FW intensity in the experimental setup, the imaging setup behind the sample is used to determine the beam radius of the focused laser beam. With the magnification of the imaging system unknown, we can compare the image size on the camera chip of an object to its real-world object size. As a reference object, a metasurface with a side length of 100 μm is taken to determine the magnification. Lastly, by applying a gaussian fit to the imaged focus of the laser beam, the beam radius (taking the $1/e^2$ -convention) and thus the beam area is determined. After dividing $P_{\text{FW, peak}}$ by the beam area, the FW intensity in the experimental setup is determined as $I_{\text{FW, setup}} \approx 0.19/0.095 \frac{\text{GW}}{\text{cm}^2}$ during the THG/SHG measurements. Dividing the intensity from the simulations by the measured intensity and cubing/squaring the resulting ratio provides the scaling factor for the THG/SHG measurements. The resulting THG/SHG nonlinear conversion efficiencies for the co- and cross-polarization from the C4 structures in hexagonal lattice/C3 structures in square lattice are shown in figure 4.18 (right). Additionally, a power dependent measurement series from 20 mW to 200 mW in 10 mW steps is done for the cross-polarized diffraction order from the C4 structures in the hexagonal lattice (bottom-left).

After fitting a linear function onto the double-logarithmic representation of the power dependent measurement, the slope is found to be 3.044 (R-squared value of 0.993), illustrating the expected third-power dependence on the incident power as expected for THG. Furthermore, the calculated conversion efficiency in the co- and cross-polarization channel within the same order of magnitude compared to the simulated results that were listed in table 4.5. The co- and cross-polarization nonlinear conversion efficiency are measured to be $0.3 \cdot 10^{-8}$ (simulated as $\eta_{\text{THG, co}} = 0.48 \cdot 10^{-8}$) and $5.52 \cdot 10^{-8}$ (simulated as $\eta_{\text{THG, cross}} = 5.2 \cdot 10^{-8}$) at the design wavelength of 1200 nm. Note that the strong undiffracted THG signal is not included in these evaluations. Additionally, we find that the conversion efficiency of the SHG signal is similar to the THG conversion efficiency. Table 4.7 contains several other publications that used Si nano-structures to generate TH and evaluated the conversion efficiency. The structures used different mode excitation schemes, e.g. Mie-resonances and qBIC-resonances, resulting in a widely varying range for the conversion efficiency. As its last column, the table contains the upscaled TH conversion efficiency at $I_{\text{FW}} = 1.33 \frac{\text{GW}}{\text{cm}^2}$ to put the efficiency of the structures used in this work into perspective. Note that I only evaluated the diffraction orders - leaving out the strong undiffracted THG signal - and that some literature values are already working close to the efficiency-saturation region of their structures, making the upscaling nonsensical as the given values in the last column might not be reachable. All in all, despite not confining the fundamental wavelength within the nano-structure's small Si-volume, the metasurface shows a respectable nonlinear conversion efficiency.

Table 4.7: THG conversion efficiency comparison of Si nano-structures from various publications. The mode-excitation schemes used vary from sharp resonances like Fano- or qBIC-modes to broader Mie-type modes. The table shows the THG wavelength, the reported conversion efficiency at the given fundamental wave intensity as well as an upscaled efficiency to match the simulation intensity of the structures presented in this chapter. Note that the upscaling does not take into account at what point some structures go into the efficiency-saturation region.

source	λ_{THG} [nm]	$I_{\text{FW}} \left[\frac{\text{GW}}{\text{cm}^2} \right]$	η_{THG}	η_{THG} at $1.33 \frac{\text{GW}}{\text{cm}^2}$
[73]	528	0.11	$0.9 \cdot 10^{-6}$	$1.59 \cdot 10^{-3}$
[74]	343	0.7	$1.13 \cdot 10^{-5}$	$7.75 \cdot 10^{-5}$
[75]	417	0.42	$1.8 \cdot 10^{-6}$	$5.72 \cdot 10^{-5}$
[76]	517	3	$4 \cdot 10^{-5}$	$3.48 \cdot 10^{-6}$
[77]	427	0.52	$1.76 \cdot 10^{-7}$	$2.94 \cdot 10^{-6}$
[28]	520	1.2	$2.8 \cdot 10^{-7}$	$3.81 \cdot 10^{-7}$
[78]	477	2.59	$1 \cdot 10^{-6}$	$1.35 \cdot 10^{-7}$
[79]	413	0.06	$1 \cdot 10^{-11}$	$1.2 \cdot 10^{-7}$
[80]	449	3.2	$1.2 \cdot 10^{-6}$	$8.61 \cdot 10^{-8}$
This work [68]	413	0.19	$1.61 \cdot 10^{-10}$	$5.52 \cdot 10^{-8}$
[81]	420	5.5	$1 \cdot 10^{-7}$	$1.41 \cdot 10^{-9}$
[82]	427	23.3	$2.2 \cdot 10^{-6}$	$4.09 \cdot 10^{-10}$
[83]	467	33	$1.1 \cdot 10^{-6}$	$7.2 \cdot 10^{-11}$

4.6 Conclusion

In this chapter, I experimentally investigated the THG phase response of silicon meta-atoms with different rotational symmetries in square and hexagonal lattice configurations. These structures are fabricated on top of a reflective silver-layer that enhances THG by 3 orders of magnitude compared to similar structures placed on a transparent substrate [71]. This work covers C2-C4 rotationally symmetric structures in a square lattice as well as C2 and C4 structures in a hexagonal lattice. By carefully designing the meta-atom's structural parameters, certain nonlinear geometrical phase terms become accessible through antenna-antenna coupling in combination with a symmetry mismatch between the rotational symmetry and the lattice. These phase responses were validated by fabricating phase-gradient type metasurfaces that translate the respective nonlinear geometrical phase responses to a diffraction order with a specific k-vector.

While the selection rules that only describe individual structures already predict third order nonlinear geometrical phases for C2 structures, C4 structures are not expected to exhibit modulated co-polarized THG and for C3 structures both co- and cross-polarization are forbidden. By decoupling the latter two structure's rotational symmetry from the lattice symmetry, i.e. placing C3 structures in a square lattice and C4 structures in a hexagonal lattice, we see that these THG channels become available. Furthermore, the theoretical work derived by Bingyi Liu predicts multiple solutions for each structure that can be

accessed by properly changing their structural parameters.

While the simulations did not predict a diffraction order in the cross-polarized THG channel of the C3 structures for our particular set of parameters, we do find a strong THG diffraction order for this polarization. Additionally, the C4 structures in the hexagonal lattice unexpectedly show diffraction orders of similar strength in both halves of the k -space. This points towards unexpected wave-mixing contributions in these structures - maybe attributed to fabrication deviations - that are nevertheless covered by set of solutions derived from the formulated theory. Furthermore, the measured conversion efficiency η_{THG} for both co- and cross- polarized diffraction orders of the C4 structures closely match to the simulated efficiency.

The presented work and theory behind it extends upon a widely used tool to encode phases into metasurfaces and might enable activating certain phase responses by disturbing fabricated lattices, e.g. via strain that introduces the required symmetry mismatch between structure and lattice.

This work was published in “ACS Photonics” in 2023 [68].

5.1 Introduction

Metasurfaces introduce abrupt phase and amplitude changes to incident wavefronts, typically on sub-wavelength scales and their planar layout even circumvents the need for phase matching in nonlinear optical applications. In that regard, they can be considered as 2D-materials and because of this, even despite the presence of a substrate, they mostly exhibit out of plane symmetry [84]. As a consequence, their transmission properties are symmetric, meaning that they behave identical when input and output sides are interchanged - they are subject to optical reciprocity. This property is shared by most optical components used in labs: prisms, gratings, lenses, mirrors, anti-reflective coatings... and it is often beneficial in compact applications, reusing an optical component to undo a certain optical operation. Take the pulse shaper as an example; the input and output beam shapes are (ideally) identical which is realized by symmetric use of a couple of prisms or gratings. In contrast, non-reciprocal components do not behave the same way when the operation side is reversed. In electronics for example we have the diode which only allows a current to pass in one direction. In fluid-dynamics, there are passive devices like Tesla's macrofluidic valve. By repeatedly adding hook-shaped pockets/bypasses to a mostly straight main channel, a geometrical asymmetry along the flow-direction is introduced. While the fluid flow is mostly undisturbed in one direction, turbulence (and thereby resistance to fluid flow) is introduced when operated in reverse [85]. An example of an active non-reciprocal device is the aortic valve; when the heart contracts, the valve opens and allows for blood to flow along the arteries into the capillaries. On relaxation, this valve closes to prevent backflow, forcing rerouting of the oxygen deprived blood via the veins.

A well-known optical equivalent to these active devices is the Faraday rotator. By applying an external, static magnetic field to a ferromagnetic crystal, light that propagates along the magnetic field lines experiences circular birefringence, induced via the magneto-optical effect. As a consequence, LCP and RCP travel with different speeds, leading to a change in polarization over propagation distance. Interestingly, this effect is not reversed when the propagation direction changes. Instead, the polarization rotation is additive. By designing a device that rotates the polarization by 45° on a single pass, a back-reflection passing this crystal in opposite direction is now rotated by a total of 90° . By combining a Faraday rotator with a polarizing beamsplitter (= optical isolator) one can prevent back-reflections from entering a laser cavity, which can otherwise cause destabilization of the laser system [86]. Similarly, back-reflections at optical fibers facets can increase the bit error rate by creating ghosting signals [87], especially at high data transfer rates and could be prevented by such optical isolators.

While the Faraday Rotator is an active component, requiring an external magnetic field as well as a certain propagation length to fully take effect - making the component bulky - passive non-reciprocal metasurfaces have also been explored. Yang et al. used magnetic nano-antennas in the low GHz regime, consisting of cuboid shaped lanthanum-doped barium hexaferrite in order to leverage said magneto-optical effect without the need for external magnetic fields in addition to subwavelength thickness [88]. In forward and backwards propagation, the structures show difference resonance frequencies, thereby introducing different transmission and phase that were used for directionally depended holography.

Aside from magnetism used to introduce non-reciprocity, Tripathi et al. use VO_2 -coated substrates in combination with Si nano-posts that show directionally dependent transmission [89]. The structures can be designed in such a way, that the direction of incidence shows different near-field distributions in the nano-pillars. If for one direction a strong field enhancement near the VO_2 surface is achieved, the corresponding absorption increases. As a consequence, the VO_2 layer heats up more and induces a phase transition that was exploited to reduce the metasurface transmission. The non-reciprocity of the unit cell is therefore a consequence of asymmetric near field excitations. In a similar manner, Mathew et al. used Si nano-posts on ITO-coated substrates to leverage the nonlinear optical refractive index in the epsilon near zero (ENZ) regime and also making use of asymmetric field enhancement depending on the direction of incidence [90]. For a low intensity regime, the device behaves reciprocal, whereas higher power and therefore larger refractive index changes result in a strong transmission contrast between forwards and backwards operation. The quick response time of the ITO makes this design suitable for ultra-fast optical switching.

In 2019, Frese et al. realized a two-layer plasmonic Janus metasurface, acting as an optical one-way transmission channel which is designed to generate a single hologram for a given input polarization [91]. In reverse direction, the same incident linear polarization does not retrieve the hologram, potentially making it an anti-counterfeiting feature. This design works by introducing a layer acting as a linear polarizer; depending on if this layer is addressed first or last, the allowed polarization conversion in the device is altered.

While this is not an exhaustive list, we can see that there are several ways of introducing non-reciprocity, mostly relying on specially designed light-matter interactions. The metasurface presented in this chapter also makes use of intermediate polarization filtering, analogous to the work from Frese et al. We extend this design by an additional layer, thereby realizing a three layer plasmonic Janus metasurface. The increased design complexity goes hand in hand with an increase in the information capacity. By encoding certain phase information into the first and third layer we make use of three distinct polarization channels and also introduce wavelength multiplexing at 900 nm and 1200 nm. These additions extend the number of encoded holograms from one to six. While this design does not allow for ultra-fast optical switching because of the absence of any nonlinear effects, it allows functionality at low peak power continuous wave operation and can function as a miniaturized optical isolator.

5.2 Theory

Within the confines of this work, we assume that the Janus metasurface consists of three layers whereas each individual layer is subject to reciprocity. Furthermore, the first and last layer are able to generate co- and cross-polarization by use of L-shaped nano-antennas, given by the off-diagonal transmission matrix elements. More precisely, input x-polarized light is converted to both x-polarization (co-polarization) and y-polarization (cross-polarization). Similarly, incident y-polarization can partially be converted to x-polarization. The transmission matrix of these L-shaped optical elements is generally described by:

$$t_L^F = \begin{pmatrix} t_{L,xx}^F & t_{L,xy}^F \\ t_{L,yx}^F & t_{L,yy}^F \end{pmatrix}. \quad (5.1)$$

The superscript F refers to forward propagation and can be contrasted by B if backwards propagation through the system were to be considered. The subscript L refers to the L-shaped structures and the additional subscript of the matrix elements refers to the linear output- and input-polarization state, respectively. Furthermore, reciprocity of an optical element gives the following relation [91]:

$$\begin{pmatrix} t_{xx}^F & t_{xy}^F \\ t_{yx}^F & t_{yy}^F \end{pmatrix} = \begin{pmatrix} t_{xx}^B & -t_{yx}^B \\ -t_{xy}^B & t_{yy}^B \end{pmatrix} \quad (5.2)$$

When changing between forward and backward propagation, we can see that the diagonal matrix elements remain identical, meaning that no difference in co-polarization transmission exists between forward and backward propagation. The off-diagonal elements on the other hand swap positions and invert their sign. Note that the sign-change is a consequence of the change in coordinate-system definition once the propagation direction is inverted. Additionally, in the case of optical elements exhibiting out-of-plane symmetry (as is typically the case for single layer planar structures, despite their air-substrate), the off-diagonal elements are identical $t_{yx} = t_{xy}$, meaning that they do not impart independent phases. This constraint also applies to the individual layers presented here.

The middle layer is supposed to act as a linear polarizer, only allowing transmission for the t_{xx} matrix element:

$$t_D^F = \begin{pmatrix} 1 & 0 \\ 0 & 0 \end{pmatrix}. \quad (5.3)$$

The polarization filtering is done using dimer-structures, hence the subscript D . From now on, we differentiate between layer L_a (above) and layer L_b (below) as the upper and lower layer containing L-shaped nano-antennas in this three-layer stack.

Calculating the total forward propagation transmission T^F through the entire layer system

for any pair of input- and output-polarizations, we get:

$$\begin{aligned}
 T^F &= t_{Lb}^F \cdot t_D^F \cdot t_{La}^F \\
 \langle X | T^F | X \rangle &= T_{XX}^F = t_{La,xx}^F t_{Lb,xx}^F \\
 \langle Y | T^F | X \rangle &= T_{YX}^F = t_{La,xx}^F t_{Lb,yx}^F \\
 \langle X | T^F | Y \rangle &= T_{XY}^F = t_{La,xy}^F t_{Lb,xx}^F \\
 \langle Y | T^F | Y \rangle &= T_{YY}^F = t_{La,xy}^F t_{Lb,yx}^F
 \end{aligned}$$

Combining these contributions, we get the forward propagation transmission matrix of the Janus metasurface T_{JM}^F as:

$$T_{JM}^F = \begin{pmatrix} T_{XX}^F & T_{XY}^F \\ T_{YX}^F & T_{YY}^F \end{pmatrix} = \begin{pmatrix} t_{La,xx}^F & t_{Lb,xx}^F & t_{La,xx}^F & t_{Lb,xy}^F \\ t_{La,yx}^F & t_{Lb,xx}^F & t_{La,yx}^F & t_{Lb,xy}^F \end{pmatrix} \quad (5.4)$$

From the matrix T_{JM}^F we can see that a transmission from x- to y- polarization does not carry the same information as a transmission from y- to x- polarization $T_{YX}^F \neq T_{XY}^F$, thus breaking the reciprocity from the individual layers. Note that the presence of the dimer-layer is what enables this non-reciprocal behavior.

Similar to the forward propagation, the transmission matrix for backward propagation is:

$$T_{JM}^B = \begin{pmatrix} T_{XX}^B & T_{XY}^B \\ T_{YX}^B & T_{YY}^B \end{pmatrix} = \begin{pmatrix} t_{Lb,xx}^B & t_{La,xx}^B & t_{Lb,xx}^B & t_{La,xy}^B \\ t_{Lb,yx}^B & t_{La,xx}^B & t_{Lb,yx}^B & t_{La,xy}^B \end{pmatrix} \quad (5.5)$$

Since reciprocity still holds for an individual layer, if we apply relation 5.2 to every layer, we can rewrite the backwards propagation as

$$T_{JM}^B = \begin{pmatrix} t_{Lb,xx}^F & t_{La,xx}^F & -t_{Lb,xx}^F & t_{La,yx}^F \\ -t_{Lb,xy}^F & t_{La,xx}^F & t_{Lb,xy}^F & t_{La,yx}^F \end{pmatrix}. \quad (5.6)$$

We can see that compared to the transmission in forward direction, the off-diagonal elements swapped positions and inverted their sign. This means, when the propagation direction changes, the encoded holograms will appear mirrored and the requirement for retrieving these holograms changes, e.g. from incident x-polarization and detected y-polarization to incident y-polarization and detected x-polarization. Since the off-diagonal elements carry different phase information, they too become accessible as independent channels for holography, allowing for - in principle - four different holography channels. In the next subsection I will explain, why we only get access to three of them with the presented layout.

5.3 Design

One shortcoming of plasmonic nano-particles is that their underlying resonance does not facilitate a full 2π phase coverage by itself. The previous chapters already showed, that rectangular structures in combination with circular polarization can achieve this by making use of the geometrical phase. But here we are interested in linear polarization states, since the dimer-layer responsible for transmitting only x-polarization is what introduces the non-reciprocity of the three layer design.

Another way of achieving a 2π phase coverage is by using V-shaped nanostructures, defined by an arm length h and an angle Δ between those arms [63, 91–93]. Figure 5.1 illustrates that these structures mainly support two different modes indicated by two distinct excited surface currents; a symmetric and an antisymmetric mode. A larger current density corresponds to a lighter color. We can see that the symmetric mode generates two in-phase dipoles along each arm of the structure, resonant to the arm length h , indicated by the current density which falls off at the joint and end of an individual arm. In contrast, the antisymmetric mode shows the largest surface current in the joint region of the V-shape and the excited current follows the entire length of the structure, giving a resonance wavelength proportional to $2h$.

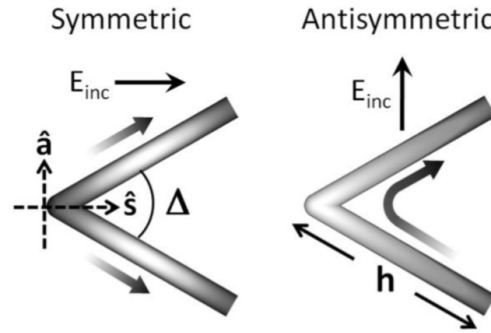


Figure 5.1: Illustration of symmetric and antisymmetric mode excitation in plasmonic V-shaped nano-antennas with arm-length h and angle Δ . The gray-scaled color in the antenna illustrates the local current density with white referring to large and black corresponding to small currents. The symmetric mode is excited by an electric field polarized parallel to the $\hat{s}$ -axis, generating two dipoles along each arm of length h . The asymmetric mode results from the excitation of the entire structure length $2h$. [93] (© 2013 IEEE. Reprinted with permission.)

As a consequence, the resonance wavelength for these two modes differs by a factor of ≈ 2 resulting in different phase and amplitude response [92–94]. In this work, we considered the special case of L-shaped structures ($\Delta = 90^\circ$, with one arm facing the $+x$ -direction and the other the $+y$ -direction). When exciting a plasmonic L-shaped structure with a polarization parallel to one of its arms, the induced current partially continues along the shape of the structure and allows coupling to the respective cross-polarization. On its own, this mode only covers a $0 - \pi$ phase range. But when rotating an L-shaped structure by

90° (one arm faces the +x-direction, another the -y-direction), one still leverages/excites the e.g. x-oriented arm, but the induced current flows in negative y-direction. Since we still couple to the same mode but reroute the current, the radiated electromagnetic wave is 180° phase shifted with respect to the unrotated L-shaped structure. By designing a set of parameters that sufficiently cover a $0 - \pi$ phase range and rotating these structures by 90°, we extend the viable phase range by π and get access to the full 2π -range. Note that this “geometrical trick” works by indirectly coupling to the second arm of the structure. For this reason, the co-polarization channel does not benefit from this effect, as it necessitates a dipole parallel to the incident beam. Consequently, the t_{xx} -channels derived in the transmission matrix 5.6 do not contribute in a meaningful way to the phase range required to encode holograms. Additionally, because of the dimer layer, the t_{yy} -channels are also irrelevant for the suggested structures. Leaving out the co-polarization conversion from each layer, thereby fully neglecting the T_{XX} -channel, the relevant phase contributions for the holograms in forward direction are $t_{Lb,xy}^F$ as well as $t_{La,yx}^F$ and the combined $t_{La,yx}^F t_{Lb,xy}^F$ transmission coefficients. In backward direction, these are $t_{La,xy}^B$ as well as $t_{Lb,yx}^B$ and $t_{Lb,yx}^B t_{La,xy}^B$.

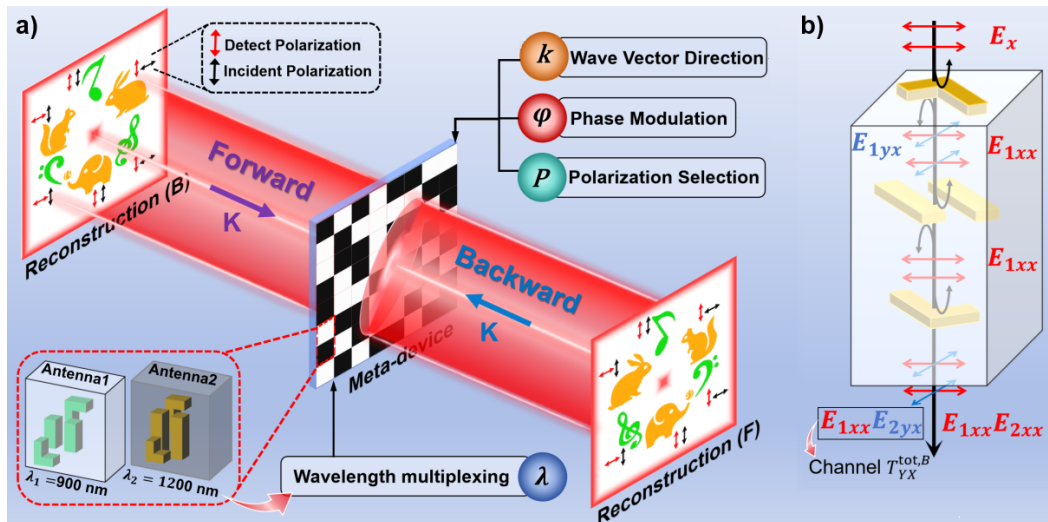


Figure 5.2: a) Schematic illustration of the cascaded plasmonic Janus metasurface (CPJM), encompassing wavelength, polarization and k-vector direction as multiplexing parameters. A total of six holographic images are accessible via different wavelength- and polarization-settings. By switching between forward and backward propagation, the holograms are mirrored and generation of a particular hologram is done by interchanging input-output polarization. The wavelength multiplexing is realized by staggering different sized antenna-layers into the 2D metasurface, exhibiting resonance features near 900 nm or 1200 nm, respectively. b) Visualization of the three layer stack performing polarization conversions and filtering to generate the transmission channel T_{YX}^B .

Figure 5.2 a) captures the working principle of the proposed cascaded plasmonic Janus metasurface (CPJM). The multiplexing encompasses the k -vector direction, co- and cross-

polarized transmission channels as well as wavelength multiplexing, aiming for operation at 900 nm and 1200 nm. Backward incidence is defined as light incident from the air side, and forward incidence as light incident from the substrate side. A total of six holograms can be generated with different wavelength and input/output polarizations. Upon changing the propagation direction, the holograms are mirrored and the required incident and detecting polarization interchange. Panel b) illustrates the polarization conversion for the T_{YX}^B transmission channel. Incident x-polarized light passes the layer L_a and is partially converted to y-polarization. Here, the generated y-polarization is filtered due to the dimer structure. The transmitted x-polarization then interacts with the last layer, again causing a partial conversion to y-polarization. In this particular channel, the cross-polarization conversion from Layer L_b can be used to encode a hologram. Similarly, layer L_a can be used to generate a hologram as well as the combined cross-polarization phases from layer L_a and L_b .

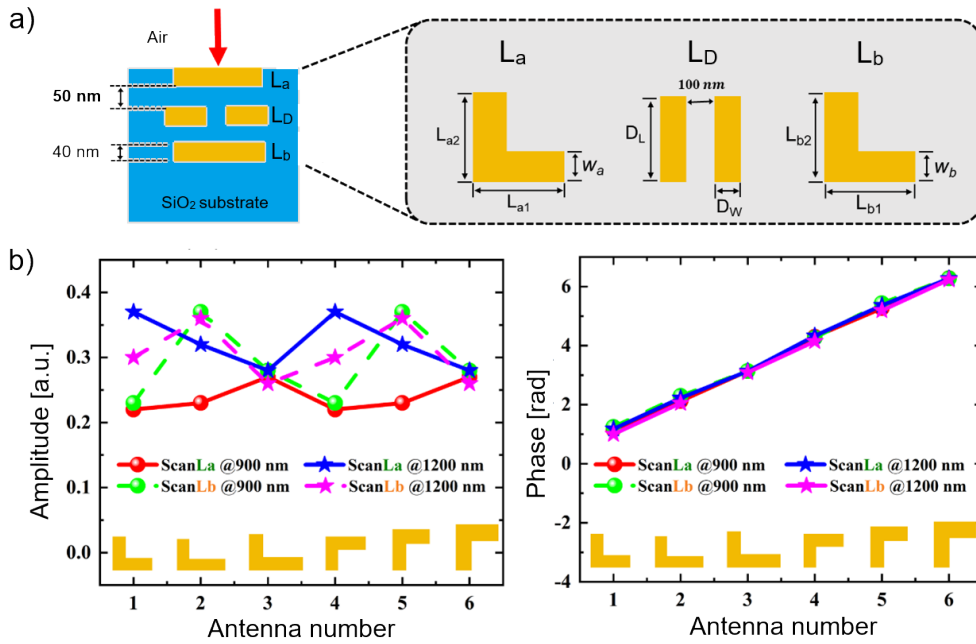


Figure 5.3: a) Schematic illustration of the three-layer design in the SiO₂-substrate. The layers consist of 40 nm thick gold-structures, each spaced by 50 nm. b) Transmitted electric field amplitude and phase of the optimized L-shaped nano-antennas. Including the 90° rotation of the L-shaped antennas (antennas 4-6), a good 2π -phase coverage is achieved.

Figure 5.3 a) shows the parametrization of the individual layers. The unit cell's periodicity is 550 nm, the antenna height is 40 nm and the layers are embedded in SiO₂ spaced by 50 nm, each. In order to optimize the structural parameters of each layer, the entire three layer stack is simulated in RCWA simulations to account for inter layer coupling (simulations performed by Guocui Wang). To speed up this optimization, starting parameters are

taken from single layer transmission evaluations. After that, only the layers L_a and L_b are sequentially optimized. When sweeping the geometrical parameters of e.g. layer L_a (those only including the two arm lengths), the parameters of layer L_b were kept fixed at the parameters retrieved from the single layer optimization. As an example, optimization of the layer L_a at 900 nm wavelength entailed fixed arm lengths of layer L_b (both being 160 nm) and sweeping the arm lengths of layer L_a from 60 nm - 450 nm, respectively. Conversely, when optimizing layer L_b , the structural parameters of layer L_a are fixed. From the 2D parameter optimization, three structures are selected to cover a π -phase range while aiming to keep a roughly constant transmission amplitude. By rotating the selected structures, the phase range is extended by π . Panel b) shows both the transmission coefficients' amplitude as well as the phase of channel T_{XY} and T_{YX} for the optimized antenna sets, including their 90° rotated variants (antennas 4 - 6). Additionally, table 5.1 shows the lateral geometrical parameters of the optimized antennas, excluding the 90° rotated variants.

Table 5.1: Lateral geometrical properties of the incorporated nano-antennas for each layer. Only 3 distinct L-shaped antennas are used per wavelength per layer. Introduction of 90° rotation of these L-shaped structures extends the phase range to 2π .

Antenna		$\lambda = 900 \text{ nm}$				$\lambda = 1200 \text{ nm}$			
	L_a	L_{a1} [nm]	135	170	195	L_{a1} [nm]	240	250	385
		L_{a2} [nm]	135	125	155	L_{a2} [nm]	195	295	230
		W_a [nm]	80	80	80	W_a [nm]	120	120	120
		L_{D1} [nm]	200	200	200	L_{D1} [nm]	380	380	380
	L_D	W_D [nm]	160	160	160	W_D [nm]	180	180	180
	L_b	L_{b1} [nm]	130	130	190	L_{b1} [nm]	290	235	210
		L_{b2} [nm]	115	140	135	L_{b2} [nm]	275	275	210
		W_b [nm]	80	80	80	W_b [nm]	120	120	120

With the building blocks of the CPJM introduced, the next challenge is the encoding of multiple holograms. The phase retrieval algorithm is implemented by Guocui Wang and is shown in figure 5.4. The algorithm randomly initialized two phase profiles (ϕ_a and ϕ_b) corresponding to the argument of the cross-polarization transmission terms of layer L_a and layer L_b . The third hologram for a particular wavelength is generated by the sum of these two phase masks $\phi_c = \phi_a + \phi_b$. Given an electric field amplitude, that is reapplied on every iteration-cycle, the Fast Fourier Transform of the three input light fields is taken, yielding the currently achieved holograms. A mean-squared error function (E_k with $k = a, b, c$) determines the difference of the holograms to the target images. Here, a gradient descent optimization is applied to the total error function E_t - the sum of the three individual error functions E_k - taking the derivative based on the phase mask value $\Phi = \phi_a + \phi_b$. If the resulting error is deemed small enough, the algorithm terminates, otherwise the current phase profiles (altered by the gradient descent optimization) are reused in the next

iteration cycle. The optimization at 900 nm and 1200 nm is done independently and the optimized phase profiles for these two wavelength are randomly staggered in the 2D plane to construct the final metasurface design.

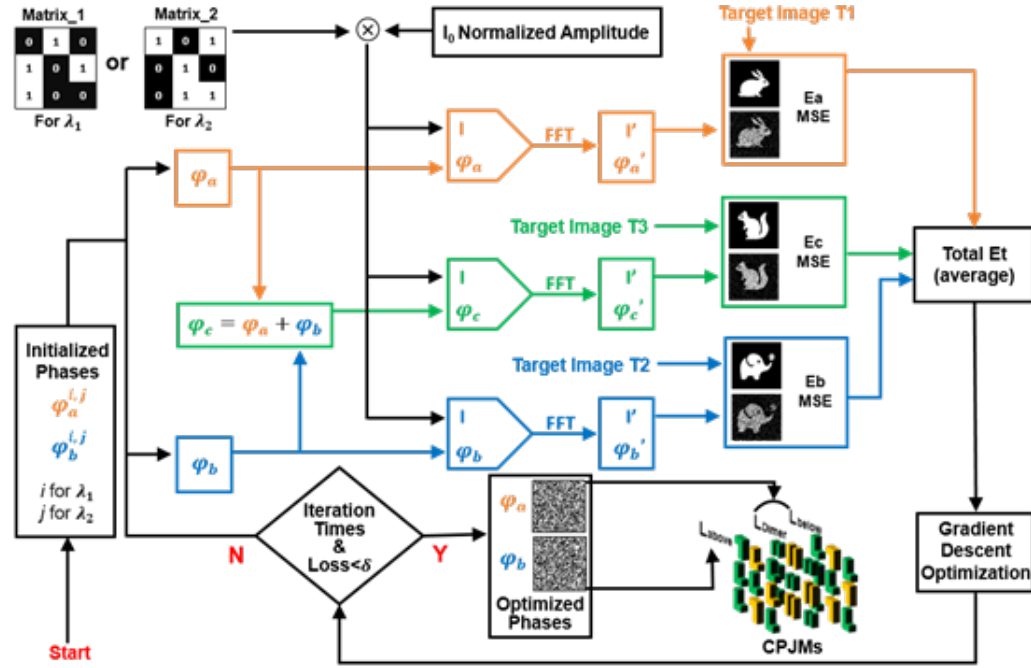


Figure 5.4: Flowchart of the retrieval optimization algorithm for the CPJM. The optimization for the 900 nm and 1200 nm operating wavelength is done separately, with randomly initialized phases ϕ_a and ϕ_b for layer L_a and L_b , respectively. The third hologram is generated via phase ϕ_c , representing the sum of ϕ_a and ϕ_b . In each iteration cycle, the incident field amplitude is reapplied whereas the calculated phase from the previous calculation step is reused. The phase profiles are improved based on a gradient descent optimization of the mean-squared error (MSE) between the target images and the current generated holograms.

The expected behavior of an example-hologram is illustrated in figure 5.5 based on a 50 x 50 pixels meta-atom array. The top row shows the target images, the second row shows the generated holograms based on the phase profiles acquired from the retrieval algorithm and the last row incorporates the optical properties of the targeted nanostructures from table 5.1. Note that the co-polarization channel T_{yy} shows worse hologram visibility compared to the cross-polarization channels; one the one hand because it requires interaction with both layers containing L-shaped antennas and on the other hand due to the presence of the 0th diffraction order. The layer alignment accuracy might arguably be the main challenge of the fabrication. In figure 5.6 the expected hologram quality for a successive layer-layer displacement of 100 nm is illustrated, causing a 200 nm displacement between L_a and L_b . The quality of the cross-polarized transmission channel still shows good visibility, as the associated holograms from layers L_a and L_b operate independently, whereas the

co-polarization channel becomes a little less pronounced. Note that this simulation does not capture the intended wavelength-multiplexing; due to the staggering of structures designed for 900 nm and 1200 nm, a misalignment of $P/2$ between L_a and L_b is expected to heavily reduce the T_{YY} -channel hologram quality.

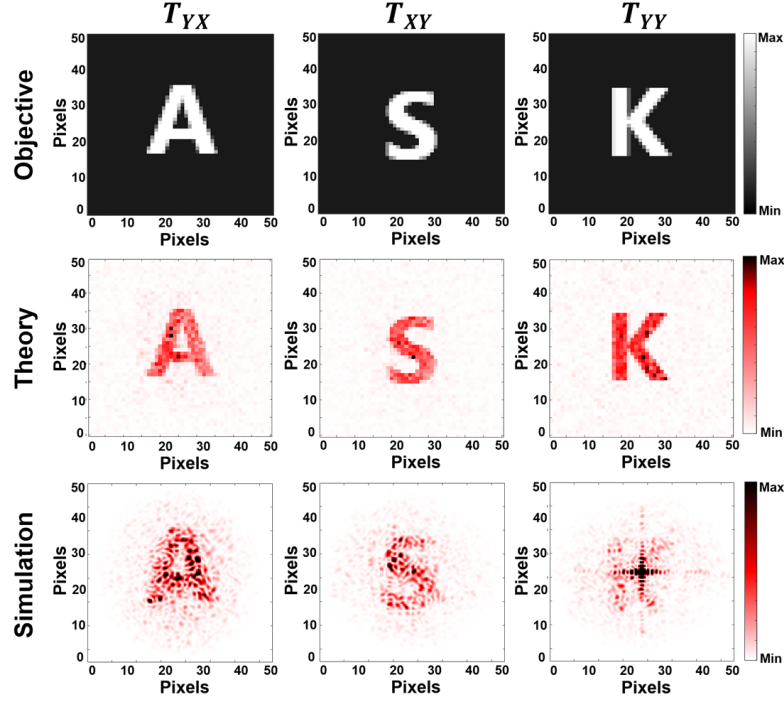


Figure 5.5: Example holograms to test the functionality of the described algorithm as well as the optimized antenna-parameters on a 50 x 50 pixels nanoantenna array. The top row shows the target images for the transmission channels T_{YX} , T_{XY} and T_{YY} , respectively. The middle row shows the generated holograms based on the optimized phase profiles from the described retrieval algorithm. The bottom row considers the transmission amplitude and the phase of the nano-antennas chosen for fabrication.

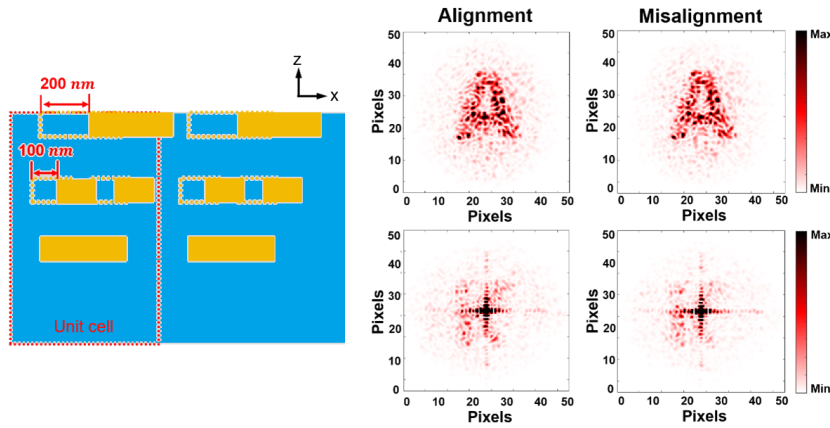


Figure 5.6: Fabrication error evaluation of 100 nm displacement per layer in x-direction on a strictly periodic unit cell. left: illustration of the misalignment in the three layer stack. right: comparison between holograms in the aligned and misaligned case for the cross- and co-polarization channel.

5.4 Fabrication

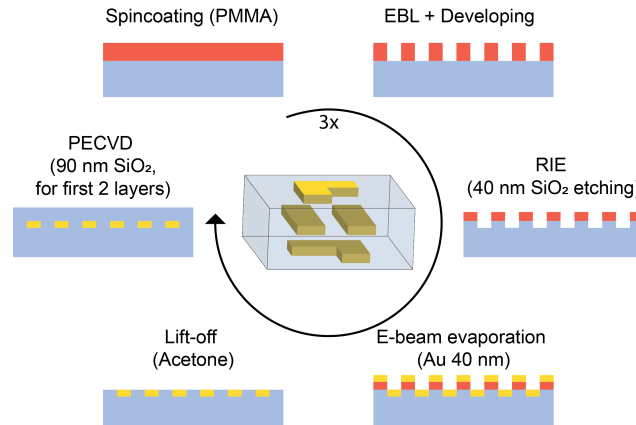


Figure 5.7: Schematic fabrication workflow. After performing standard e-beam lithography (EBL) and development, 40 nm deep holes are etched into the SiO_2 substrate. The evaporation of 40 nm Au and the following lift-off ideally lead to a flat surface that allows easier fabrication of the following layers. A 90 nm layer of SiO_2 is deposited via plasma-enhanced chemical vapor deposition (PECVD). The entire process is repeated two additional times but the SiO_2 deposition is left out after patterning the last layer thus exposing the topside of the Au nanostructures to air.

Figure 5.7 shows the schematic workflow for the sample fabrication. I spin-coated a 260 nm PMMA e-beam resist layer (baked at 180 °C for 2 min) and added a 40 nm Elektra92 layer (baked at 90 °C for 2 min) to improve conductivity. During the first e-beam lithography

(EBL, dose of $460 \frac{\mu\text{C}}{\text{cm}^2}$ at 50 kV) both the L_b -layer as well as multiple alignment markers for subsequent layers are patterned. After patterning, the Elektra92 is removed (rinse with DI-water for 60 s) and the e-beam resist is developed (90 s MIBK, 20 s IPA). The pattern is transferred into the SiO_2 substrate via reactive ion etching (RIE) using a combination of Ar- and CHF_3 -gas. After etching 40 nm of SiO_2 , 40 nm of Au are deposited via e-beam evaporation. This way, one minimizes a severely increasing surface roughness from the multi-layer lithography process. Note that the Ar-gas quickly etches the PMMA mask, therefore it is important to spin coat a sufficiently thick PMMA layer that allows for a proper lift-off of the Au-layer. The etch rate of PMMA for the etching recipe is roughly twice as large as the SiO_2 etch rate leading to a remaining PMMA layer of around 170 nm. After completing the lift-off in acetone at 60 °C, I deposited a 90 nm thick SiO_2 layer using PECVD at 300 °C. The described process is repeated two additional times. Subsequent patternings include a writefield alignment on the fabricated alignment markers, allowing for precise positioning of the L_D - and L_a -layer over the first patterning. Note that no SiO_2 layer is deposited after the last patterning, leaving the topside of the L_a -layer exposed to air.

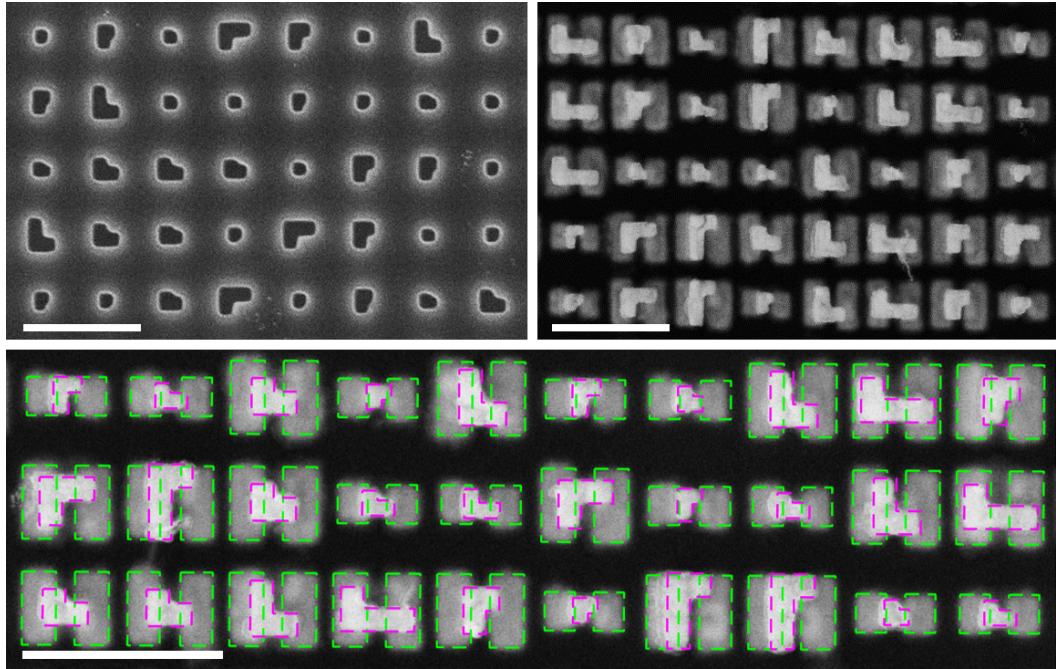


Figure 5.8: SEM micrograph of (top-left) etched holes after the first patterning procedure and (top-right) completed sample. (bottom) SEM image of completed sample with corresponding GDS-file section from layers L_B and L_D . The scale bar represents 1 μm .

Figure 5.8 (top-left) shows an SEM micrograph of the first metasurface layer (L_B) after RIE etching. This particular panel refers to an older sample-iteration where the arm lengths of the L-shaped structures were overall smaller, partially leading to apparent square-shaped structures after fabrication. (top-right) shows an image of the last layer captured at a

higher acceleration voltage of 10 kV allowing to see the dimer layer underneath as well. The bottom image shows the top layer and middle layer of the sample with an overlay from the corresponding GDS files (the patterning file-format used for the EBL). The GDS files are collectively stretched and rotated to match the dimensions and orientation of the *dimer*-antennas in the SEM image since charging effects tend to slightly distort the acquired images. With the dimer structures adjusted to its respective GDS layer, the total alignment error is visualized by the offset between the marked and patterned L-shaped structures. The offset is evaluated on 13 antennas in the presented image and evaluates to roughly 10 nm. Note that this evaluation makes the worst-case assumption of systematic displacement by only evaluating/averaging the *absolute* displacement without regards to the displacement direction that might otherwise average out to an even lower value. I want to emphasize that grid-snapping - the placement of individual nano-antennas on a coarse grid equal to the used step size in the e-beam lithography by 'rounding' the vortex positions of their polygons - can also add to this effect. The step size used during patterning was 2.5 nm. Since even with this evaluation, the displacement is found to be well within the targeted confines, it is not further investigated.

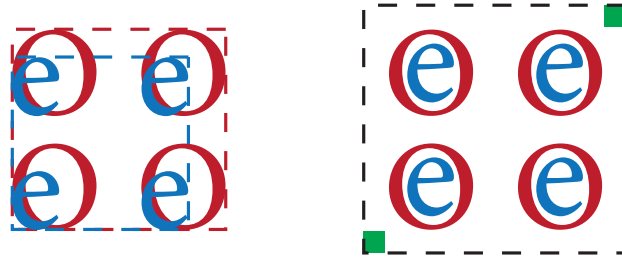


Figure 5.9: The center-alignment of two periodic structures (“O” and “e”) in two different GDS layers (red and blue) is desired. (left) Systematic misalignment of these layers introduced by un-uniform smallest-fitting rectangles created around the respective files. (right) The addition of anchoring structures (green squares) outside of the periodic array can help to unify the bottom-left corner of the individual layer, resulting in easy center-alignment.

I want to add another important detail when aligning multiple layers on top of each other. Some EBL patterning systems encompass the to-be-patterned GDS file in a smallest-fitting rectangle. The coordinate at which this GDS file is placed on a substrate then refers to the bottom-left corner of said rectangle. In our case, if the individual layers have structures of varying lateral extend, the bottom-left corner of multiple GDS files might not be uniform. This can already result in a small misalignment. To fix this, we can manually add “anchoring”-structures to the individual GDS files, preferably to the bottom-left of the main structure. This forces the fitting rectangle to move its bottom left corner further outwards to the anchoring-structure and results in easier alignment. This concept is illustrated in figure 5.9; the idea is to center-align two different layers, one in red, containing periodic arrangements of the letter “O” and the other in blue with the letter

“e”. The smallest fitting rectangle is shown as a dashed box in the color of the respective layer. On the left, the bottom-left most corner of these rectangles overlap but result in bad alignment of the letters. By adding anchoring structures, shown as a green box in the bottom left and top right corner, which is added to every layer, the smallest fitting rectangle is now uniform and shown as a dashed black box. Note that only the bottom left anchoring structure is required for this to work. Considering the design parameters of layer L_b and layer L_D , this effect would already introduce a systematic misalignment of -52.5 nm in x-direction and -45 nm in y-direction.

5.5 Experimental setup and measurements

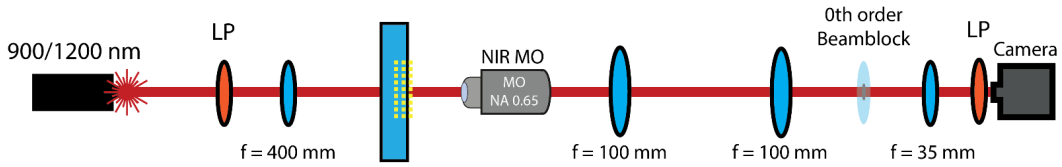


Figure 5.10: Schematic of the experimental setup. A linear polarizer (LP) and half-wave plate (HWP) are used to set the input polarization. The holograms are imaged with an NIR microscope objective (MO, NA = 0.65) and a 4f-setup in combination with an additional imaging lens in a 2f-2f configuration. The 0th order beam is blocked in the Fourier plane using a circular silver aperture.

The optical setup is shown in figure 5.10. A lens with a focal length of 400 mm focuses the beam down to the size of the metasurfaces. Imaging of the holograms is done using an NIR microscope objective (NA = 0.65, capturing the required NA of 0.6 from the hologram design) in combination with a 4f-setup as well as an additional imaging configuration. A 100 nm thick, circular silver aperture behind the 4f-setup in the Fourier plane blocks out the 0th diffraction order. The filtered holograms from this plane are then imaged with a 35 mm focal length lens in a 2f-2f configuration onto a CCD camera (Zyla5.5 from Andor). A linear polarizer between the sample and the camera allows separation of the individual polarization channels, while a half-wave plate (HWP) in front of the sample sets either x- (horizontal) or y- (vertical) polarization.

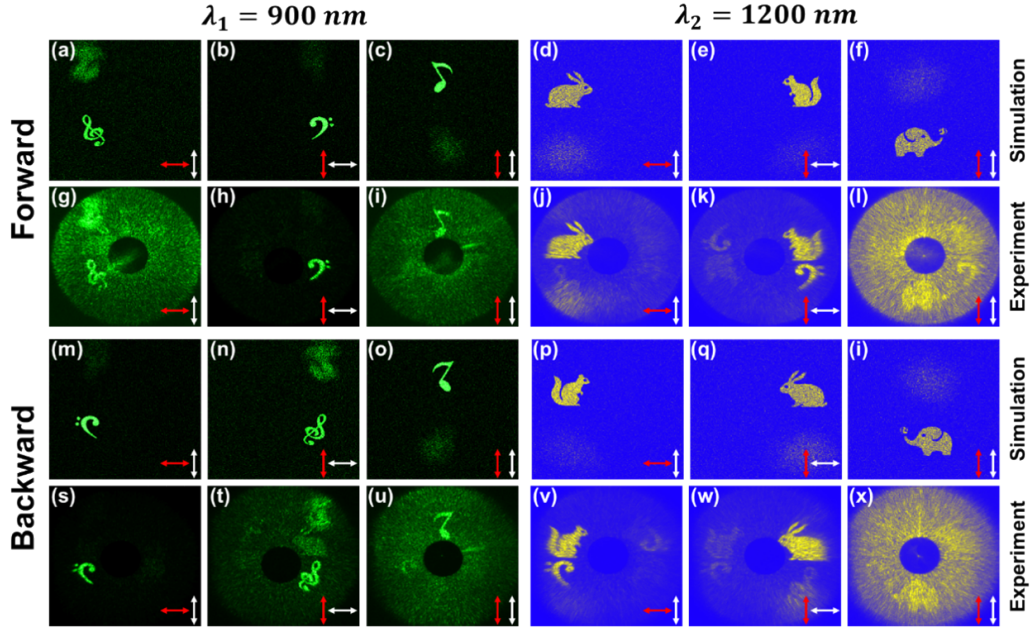


Figure 5.11: (a - f) and (m - i) show the designed holograms in forward and backward propagation direction, respectively. (g - l) and (s - x) correspond to the measured holograms. The red (white) arrow represents the detected (incident) linear polarization state. The holograms show a treble and bass clef as well as an eighth note at 900 nm and a rabbit, a squirrel and an elephant at 1200 nm. The designed holograms require an NA of 0.6 to capture. The holograms expected in forward propagation are mirrored to the backward propagation holograms. Additionally, the holograms in the cross-polarization transmission channels are interchanged between forward and backward propagation.

Figure 5.11 shows the designed and measured holograms at 900 nm and 1200 nm in forward and backward propagation for the T_{XY} , T_{YX} and T_{YY} channels, respectively. At 900 nm operation wavelength, these include a treble clef and a bass clef for the cross-polarization channels and an eighth note for the co-polarization channel, respectively. At 1200 nm, a rabbit and a squirrel are designed for the cross-polarization as well as an elephant for the co-polarization channel. We can see, that the holograms mostly match the simulated images. The cross-polarized transmission channels show a 'swapping' of the respective holograms when changing from forward to backward incidence. Furthermore, the expected mirroring that originates from the changing direction of incidence can also be observed. The elephant from the co-polarization channel at 1200 nm has a particularly low visibility, even showing transmission of the 0th diffraction order through the 100 nm thick silver aperture that is used as a k-space filter. On the other hand, the co-polarization channel at 900 nm is nicely visible. The T_{XY}^B channel that is supposed to show a squirrel at 1200 nm also clearly shows the bass clef corresponding to the hologram intended for 900 nm fundamental wavelength. Similarly, the bass clef is faintly visible in the T_{YY}^F channel, indicating cross-talk in these channels.

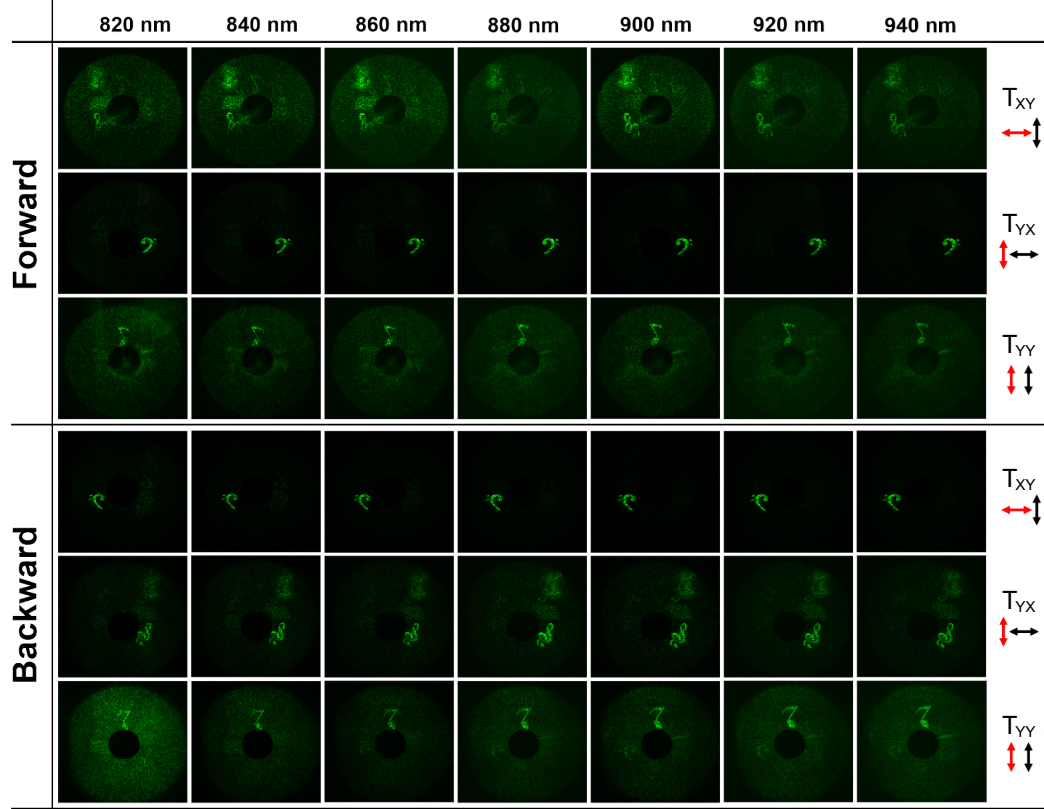


Figure 5.12: Holograms observed for a wavelength sweep from 820 nm to 940 nm in 20 nm steps. The red (black) arrow represents the detected (incident) linear polarization state. The quality of the holograms remain mostly unaffected within the tested wavelength range.

To see whether the cross-talk in channels T_{YX}^F and T_{XY}^B changes with the incident wavelength, the measurements from figure 5.11 are repeated at different fundamental wavelengths, ranging from 820 nm to 940 nm (see Fig. 5.12) and from 1160 nm to 1280 nm (see Fig. 5.13) in steps of 20 nm. Note that the wavelength is not swept entirely from 900 nm to 1200 nm as the utilized laser system has a gap in the accessible wavelength range between 940 nm and 1160 nm, separating the signal and idler wavelength. Each row in the figure is normalized to the highest detected signal within that particular row. We can see that the holograms designed for 900 nm remain mostly unaffected by a change in wavelength. The holograms designed for 1200 nm on the other hand show that the bass clef is less pronounced as the wavelength is increased, nearly vanishing at 1280 nm in the T_{YX}^F - and T_{XY}^B -channels. Similarly, the bass clef in the T_{YY}^F -channel also seems to vanish at 1260 nm, but starts to show the squirrel for even larger wavelengths instead.

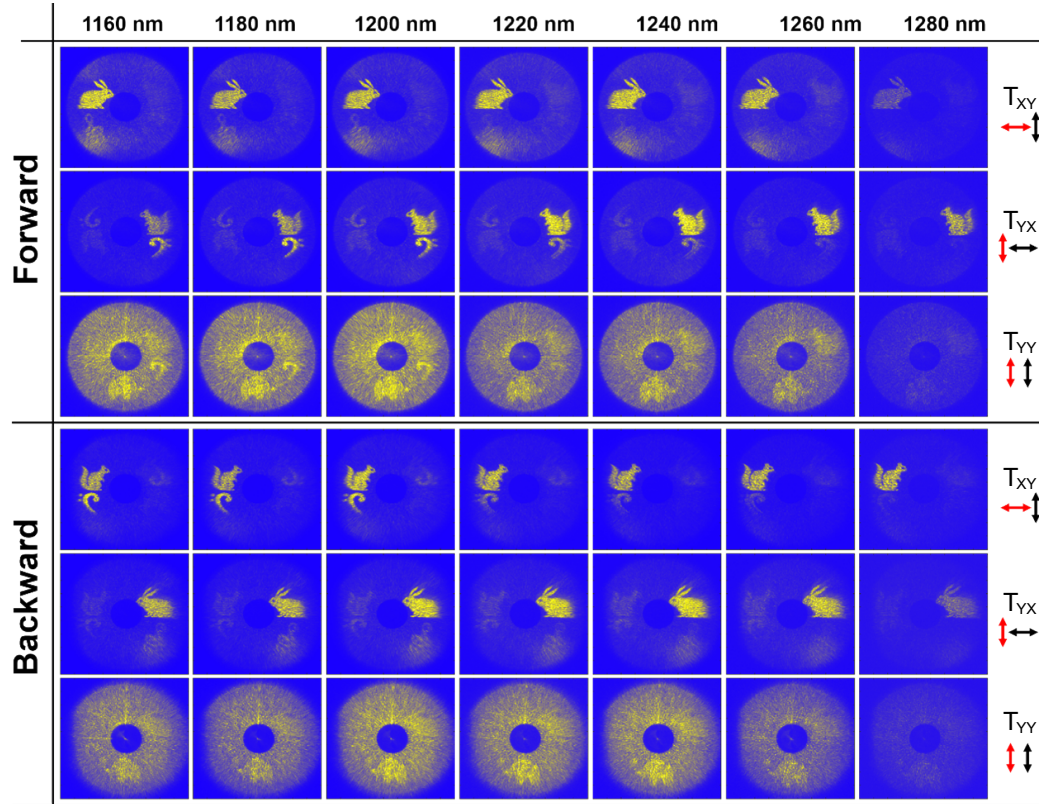


Figure 5.13: Holograms observed for a wavelength sweep from 1160 nm to 1280 nm in 20 nm steps. The red (black) arrow represents the detected (incident) linear polarization state. The hologram designed to show the squirrel shows a decrease in cross-talk (vanishing bass clef) as the wavelength is increased. Additionally, the contrast of the hologram showing the elephant seems to improve for an increasing wavelength.

One shortcoming of the established fabrication is the heat treatment of the gold antennas during deposition of the SiO_2 layer in the PECVD. As discussed in the pulse shaping chapter, treatment of these antennas already shows a change in their optical behavior at temperatures of 200 °C. In addition to that, since the layer L_a is not covered with a spacer layer, these antennas were not exposed to the same temperature. Since only 6 distinct antenna parameter sets are used to cover the 2π phase range, introducing a performance shift in one or two of these parameter sets can already result in a rapid decline of hologram quality. Keep in mind that half of these parameter sets are the rotated version of other parameters. Therefore a change in optical performance immediately impacts both the rotated and the unrotated antenna design. Ironically, layer L_a contains the phase information for the bass clef and the squirrel - the channel that exhibits the most cross-talk - and is also the layer that was not treated at 300 °C. Following the observation from the pulse shaping chapter and assuming that the optical properties of this layer would blue-shift after treatment, the strong cross-talk is expected at smaller wavelengths and would theoretically improve the hologram quality at 1200 nm. However, I did not test this

assumption.

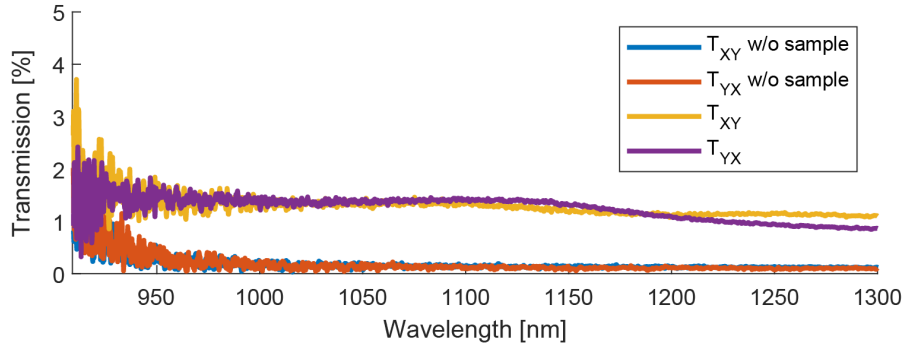


Figure 5.14: FTIR transmission measurement of the samples cross-polarization channels T_{YX} and T_{XY} in forward direction. Crossed polarizer settings without a sample are taken for reference. The sample's transmission is around 1.5% from 900 nm to 1200 nm, captured with a numerical aperture of 0.5. Note that below 900 nm, the detected signal shows a lot of noise.

Lastly, figure 5.14 shows the transmission of both cross-polarization transmission channels, captured via FTIR measurements (Bruker Vertex 70 with a Hyperion microscope) in forward direction. To show that crossed polarizer settings in the measurement drop off to 0%, measurements without the sample are added for reference. The detected transmission is an indicator for the conversion efficiency in these two channels, provided that cross talk is not severe. Based off of figure 5.3 we expect a transmission efficiency of $\approx 9\%$ (corresponding to the plotted transmission amplitude of 0.3), while the measured transmission is only around 1.5 % between 900 nm and 1200 nm. Note that the detecting numerical aperture of the microscope objective is 0.5 while the samples require a numerical aperture of up to 0.6 to be fully captured, consequently underestimating the conversion efficiency a bit. Nevertheless, we see that the measured transmission is constant but lower than anticipated, which might be linked to the high temperature treatment.

5.6 Conclusion

In this chapter I presented a triple layer plasmonic Janus metasurface that breaks reciprocity, i.e. its optical properties are dependent on the direction of incidence. This functionality comes at the cost of two overlay lithography steps with a targeted misalignment of less than 100 nm - a precision readily achievable with modern lithography systems. The three layer system is capable of generating six holograms, multiplexed in two different wavelengths and three polarization channels. While the transmission matrix of typical metasurfaces - despite their air-substrate interface - exhibit out-of-plane mirror symmetry [84], meaning that the optical behavior in forward and backward direction as well as their off-diagonal transmission matrix elements are identical, here we realize non-reciprocity by introducing

polarization filtering inside the three layer system. This work can be considered as an extension of the two layer system realized by Frese et al. [91], which incorporates a single hologram on the account that only one layer of L-shaped antennas is present. In our design, the realization of three holograms per wavelength is possible because of the presence of an additional layer consisting of L-shaped plasmonic nano-antennas.

And while the measured cross-polarization transmission is by a factor of up to 6 lower than anticipated, we nevertheless found high quality holograms with low crosstalk in all but one cross-transmission polarization/wavelength channel. The strong cross talk observed in one of these six transmission channels belongs to the last fabricated layer (L_a) and is found to vanish for wavelengths larger than 1280 nm. Arguably, the cross-talk from said channel could be fixed by high temperature treatment of the last layer, causing a blue shift of the optical response and thereby improving the hologram quality at 1200 nm. However, as the heat treatment is a rather uncontrolled way of adjusting the sample performance, the fabrication would benefit from a lower temperature SiO_2 deposition process to avoid altering the optical properties of the gold antennas.

While the observed shortcomings are mostly on the efficiency and fabrication side, to which improvements were suggested, design wise we find that the sample performs as intended; we see multiple independent holograms and directionally dependent access to these holograms with respect to incident and detected polarization. The design features asymmetric processing of information and could in principle be extended to another independent hologram channel provided that the co-polarization from a single layer can cover a full 2π phase range. Additionally, while plasmonic structures are easier to fabricate than dielectric nano-pillars, they also impose a restriction on simultaneous transmission amplitude and phase requirements. In addition to an increase in fabrication complexity, a fourth hologram channel would also come at an increase in design complexity, as the $t_{La,xx}$ and $t_{Lb,xx}$ channels impact all but the T_{YY} transmission channels. Overall, the presented functionality can be convenient in compact, laser based systems by acting as optical isolators by preventing back reflections into sensitive laser cavities.

A manuscript on this work is currently handed in to “Advanced Materials”.

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Publications and conference contributions

Publications

Liu, B.; **Geromel, R.**; Su, Z.; Guo, K.; Wang, Y.; Guo, Z.; Huang, L.; Zentgraf, T. “Non-linear Dielectric Geometric-Phase Metasurface with Simultaneous Structure and Lattice Symmetry Design”. *ACS Photonics* **2023**, 10, 4357-4366.

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Currently submitted manuscripts

Spedt, V.; Meier, F.; **Geromel, R.**; Mahler, P.; Henksmeier, T.; Reuter, D.; Zentgraf, T.; Meier, C. “Hybrid GaAs (111)/GaAs (100) PIN photodiodes for metasurface-assisted nonlinear upconversion detection”. *Opt. Express*.

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Conference contributions

Geromel, R.; Georgi, P.; Protte, M.; Bartley, T.; Huang, L.; Zentgraf, T. “Dispersion Control with Integrated Plasmonic Metasurfaces” [conference talk]. *CLEO 2023: Conference on Lasers and Electro-Optics*, San Jose, CA, United States **2023**.

Geromel, R.; Georgi, P.; Protte, M.; Lei, S.; Bartley, T.; Huang, L.; Zentgraf, T. “Optical pulse-shaping with plasmonic metasurfaces” [invited talk]. *META 2023: 13th International Conference on Metamaterials, Photonic Crystals and Plasmonics*, Paris, France, **2023**.

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