CONTROLLING LIGHT WITH SPATIALLY AND TEMPORALLY PHASE-MODULATED METASURFACES

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Xuexue Guo

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The dissertation of Xuexue Guo was reviewed and approved by the following:

Xingjie Ni
Assistant Professor of Electrical Engineering
Dissertation Advisor
Chair of Committee

Zhiwen Liu
Professor of Electrical Engineering

Noel Christopher Giebink
Associate Professor of Electrical Engineering

Christine Keating
Distinguished Professor of Chemistry

Kultegin Aydin
Professor of Electrical Engineering
Head of the Department of Electrical Engineering
Metasurface – consisting of artificially engineered subwavelength meta-atoms arranged on a surface – has revolutionized the way of manipulating light properties, such as amplitude, phase and polarization, and has impact in many fields ranging from material science, optics and photonics, quantum optics to nanoscience and life science. Since its emergence in the early 2010s, optical metasurface grows exponentially, with demonstrations of achromatic lens, skin cloak, compact holograms, optical vortex phase plate, beam steering, mode/polarization converter, efficient nonlinear generations, etc. However, current metasurfaces are designed based upon spatial structure engineering, such as geometries and orientations of constituent meta-atoms, and their properties are fixed at the time of fabrication. This limits their applications in emerging technologies requiring active control of light, such as virtual reality, augmented reality, holographic display, tunable and adaptive optics and light ranging and detection (LIDAR). On the other hand, due to the lack of efficient temporal modulation technique, the study of novel physical phenomena such as nonreciprocity, temporal refraction and topological photonics based on time-varying optical metasurfaces is limited to theoretical proposals. In this dissertation, the largely uncharted properties of spatially and temporally phase-modulated metasurfaces have been explored. I will show how spatiotemporal phase gradient metasurfaces break Lorentz reciprocity and realize novel nonreciprocal light propagation. Another interesting phenomenon of time-varying systems is that parametric resonances can be leveraged to enhance nonlinear generations. I created a miniature parametric oscillator based on optical metasurfaces and demonstrated extraordinary nonlinear behaviors. In addition, optical metasurfaces integrated photonic chips have been studied, offering a promising platform for manipulating light both in free space and waveguides. Leveraging the versatile and compact metasurface, different on-chip functions, including beam deflection, focusing and orbital angular momentum (OAM) lasing, have been demonstrated.
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Chapter 1

Introduction

1.1 Metamaterials and Metasurfaces – Controlling Light at the Nanoscale

The emerging field of metamaterials and metasurfaces has captured the imaginations of scientists and engineers in the hope to create materials where the interaction between electromagnetic waves and matter is fundamentally altered to produce fascinating new effects. A metamaterial, consisting of engineered subwavelength metallic/dielectric structures also known as meta-atoms, is an artificial material whose properties are determined by the structuring of the material rather than the constituent materials. During the last 20 years, this field has attracted great interest and demonstrated some ground-breaking electromagnetic phenomena and useful applications. One of the major feats of the metamaterial community is achieving materials with negative refractive index \(^1\text{–}^4\), which is traditionally tied to super-resolution optical imaging \(^5\text{–}^6\). The other hallmark is achieving a cloaking device by spatially changing material properties in a well-designed manner \(^7\text{–}^8\). However, the potential applications of metamaterials are inherently limited by the high loss / dispersion related to the resonant metallic structures and the propagation effect, and the demanding fabrication process required by three-dimensional (3D) structures.

Planar metamaterials, also known as metasurfaces, not only provide a practical solution to some of the challenges facing 3D metamaterials, but also represent a new paradigm in the physics of metamaterials. Metasurface distinguishes itself from conventional metamaterials by displaying spatially varying optical responses over an optically thin interface. Arrays of meta-atoms with subwavelength separation can have spatially varying geometries or material compositions, thus leading to spatial-variant optical responses, \(i.e.\) amplitude, phase and polarization. Thanks to the subwavelength thickness of metasurfaces, the undesirable loss can be greatly suppressed. And the planar structures can be readily fabricated using established
semiconductor process. In summary, metasurfaces inherit the novel properties of bulk metamaterials, such as enhanced light-matter interactions and designable material permittivity / permeability, and overcome the challenges facing bulk metamaterials. Therefore, in recent 10 years metasurfaces have dominated the general field of metamaterials research and opened up numerous application opportunities.

Historically, the origin of metasurfaces can be dated back to the early 20th century when Robert W. Wood discovered dark region in the reflectance spectra of a subwavelength metallic grating. Later, based on the simple grating structures, frequency-selective surfaces (FSS) have been invented and broadly used in microwave absorption, radar cross section reduction and other millimeter and terahertz wave applications. In optical frequencies, this concept remained dormant until the development of optical metamaterial. Two pioneering works have laid the foundation for optical metasurfaces: in the early 2000s, Hasman’s group rediscovered Pancharatnan-Berry phase in optical grating and used it for polarization control; ten years later, Capasso’s group demonstrated generalized laws of refraction based on space phase gradient plasmonic metasurfaces. Since then, this field grows exponentially, with demonstrations of achromatic lens, skin cloak, compact holograms, optical vortex phase plate, beam steering, mode / polarization converter, efficient nonlinear generations and many interesting physical phenomena such as optical spin Hall Effect and bound states in the continuum.

1.2 Space-time Metasurfaces

As discussed above, metasurfaces based on meta-atoms array displaying spatial-variant phase/amplitude/polarization responses have provided a powerful means to manipulate the wavefront of light, ushering in a new era for flat optics. Despite these impressive achievements, most metasurfaces are static and passive in nature, which hinders their applications in areas demanding active and reconfigurable components, such as optical / quantum computation and communications, light detection and ranging (LiDAR), augmented reality and holographic display. On the other hand, as governed by Maxwell’s
equation, time plays an equivalently important role in the description of electromagnetic waves, hence it is essential to factor time in for the design of metasurfaces. For example, taking advantage of space-time duality, temporal imaging \(^{30}\) and cloaking \(^{31}\) have been demonstrated in conventional optical systems. Structuring metasurfaces in time domain will bring forth new physical effects, such as nonreciprocity \(^{32}\) and Doppler-like frequency shift \(^{33,34}\), and give rise to novel optical devices like optical isolators without the need of magnetic field. Both the vast range of applications and the fascinating new physics arisen from space-time metasurfaces have driven the exploration of this emerging field. This section summarizes the current state of the field and provides an overview on our endeavors to build spatially and temporally phase-modulated metasurfaces for purposes of breaking reciprocity and nonlinear generation enhancement.

1.2.1 Dynamically Modulated Metasurfaces

In contrast to the intensive demand for spatiotemporal metasurfaces, relevant researches have lagged behind due to the difficulties in implementing efficient and fast temporal modulation on the ultra-thin metasurfaces especially in optical regime. Commonly used technologies can be summarized into three categories based on the external stimuli used to induce dynamic changes in material or structure properties.

The mechanical approach modifies the electromagnetic (EM) response of metasurfaces by altering the geometrical structures and spatial arrangement of meta-atoms \(^{34,35}\). In particular, well-established micro-electro-mechanical (MEMS) and nano-electro-mechanical (NEMS) technologies provide nanometer scale control on the positioning of nanostructures at a frequency ranging from kilohertz to megahertz, leading to compact and light-weight tunable / reconfigurable devices that operate at moderate speed. For example, A. L. Holsteen \textit{et al.} experimentally demonstrated dynamic beam shaping and temporal color mixing based on silicon nanowire meta-atoms that are movable above a silicon substrate \(^{36}\). In addition, stretching of flexible substrates can change the periodicity of meta-atoms array, thus changing the near-field and far-field interactions between meta-atoms and the EM response of the whole metasurfaces \(^{34,35}\). Tunable metalens \(^{37}\),
hologram 38, color filter 39 and beam steering device 40 have been demonstrated based on stretchable substrates.

The mature electronic and semiconductor technologies have laid a groundwork for modulating the metasurfaces made of electrically responsive materials, such as varactor / PIN diodes, liquid crystal, transparent conducting oxides (TCOs), doped semiconductors and two-dimension (2D) materials. In the microwave regime, metasurfaces consisting of varactor / PIN diodes meta-atoms can be tuned by applied voltage 29,41. But such design strategy cannot be extended to terahertz (THz) and optical regime due to the lack of suitable varactor diodes that can respond at such a fast speed. For THz and optical applications, doped semiconductors 42,43, 2D materials 44–46 and TCOs 47–49 are excellent choices, because their conductivities can be tuned with carrier doping through electrical gating. In particular, epsilon-near-zero (ENZ) materials made by doped semiconductors or transparent conducting oxides have shown prominent permittivity change when operating near ENZ point 50. Additionally, since the resonant properties of meta-atoms are sensitive to their ambient dielectric properties, incorporated with electrically controlled liquid crystal, metamaterials and metasurfaces have shown various dynamic functions, such as spatial light modulation 51, color tuning 52 and polarization rotation. Overall, electrical tuning method features moderate to high modulation speed (MHz to GHz), broad bandwidth and large dynamic range. However, it hit a wall in terms of creating ultrafast (THz) modulation.

Optical actuation is by far the most versatile modulation method that covers a broad range of modulation speeds and works for almost all kinds of materials 53,54. According to different mechanisms underlying the change in material properties, it can be further classified into: thermal-optical nonlinearity 53,55, optical induced phase change 56–59, photo-carrier doping and optical nonlinearity (Fig. 1-1). Thermal-optical effect is normally used for low frequency modulation (kHz) 60 due to the long relaxation time from phonon-phonon scattering. It is possible to use pulsed laser heating on nanostructures to boost and speed up the modulation via ultra-fast heating 61,62 and thermal expansion 63. Thanks to the large dielectric property change induced by local temperature change over a threshold value $T_c$. Phase change materials, like VO$_2$ 64
and germanium (Ge)–antimony (Sb)–tellurium (Te) alloys, hybrid with optical nanoantennas provide a promising opportunities for optically tunable applications in the near infrared region. But the energy required to induce phase change is generally higher than that of other optical modulation mechanisms. Some novel physical effects such as nonreciprocity and frequency shifting may only be accessible at high modulation speed, where it is inevitable to use ultrafast (10 fs – 0.5 ps) photo-carrier excitation or optical nonlinear effect in metallic and dielectric meta-atoms. In particular, the promising optical functionalities and low loss offered by metasurfaces made of high index dielectrics, combined with the CMOS (complementary metal oxide semiconductor) compatibility, have triggered a surge of effort on dynamically modulated dielectric metasurfaces. Assisted by the resonant dielectric meta-atoms, prominent intensity dependent refractive indices change can be induced by nonlinear Kerr effect and two photon absorption (TPA), leading to modification of the resonance and thus facilitating the phase and amplitude tuning of the transmitted / reflected light. In addition, free-carrier injection induced by optical pumping in dielectrics can cause a larger index change than that of metals, because the low electron density of dielectrics makes it more sensitive to photo excited carrier density change. Recently, ENZ material has gained more attention due to its extraordinary nonlinear properties arisen from plasmon-induced hot electrons. As the permittivity approaches zero, the boundary condition ensures that the electric field inside the ENZ material skyrockets, therefore the nonlinear interactions become significantly stronger.

Although we have witnessed great progress in the field of dynamic metasurfaces, most efforts are devoted to upgrading the static metasurfaces to active ones, demonstrating tunable and reconfigurable spatial light modulation functions such as phase/amplitude/polarization modulation, adaptive optics, tuning spectral response and thermal emission. Entirely new physical effects will emerge under ultrafast temporal modulation through photo-carrier doping and optical nonlinear effect, such as breaking Lorentz reciprocity and inducing frequency shift, which is the focus of this dissertation.
1.2.2 Space-time Phase Gradient Metasurfaces for Breaking Lorentz Reciprocity

Lorentz reciprocity is a fundamental and universal rule governing the electromagnetic wave behaviors in a linear and time invariant system\textsuperscript{32}. Breaking Lorentz reciprocity can achieve optical isolation which is key to protecting laser system and building full-duplex communication system. Conventionally, optical isolators are built upon bulky magneto-optic crystals with asymmetric dielectric properties, which exhibits large loss and hinders on-chip integration. Breaking time reversal symmetry is an alternative approach to nonreciprocity. By introducing a temporally phase modulation to the static space phase gradient metasurfaces, several groups have predicted nonreciprocal light reflection/refraction/transmission\textsuperscript{78,79} based on space-time metasurfaces and experimentally demonstrated nonreciprocal microwave propagation\textsuperscript{80}. It should be noted that in order to achieve efficient isolation for the forward and backward propagating optical waves, temporal modulation frequency needs to exceed the signal bandwidth, which requires ultrafast optical modulation technologies. Some have avoided the demanding ultra-high frequency modulation requirement by using narrow band optical signals in waveguide systems\textsuperscript{81–84}. However, it remains a
challenge to experimentally demonstrate nonreciprocal free-space light propagation based on space-time phase gradient metasurfaces. We will introduce an ultrafast (~ 2.8 THz) temporal modulation method based on nonlinear Kerr effect and create a spatiotemporal phase modulated metasurface for nonreciprocal light reflection in the near infrared. We also observed the effect known as time refraction, which states that when light crosses a temporal boundary, its frequency will change.

1.2.3 Parametrically Excited Metasurfaces for Second Harmonic Generation

As widely explored in mechanical and electronic systems, parametric oscillations leverage the temporal modulation of a system to generate or amplify nonlinear signals. Due to the resonance enhanced local field and relaxed phase matching conditions, metasurfaces have made tremendous progress in nonlinear optics. Traditionally, optical parametric oscillators are constructed with large optical cavities and bulky nonlinear crystals, which limits their miniaturization and on-chip integration. Recent studies on nonlinear metasurfaces - consist of artificial subwavelength building blocks known as meta-atoms - have shown great potential for boosting nonlinear generation without phase matching constraint, and agile control over the properties of nonlinear signals. Leveraging the resonance enhanced local field, nonlinear signals are enhanced within a subwavelength interaction length, which is essential for reducing device footprint. By adding the temporal degree of freedom to metasurface design, we can unlock entirely new ways for engineering nonlinear generations. We designed amorphous silicon (α-Si) meta-atoms showing prominent time-varying resonance under travelling-wave modulation, and experimentally demonstrated greatly enhanced second harmonic generation (SHG).
1.2.4 Controlling Free Space Light with Guided-wave-driven Metasurfaces

The ever-growing demand for photonic integrated circuits (PICs) to complement and replace electronic chips has triggered the development of various compact, lightweight and integratable functional devices, among which optical metasurfaces are promising candidates for revolutionizing the next generation integrated photonics. So far, most metasurfaces work with free space light input, which makes it difficult for on-chip integration. We combined synergically two powerful, complimentary technologies: integrated photonics and metasurfaces, and developed a hybrid architecture where metasurfaces are directly driven by guided waves to realize complex free-space functions. We placed subwavelength-sized meta-atoms on top of photonic integrated components. In contrast to existing metasurfaces that operate with both input and output light in free space, our integrated metasurface bridges guided waves inside a waveguide with free-space ones. Through it the guided light is tapered into free space and molded into desired light fields. Meanwhile, multiple metasurfaces can be connected via waveguides to achieve different free-space functions simultaneously.

1.3 Research Objectives and Organization of the Dissertation

The primary goal of this dissertation is to provide a comprehensive study on space-time metasurfaces, including design principles, simulations, temporal modulation implementations, sample fabrications and experimental demonstrations. I will investigate in general the novel physical phenomena and new applications brought forth by ultrafast temporal modulation of optical metasurfaces. In addition, to further extend the potential of metasurfaces for next-generation optics and photonics application, guided-wave-driven metasurfaces are developed and demonstrated.

First, chapter 2 to 4 investigate the novel physical effects, such as nonreciprocity, time refraction and parametric oscillation, based on space-time phase gradient metasurfaces. Chapter 2 discusses potential means for creating ultrafast temporal modulation which makes those novel physical phenomena accessible.
Chapter 3 starts with theoretical introduction on Lorentz reciprocity and available methods to break it, with an emphasis on the relation between time-reversal symmetry and reciprocity. Then it elaborates on how to design and implement the spatiotemporal phase modulated metasurfaces and demonstrate nonreciprocal light reflection and time refraction. Chapter 4 introduces another interesting effect induced by temporal modulation - parametric oscillation and enhanced second harmonic generation.

Second, chapter 5 investigate a new type of metasurfaces that work with both guided waves and free space waves. It starts with an introduction to the general design methodology, then demonstrates the manipulation of guided waves, such as beam deflection and focusing. Finally, it discusses how to leverage the broken spatial inversion symmetry induced by metasurfaces and demonstrate orbital angular momentum microring laser. Finally, I conclude in chapter 8 by summarizing all novel physical phenomena demonstrated on space-time metasurfaces.
Chapter 2

Ultrafast Temporal Phase Modulation of Optical Metasurfaces

This chapter presents the principle and implementation of the ultrafast temporal modulation based on third order nonlinear Kerr effect, which is the enabling technology for space-time metasurfaces. Third order nonlinearity exists in all kinds of materials, making it a versatile modulation mechanism for various applications. Discussions will be made to establish potential ways for enhancing effective temporal modulation based on the choice of materials and structures. In particular, I will describe the design of metasurfaces with strong temporal permittivity/phase modulation based on dielectric materials – amorphous silicon, and the experimental implementation of heterodyne optical modulation.

2.1 Background

2.1.1 Dynamic Optical Modulation

The increasing demand for tunable and reconfigurable micro- and nanophotonic devices drives the development of efficient modulation methods to change optical properties in a controllable manner, among which optical modulations are the most flexible and versatile ones. Under intense light excitation, dielectric properties of materials will undergo various changes, depending on the dominant light-matter interactions happening in the specific material. In general, light induced property change can be represented by the nonlinear polarization equation:

\[ P = \chi^{(1)}E(\omega) + \chi^{(2)}E(\omega)E(\omega) + \chi^{(3)}E(\omega)E(\omega)E(\omega) + \ldots \]  

(2-1)
where $\chi^{(n)}$ is the nth order optical susceptibility. The first term on the right-hand side of equation (2-1) determines the linear refractive index as $n = \sqrt{1 + \chi^{(1)}/\varepsilon_0}$ ($\varepsilon_0$ is the vacuum permittivity). Second order nonlinear process often generates new frequencies, which cannot make effective contribution to the linear dielectric property of the material, unless one uses a dc electric field to generate a frequency component at $\omega$, which is widely used in active terahertz metadevices \cite{42} based on electro-optical modulation. The same holds for other even order nonlinear processes. In contrast, first and third order (odd order) processes contributes to the dielectric property tuning without frequency conversions. In particular, third order nonlinear effect stands out due to its universality, ultrafast modulation speed and large modulation depth \cite{53,54}. Equation (2-1) presents a macroscopic picture of light-matter interactions where details of interactions are embedded in the simplified expression of nonlinear susceptibilities. But it is not informative enough to cast light on different light modulation mechanisms and their performance metrics, such as the energy consumption, modulation depth ($\Delta n$) and the recovery time of response. We need to look further into details of physical mechanisms underlying the ultrafast (hundreds of GHz to THz) optical modulation.

According to a generalized Drude-Sommerfeld model \cite{98}, material optical properties originate from the optical responses of bound and free electrons in the valence and conduction band, respectively.

\[
\varepsilon(\omega) = \varepsilon_\infty - \frac{\omega_p^2}{\omega(\omega + i\Gamma)}
\]  

(2-2)

where $\varepsilon_\infty$ is the permittivity at high frequency (i.e. related to inter-band transitions), $\omega_p = \sqrt{e^2 N / \varepsilon_0 m^*}$ is the plasma frequency ($e$ is electron charge, $N$ is free carrier density and $m^*$ is effective mass) and $\Gamma$ is the damping rate. The refractive index can be tuned by varying the plasma frequency $\omega_p$ and damping rate through photo injection of free carriers. In general, three physical effects are responsible for the change of optical properties \cite{71,99,100} in a strongly excited semiconductor: (1) state and band filling, (2) renormalization of the band structure, and (3) the free-carrier response (related to the Drude term in equation (2-2)). The first two factors are included in the first term in equation (2-2), being important when the free-carrier density
($>10^{20}\ \text{cm}^{-3}$) is comparable to the capacity of the conduction band, and the wavelength exhibits large dispersion $(dc/d\omega)$ \textsuperscript{91}. The third contribution plays the most important role in the infrared range at relatively low intensities \textsuperscript{101}. In order to describe the dynamics of light-induced electron-hole plasma generation, we used the rate equation introduced in the literature \textsuperscript{99,102},

\[
\frac{dN_{fc}}{dt} = -\frac{N_{fc}}{\tau} + \frac{W_1}{h\omega} + \frac{W_2}{2h\omega}
\]

(2-3)

where $W_1,2$ are the volume-averaged dissipation rates due to one- and two-photon absorption (TPA), respectively, and $\tau$ is the free-carrier relaxation time. The dissipation rates are written as

$W_1 = \frac{\omega}{8\pi}\langle I \rangle \text{Im}(\varepsilon)$ and $W_2 = \frac{\omega}{8\pi}\langle I^2 \rangle \text{Im}(\chi^{(3)})$, where $\langle \rangle$ denotes averaging of the exciting field intensity over the volume. Noting the different power dependence on intensity $I$, TPA will dominate over one-photon process at high intensities \textsuperscript{99}. The relaxation time of electron-hole plasma, which determines the optical modulation speed, typically lies in the range 1-100 ps \textsuperscript{99}. Some have reported sub-picosecond decay time due to Auger recombination process when the photoexcited electron-hole plasma density exceeds $10^{19}\ \text{cm}^{-3}$ \textsuperscript{102-104}. But in general, the free-carrier induced change in semiconductors is not suitable for ultrafast modulation purpose due to a relatively long relaxation time. In plasmonic materials, inter-band excitations of free electrons have a relatively slow recovery time due to a longer relaxation process as compared with intra-band excitations \textsuperscript{105,106}. Instead, the dynamics of intra-band excitation of hot-electron gas dominates the Kerr-like index change with a typical relaxation time of picoseconds.

As mentioned earlier, TPA plays an important role in the tuning of materials’ properties. According to Kramer–Kronig relation \textsuperscript{97}, an absorption induced change in the imaginary part of refractive index is accompanied by a change in the real part of index. TPA is then linked with an intensity dependent real index change. And the intensity dependent refractive index is expressed as

\[
n = n_0 + (n_2 + i\frac{\lambda}{4\pi} \beta_2)I
\]

(2-4)
where $n_0$ represents the weak-field refractive index, $n_2 = \frac{3 \text{Re}\{\chi^{(3)}(\omega, \omega, -\omega, \omega)\}}{4n_0 \text{Re}(n_0)e_0c}$ is the nonlinear Kerr index and $\beta_2 = \frac{3\pi \text{Im}\{\chi^{(3)}(\omega, \omega, -\omega, \omega)\}}{\lambda n_0 \text{Re}(n_0)e_0c}$ is the TPA coefficient. An efficient implementation of the modulation requires strong light intensity and long interaction length. In metamaterials and metasurfaces, one can design and enhance light-matter interactions by leveraging a variety of resonances and therefore increasing the refractive index change. In these structures, the intensity-dependent tuning of refractive index modifies the resonant properties, causing resonance frequency shift and spectral broadening. Therefore the self-modulation of transmission/reflection/absorption can be fulfilled by bringing the input light in or out of resonance with the structure. Unfortunately, TPA absorption generates free carriers in the conduction band, which acts as a source for free carrier absorption and deteriorates the contrast in resonance shift induced by Kerr effect. In addition, though these third order nonlinear processes are instantaneous, free carriers generated by TPA have long lifetime and will ultimately slow down the optical modulation. Measures should be taken to optimize the modulation depth and speed with judicious choice of materials and structures.

2.1.2 Materials and Structures

Deep subwavelength plasmonic nanostructures have shown Kerr-like nonlinearity originated from hot electrons excited by surface plasmon resonance enhanced light field. Intra-band optical excitation of metallic structures generates a non-thermal electron gas in its conduction band. These short-lived electrons will quickly thermalize within a sub-picosecond time frame due to electron-electron scattering, and form a hot-electron gas with a characteristic temperature $T_{e\text{hot}}$. The dynamics of hot-electron gas can be described using a phenomenological two-temperature model.
where \( E_{nt} \) is the non-thermal energy density stored in the excited electron gas, \( P \) is the time dependent absorbed power density, \( G \) is the electron-phonon coupling constant, \( C_e \) is the electron heat capacity, \( C_l \) is the lattice heat capacity and \( T_l \) is the lattice temperature. Re-writing equation (2-2) in the context of a hot-electron system, the dielectric function becomes

\[
\varepsilon(\omega) = \varepsilon_{\infty} - \frac{\omega_p^2}{\omega(\omega + i\Gamma(T_e))},
\]

where the damping rate is dependent on the electron temperature (or absorbed light intensity) and the plasma frequency can be modified by the non-parabolic conduction band of transparent conductive oxides \(118-120\). The electron-electron scattering rate \( \Gamma_{e-e} \) provides the primary source of the modification of the damping term. Due to the fact that \( \Gamma_{e-e} \propto T_e^2 \) an increased hot electron temperature leads to a larger change in the permittivity \(109,114,117\). Capabilities of metal for optical modulation \(118-120\) have been explored using plasmonic nanostructures, and three orders of magnitude increase in optical Kerr effect \( (\chi^{(3)} \approx 10^{-7} - 10^{-8} \text{ cm}^2/\text{W}) \) has been reported \(112,121\) as compared with bulk metals and dielectrics \(97\). The recovery of the permittivity happens through the electron-phonon scattering assisted thermalization process, which is in the range of several picoseconds. According to equation (2-5), the increased electron temperature results in a slower recovery process, limiting the modulation speed \(114,115\). This creates a dilemma, on the one hand, intense light excitation facilitates a large permittivity change; on the other hand, it results in a slower relaxation process. A recent paper \(122\) has demonstrated that additional pathways for hot-electron transfer can be created to expedite the equilibration process to sub-picoseconds. Some other limiting factors have to be considered in developing an ultrafast plasmonic modulator. First, intrinsic losses in plasmonic systems pose a significant challenge. Second, the ultra-small structure size limits the effective nonlinear medium volume,
resulting in small modulation depth and lowered damage threshold. Third, a tradeoff exists between the tuning speed and modulation depth ($\Delta \varepsilon$). To maximize $\Delta \varepsilon$, the hot electron temperature should be increased, resulting in a drop in energy transfer rate from electron gas to the lattice.$^{54,114}$

In recent years, epsilon near zero (ENZ) materials,$^{107,123}$ such as indium tin oxide (ITO)$^{66}$, aluminum-doped zinc oxide$^{72}$, indium-doped cadmium oxide$^{124}$ and so on, have emerged as a promising candidate for nonlinear modulation thanks to their unique properties, including enhanced nonlinear susceptibility $\chi^{(3)}$ and field intensities. Due to the vanishing of refractive index ($\text{Re}(n_0) \approx 0$), the normal definition of Kerr index$^{97}$

$$n_2 = \frac{3\chi^{(3)}}{4n_0\text{Re}(n_0)\varepsilon_0c}$$

is not suitable for describing the nonlinear index change because the assumption ($|n_2 I / n_0| \ll 1$) is violated when the linear index $n_0$ is small$^{125}$. The intensity dependent refractive index should be directly calculated from $\chi^{(3)}$.$^{123}$

$$n(I) = \sqrt{\varepsilon^{(1)} + 3\chi^{(3)}|E|^2}$$  \hspace{1cm} (2-6)

Simple ENZ thin films have shown enhanced nonlinear effect without the effort of nano-structuring. It is due to the fact that the boundary condition related to the continuity of $\hat{n} \cdot \vec{D}$ ensures that a strong normal (to the interface) component of the electric field is induced inside the ENZ materials. From a microscopic point of view, compared with metals ENZ materials have a far lower electron density and a concomitant smaller electron heat capacity $C_e$, which facilitates the increasing of hot-electron temperature and thus changing the electron scattering rate and the plasma frequency due to the non-parabolic conduction band.$^{124,126–128}$ Moreover, as indicated by the two-temperature model (2-5), the energy exchange rate between electrons and the lattice is proportional to the difference of the electron temperature increase which is a lot higher than that of the metal, therefore sub-picosecond relaxation times have been observed$^{66,126}$ compared with the few-picosecond kinetics observed in metals. However, bulk ENZ materials normally have ENZ frequencies in the near- and mid-infrared region, which imposes a spectra limitation in their applications. Although ENZ metamaterials can be engineered to shift the ENZ frequencies to other spectra
range, they inevitably use metallic materials especially for visible spectra range $^{129,130}$, which will sacrifice the sub-picosecond modulation speed.

Triggered by the observation of electric and magnetic multipolar Mie resonances in high refractive index dielectric nanoparticles $^{131}$, we have witnessed a shift to dynamic meta-devices based on dielectric nanostructures $^{132,133}$, because they show lower ohmic losses and higher damage threshold compared with plasmonic ones. In particular, silicon meta-devices have gained increasing attention due to its high refractive index, superior nonlinear properties, low loss in the telecom spectral range, CMOS compatibility and relevance to integrated photonics architectures. Optical third order nonlinear effects are especially important in silicon based devices as they enable various functions, including optical switching $^{70}$, nonlinear frequency conversions $^{90}$ and ultrafast optical modulation. According to equation (2-4), the change in the imaginary part of refractive index is determined by its TPA coefficient, which will decrease considerably as the wavelength goes beyond 2 μm, because the combined energy of two photons is less than the band-gap of silicon (1.1 eV) $^{134,135}$. Meanwhile, $n_2$ peaks around 1.8 to 1.9 μm, which shows favorable Kerr nonlinearities and maintains a low loss in the mid-infrared region $^{135,136}$. In the telecom wavelength range, silicon offers a large Kerr index, $n_2 \approx 4 \times 10^{-14}$ to $9 \times 10^{-14}$ cm$^2$ W$^{-1}$ $^{137,138}$ (versus $1 - 2 \times 10^{-15}$ cm$^2$ W$^{-1}$ for optical glasses $^{134}$), but is also accompanied by a large $\beta_2$ (5$\times$10$^{-10}$ to 9$\times$10$^{-10}$ cm W$^{-1}$ $^{139,140}$) $^{97}$. Amorphous silicon ($\alpha$-Si) provides a promising alternative to crystalline silicon ($c$-Si) thanks to its high refractive index (3.4 – 3.6), strong nonlinearities $^{141}$ and modest TPA as a result of larger band-gap (1.55 – 2.10 eV) $^{142,143}$. Strong field confinement inside the amorphous silicon resonator benefits from its high index, and prominent Mie-type resonances have been observed $^{90,144,145}$. Depending on specific growth conditions, the reported Kerr indices can be one order of magnitude larger than that of $c$-Si $^{141}$, with values as high as $7.43 \times 10^{-13}$ cm$^2$ W$^{-1}$ $^{146}$. Due to these remarkable linear and nonlinear properties of $\alpha$-Si, people have already demonstrated different integrated photonic devices with better performance than their $c$-Si counterparts, such as low loss waveguide $^{147}$, high gain parametric amplifier $^{148}$, chip-based supercontinuum generation $^{149}$, ultrafast all-optical switch $^{150}$, and so on. In addition, it can be deposited at low temperature
(200 – 400 °C) using plasma-enhanced chemical vapor deposition (PECVD) and patterned by the same technology as c-Si, making it suitable for multi-layer processing widely adopted in metasurfaces fabrication. Therefore, α-Si holds great promise for making time-variant optical metasurfaces based on the third order nonlinear processes.

2.2 Theory and Design Methodology

2.2.1 Laser-induced Dynamic Gratings

As we know, spatial grating can be created when the optical properties of matter become spatially modulated in the interference region of two coherent laser beams, which is a well-studied concept and has been widely employed in optical applications such as holography, spectroscopy, self-adaptive optical systems and telecommunications. When frequencies of the two laser beams are different, the interference produces a travelling-wave grating that can be detected and analyzed by the diffraction of a probing light beam. Without loss of generality, the electric field of the interacting laser beams can be expressed as

\[ E_j(\vec{r},t) = \left( \frac{A_j(\vec{r},t)}{2} \right) \exp \left( i\left( \vec{k}_j \cdot \vec{r} - \omega_j t \right) \right) + \text{c.c.}, \ j = 1, 2 \]  

(2-7)

where \( A \) is the wave amplitude (for a Gaussian pulse, \( A_j(\vec{r},t) = a_j f_j(t) = a_j \exp \left( -2\ln(2) \left( \frac{t}{\tau_j} \right)^2 \right) \)), \( \vec{k} \) its wavevector, and c.c. stands for the complex conjugate. The ultrafast laser pulse used in our experiment has a pulse duration of 140 fs. The intensity distribution of the total field can be represented by

\[ I(\vec{r}) = \frac{1}{Z} \left| \vec{E}_1(\vec{r},t) + \vec{E}_2(\vec{r},t) \right|^2 \]  

(2-8)
where $Z = \frac{1}{\varepsilon_0 cn}$ is the resistance of the material, and the bar denotes time averaging. Using equation (2-7) and (2-8), and averaging over times that are long compared with the optical periods $2\pi/\omega_j$ but short compared with the modulation period $2\pi/(\omega_1 - \omega_2)$ one obtains

$$I = \frac{n}{2} \varepsilon_0 c \left( a_1^2 + 2a_1 \cdot a_2 \cos \left( (\vec{k}_1 - \vec{k}_2) \cdot \vec{r} - (\omega_1 - \omega_2) t \right) + a_2^2 \right) f_1(t) f_2(t)$$

$$= \left( I_1 + I_2 + 2\Delta I \cos \left( \vec{k}_M \cdot \vec{r} - \Delta \omega t \right) \right) f_1(t) f_2(t)$$

(2-9)

The intensity $I$ exhibits a travelling-wave modulation with a grating vector

$$\vec{k}_M = \vec{k}_1 - \vec{k}_2$$

and a frequency

$$\Delta \omega = \omega_1 - \omega_2 \quad \omega_1 \geq \omega_2$$

The wave-like intensity modulation changes the optical properties, e.g. refractive index and absorption, of materials in the interference region, resulting in a moving index grating (Fig. 2-1)

$$\Delta n \propto (n_2 + i \frac{\lambda}{4\pi} \beta_2) \Delta I = \frac{n}{2} \varepsilon_0 c (n_2 + i \frac{\lambda}{4\pi} \beta_2) a_1 a_2 \cos \left( \vec{k}_M \cdot \vec{r} - \Delta \omega t \right)$$

(2-10)

The grating can be detected by a third laser beam whose diffraction angle and frequency will be modulated. Similar to the diffraction of static grating, the frequency shifts of the diffracted light from the moving grating are integer number of $\Omega$, and can be interpreted as a Doppler effect due to the motion of the grating. This dynamic refractive index modulation will in turn shift the optical response of the material, and thus laying the foundation for ultrafast optical modulation of metasurfaces.
Figure 2-1 A schematic of two pump beams with closely spaced frequencies $\omega_{p1}$ and $\omega_{p2}$ impinging on the metasurface with incident angles $\theta_1$ and $-\theta_2$, respectively. They produce travelling interference fringes with a period of $\Lambda_M$ and a speed of $v_M = \Delta \omega / k_M$ on the metasurface. The nanoantennas on the metasurface exhibit a time-variant change in the index induced by the travelling interference fringes.

### 2.2.2 Resonant Meta-atom Design – a Hybrid Approach

Hybrid metasurface made of noble metal and high-index dielectric components attract special attention as it represents a more flexible platform for the tuning of resonances. It combines the advantages of plasmonic and dielectric structures to achieve a higher damage threshold, lower loss, smaller device size and enhanced Kerr nonlinearity, etc. We used a hybrid structure consisting of $\alpha$-Si nanobar antenna / dielectric spacer / metal back plate to construct the meta-atom. With the incorporation of the dielectric spacer layer and metal back plate, multiple reflections within the spacer layer relaxes the required phase delay upon a single pass through the $\alpha$-Si resonator layer, mitigating the trade-off between phase control and efficiency present in single layer high contrast dielectric nanoantennas.
As an example, I will show the design process for the meta-atom used in Chapter 4. Fig. 2-2 a shows a schematic of a unit cell consisting of $\alpha$-Si nanobar antenna / Al$_2$O$_3$ spacer / Au back plate. In order to find the optimized parameters that can create the maximum $\Delta n$ and $\Delta \phi$ under intensity modulation, I used a commercially available finite element method (FEM) solver package – COMSOL Multiphysics. A periodic boundary condition was applied for a single building block to simplify the model and save computation memory and time. Third-order finite elements and at least 10 mesh steps per wavelength were used to ensure the accuracy of the calculated results. The experimentally obtained optical constants of gold, alumina and $\alpha$-Si were used to model the back reflector, spacer and the meta-atom. The nonlinear simulation takes two major steps in a customized model: (1) simulated the spatially dependent permittivity change using an iterative scheme – I first calculated the field distribution in the computational domain with the pump beam ($\lambda = 1550$ nm) incidence, then updated the permittivity in the Kerr medium ($n_2 = 5 \times 10^{-13}$ cm$^2$/W) with the calculated inhomogeneous field, and after that I calculated the field again with modified permittivity. I iterated over the steps above until the change of the field distribution was within a predefined tolerance. (2) Simulated the structure with the pump-induced permittivity changes with a probe light ($\lambda = 1550$nm) incidence. I obtained the reflected field distribution of a 1550nm probe beam upon incidence on the nanoantenna whose permittivity has been modified by the pump beam. By varying the size ($l_x$ and $l_y$) parameters, I mapped out the phase shift $\phi$ at different pump intensities (0 and 0.4 GW/cm$^2$), and calculated the phase shift change $\Delta \phi = \phi (I = 0.4 \text{ GW/cm}^2) - \phi (I = 0)$ (Fig. 2-2 b). The best design was found at $l_x = 600$ nm and $l_y = 650$ nm. Fig. 2-2 c displays the electric field distribution of a half of the unit cell in the $y$-$z$ plane at different pump intensities (from left to right: 0, 0.2 GW/cm$^2$, 0.4 GW/cm$^2$), where the shift of the wavefront indicates the change of phase shift $\phi$. It can also be noted that the structure supports strong magnetic dipole resonance at the wavelength 1550 nm, which is beneficial for creating large nonlinear refractive index change $\Delta n$ and $\Delta \phi$. Fig. 2-2 d shows that $\Delta \phi$ increases super-linearly with the pump intensity, leading to a strong modulation depth at a moderate light intensity.
Figure 2-2 (a) A 3D illustration of a unit cell consists of α-Si nanobar antenna. The thickness of the gold ground plate, the Al₂O₃ insulation layer and antenna are 100 nm, 50 nm and 350 nm, respectively. The square lattice periodicity is 1 µm, and the width/length of the antennas is 600 nm/650 nm. (b) Simulated dynamic phase shift change (Δφ) of the reflected optical field in a 2D parameter space spanned by \( l_x \) and \( l_y \). The white diamond marks the designed antenna that exhibits a large Δφ under the illumination of intense light (0.4 GW/cm²). (c) Simulated Δφ with modulation intensities. The inset displays electric field distribution at different modulation intensities, showing wavefront shifts induced by the nonlinear effect.

2.3 Optical Implementation

Spatiotemporal phase modulation can be created by the interference of two laser pulses with different frequencies. To generate two laser pulses with modified spectra from a single pulse, we used a technique called ultrafast pulse shaping through spectral filtering \(^{153,154}\). For a long laser pulse (beyond picosecond), pulse shaping has been done directly by elements of which the transmission is controlled externally, for example, a Pockels cell placed between crossed polarizers and driven by an electrical pulse. However, the speed limitation of electronics has prevented its application to femtosecond pulse shaping.
To achieve pulse shaping (specifically in our case frequency splitting), first the laser pulse is dispersed by a grating, then the spectrum is propagating through a mask which spatially filters the pulse. Finally, the spectral components are recollimated by a second grating (Fig. 2-3 a). The two-gating and two-lens configuration introduces zero group velocity dispersion. The mask can be designed to fulfill certain pulse shaping function. For example, a simple slit as mask acts as a spectral window. We used two spatially separated slits to filter out two spectral windows. In order to split the two spectra components, a small angle difference between the two slits was introduced, so that two pulses propagated along different paths. By adjusting the width of slit, the spectra range / pulse width can be tuned.

Fig. 2-3 b shows a folded version of the spectral filtering used in our experiment. The output of a Ti:Sapphire pulsed laser radiation (140-fs pulse width, 80-MHz repetition rate) was dispersed by a transmission grating, focused by a lens and then separated by a customized split mirror with a variable-size block attached in the center to tune the frequency separation. This split mirror works as a spatial mask that spectrally and spatially separates a single pulse into two. The separated beams were then reflected back through the lens and grating, and recombined into two pump beams with shifted central frequencies. In this way, a single laser pulse can be split into two pulses with different center frequencies. By adjusting the delay line, the temporal delay between the two pump beams can be tuned.
2.4 Conclusions

In this chapter, I have studied the theory of ultrafast optical modulation based on nonlinear Kerr effect, and different materials and structures suitable for creating large modulation depth. I described the full-wave simulation method for the design of metasurfaces and the optical implementation of travelling-wave modulation. Our method provides robust and controllable spatiotemporal modulation, of which $\Delta \omega$ and $k_M$ can be readily tuned as desired. It provides a powerful toolkit for generating dynamic modulations on various platforms, including metasurfaces and photonic integrated waveguides.
Chapter 3

Nonreciprocal Light Propagation with Spatiotemporal Phase Gradient Metasurfaces

Creating materials with time-variant properties is critical for breaking reciprocity that imposes fundamental limitations to wave propagation. However, it is challenging to realize efficient and ultrafast temporal modulation in a photonic system. In this chapter, leveraging both spatial and temporal phase manipulation offered by an ultrathin nonlinear metasurface, we experimentally demonstrated nonreciprocal light reflection at wavelengths around 860 nm. The metasurface, with traveling-wave modulation upon nonlinear Kerr building blocks, creates spatial phase gradient and multi-terahertz temporal phase wobbling, which leads to unidirectional photonic transitions in both momentum and energy spaces. We observed completely asymmetric reflections in forward and backward light propagations within a sub-wavelength interaction length of 150 nm. Our approach pointed out a potential means for creating miniaturized and integratable nonreciprocal optical components.

3.1 Background and Motivation

Reciprocity is a fundamental principle rooted in linear physical systems with time-reversal symmetry, requiring that the received-transmitted field ratios are the same when the source and detector are interchanged. However, it is preferable to break reciprocity in many practical applications, such as lasers and full-duplex communication systems, so that back-scattering from defects or boundaries can be avoided. Traditionally, nonreciprocity has been realized through magneto-optic materials which are too bulky and lossy to be integrated into modern photonic systems. In addition, nonlinear materials are employed to achieve nonreciprocity at a cost of high intensity requirement, but they suffer from poor isolation and are reciprocal to noises. In order to circumvent these limitations, more and more researches have focused on developing materials with time-variant properties in which time-reversal symmetry is
explicitly broken to achieve nonreciprocity \cite{32,162}. So far, based on strong electro-optic \cite{163,164}, acousto-optic \cite{81,165,166}, or optomechanical effects \cite{167,168} of different materials, proof of concept temporal modulation has been demonstrated at frequencies from kHz to GHz range, which is much lower than the optical frequency as a result of slow carrier injection of electro-optic modulation and low-frequency acoustic or mechanical modes in acousto-optic or optomechanical modulation. In addition, these dynamic systems suffer from limited bandwidth either due to the group velocity mismatch among photonic modes or the intrinsic narrow linewidths of acoustic and mechanical modes. Moreover, they require long interaction lengths to observe the desired effect. Nonreciprocity with a sub-wavelength interaction length and an ultrafast modulation frequency over THz bandwidth is technically challenging and has not been realized to date.

3.2 Theory

3.2.1 Lorentz Reciprocity Theorem

In electromagnetics, a reciprocal / nonreciprocal system is defined as a system that exhibits same / different received-transmitted field ratios when its source and detector are exchanged \cite{32}. Using the method presented in ref. \cite{169,170}, we started with the time-harmonic sourceless Maxwell’s equation and derived the Lorentz reciprocity theorem. We start with one solution denoted by prime, which satisfy equations

\[ \nabla \times E' = -j \omega \mu H' \] (3-1)

\[ \nabla \times H' = j \omega \varepsilon E' \] (3-2)

We set up another solution double primes in the same manner. Multiplying equation (3-1) with \(H''\) and equation (3-2) with \(E''\) and then summing them together gives

\[ H' \nabla \times E' + E' \nabla \times H' = j \omega (\varepsilon E' E' - H' \mu H') \] (3-3)

Applying the same operations with interchanged rimes yields

\[ H' \nabla \times E' + E' \nabla \times H' = j \omega (\varepsilon E' E' - H' \mu H') \] (3-4)
Subtraction equation (3-3) and (3-4) we obtain

\[ \nabla \cdot (E' \times H' - E' \times H') = j \omega (E' \varepsilon E' - E' \varepsilon E' - H' \mu H' + H' \mu H') \]  

(3-5)

If \( \varepsilon \) and \( \mu \) are scalars or symmetric tensors, the right-hand side of equation (3-5) adds up to zero, yielding the Lorentz reciprocity theorem

\[ \nabla \cdot (E' \times H' - E' \times H') = 0 \]  

(3-6)

It holds for materials with gain or loss as long as \( \varepsilon \) and \( \mu \) are symmetric.

It should be noted that reciprocity is closely related to time-reversal symmetry. Time-reversal symmetry is a fundamental principle which describes the invariance of physical process under the operation of the time reversal \( T : t \rightarrow -t \). According to ref. 32, upon time reversal operation, electric field related variables \( E, D \) and \( P \) are even (\( T\{E(\omega)\} = E^*(\omega) \)), whereas magnetic field related variables \( H, B, M \) are odd (\( T\{H(\omega)\} = -H^*(\omega) \)), and noting that \( \nabla \) operator is time reversal invariant. These properties make Maxwell’s equation (3-1) and (3-2) time reversal invariant, indicating that the time-reversal system will have the same electromagnetic solutions as the direct system. And using the same mathematical operations, we obtain

\[ \nabla \cdot (E \times H^* - E^* \times H) = j \omega (E^* D - ED^* - H^* B + HB^*) \]  

(3-7)

For a bianisotropic medium, its constitutive parameters are

\[ D = \varepsilon(F) \cdot E + \zeta(F) \cdot H \]  

(3-8)

\[ B = \zeta(F) \cdot E + \mu(F) \cdot H \]

\( \varepsilon \) is permittivity tensor, \( \mu \) is permeability tensor, \( \zeta \) is magnetic-to-electric coupling tensor and \( \xi \) is electric-to-magnetic coupling tensor. And \( F \) represents the external bias. The same for the time-reversed medium:

\[ D' = \varepsilon(-F) \cdot E' + \zeta(-F) \cdot H' \]  

(3-9)

\[ B' = \zeta(-F) \cdot E' + \mu(-F) \cdot H' \]

Applying time-reversed operations on (3-8):
\[ D^* = \bar{\varepsilon}(-F) \cdot E^* + \bar{\varepsilon}(-F) \cdot (-H^*) \]
\[ B^* = \bar{\varepsilon}(-F) \cdot E^* + \bar{\mu}(-F) \cdot (-H^*) \]  
(3-10)

Complex conjugating (3-10),

\[ D = \bar{\varepsilon}^*(-F) \cdot E - \bar{\varepsilon}^*(-F) \cdot H \]
\[ B = -\bar{\varepsilon}^*(-F) \cdot E + \bar{\mu}^*(-F) \cdot H \]  
(3-11)

Comparing equation (3-11) with (3-8), we find

\[ \bar{\varepsilon}^*(-F) = \bar{\varepsilon}(F) \]
\[ \bar{\mu}^*(-F) = \bar{\mu}(F) \]
\[ \bar{\varepsilon}^*(-F) = -\bar{\varepsilon}(F) \]
\[ \bar{\mu}^*(-F) = -\bar{\mu}(F) \]

(3-12)

Complex conjugating (3-12) and applying the dummy transformation \( F \rightarrow -F \),

\[ \bar{\varepsilon}(F) = \bar{\varepsilon}^*(-F) \]
\[ \bar{\mu}(F) = \bar{\mu}^*(-F) \]
\[ \bar{\varepsilon}(F) = -\bar{\varepsilon}^*(-F) \]
\[ \bar{\mu}(F) = -\bar{\mu}^*(-F) \]

(3-13)

Assuming lossless system, then equation (3-13) becomes

\[ \bar{\varepsilon}(F) = \bar{\varepsilon}(-F) \]
\[ \bar{\mu}(F) = \bar{\mu}(-F) \]
\[ \bar{\varepsilon}(F) = -\bar{\varepsilon}(-F) \]
\[ \bar{\mu}(F) = -\bar{\mu}(-F) \]

(3-14)

Substituting equation (3-14) to (3-10), we obtain the time-reversed counterparts of \( D \) and \( B \):

\[ D^* = \bar{\varepsilon}(-F) \cdot E^* + \bar{\varepsilon}(-F) \cdot H^* \]
\[ B^* = \bar{\varepsilon}(-F) \cdot E^* + \bar{\mu}(-F) \cdot H^* \]

(3-15)

Substituting (3-8) and (3-15) into (3-7), we obtain
\[
\n\n\n\]

Applying the transposed identity \( a \cdot \mathcal{F} b = (a \cdot \mathcal{F} b) \) to terms on the right-side of (3-16):

\[
\n\n\n\]

Due to reciprocity, the right-side of equation (3-17) vanishes, therefore, the following relations must hold for arbitrary fields:

\[
\begin{align*}
\bar{\varepsilon}(F) &= \bar{\varepsilon}_r(-F) \\
\bar{\mu}(F) &= \bar{\mu}_r(-F) \\
\bar{\xi}(F) &= -\bar{\xi}_r(-F) \\
\bar{\zeta}(F) &= -\bar{\zeta}_r(-F)
\end{align*}
\]

(3-18)

Noting that transposed dyadics in equation (3-18) correspond to propagation in the reverse direction to that of the wave associated with the un-transposed dyadics. So that time-reversal symmetry criterion for reciprocity is with reversed sign of \( F \), wave propagating in reversed directions see the same effective media.

Taking the transmission through a linear and lossless optical media as an example, the time-reversed process can be viewed as flipping the direction of propagation of light. With the media remain unchanged and no external force \( F = 0 \), this back-propagation retraces the path in the present time frame, and the system is deemed as time-reversal symmetric and reciprocal.

The consequences of Lorentz reciprocity are easier to grasp using a concrete example. Considering a plane wave impinging on, reflection from and transmission through a slab of optical media. Defining incident wave amplitudes by \( a \) and reflected wave amplitudes by \( b \), the tangential components of the wave have the form
\[ E_{y,1}(x, y, z) = (a_i e^{-ikz} + b_i e^{ikz}) e_{y,1}(x, y) \]  \tag{3-19}

\[ H_{x,1}(x, y, z) = (a_i e^{-ikz} - b_i e^{ikz}) h_{x,1}(x, y) \]  \tag{3-20}

Noticing that \(a_1\) and \(a_2\) can be chosen as independent variables, \(b_1\) and \(b_2\) as dependent variables, the latter being linear functions of the former. The system can be described by the scattering matrix

\[
\begin{pmatrix}
    b_1 \\
    b_2 \\
\end{pmatrix} =
\begin{pmatrix}
    S_{11} & S_{12} \\
    S_{21} & S_{22} \\
\end{pmatrix}
\begin{pmatrix}
    a_1 \\
    a_2 \\
\end{pmatrix}
\]

\[ B = SA \]  \tag{3-21}

Here, \(A = (a_1, a_2)^T\) and \(B = (b_1, b_2)^T\). Diagonal elements are the reflection coefficients back into the respective port, while off-diagonal elements are transmission coefficients from one port to the other.

Figure 3-1 TE wave incident (not necessary to be normal) on a slab. It represents a two-port system.

Recalling equation (3-5) in integration form and using equation (3-19) and (3-20), we obtain

\[
\int_E E_x^s \times H_y^s \cdot ds \rightarrow -\left(A_T^s + B_T^s\right)(A^s - B^s)
\]

\[
\int_E E_x^s \times H_y^s \cdot ds \rightarrow -\left(A_T^s + B_T^s\right)(A^s - B^s)
\]  \tag{3-22}

The reciprocity theorem gives

\[ B_T^s A^s = A_T^s B^s \]  \tag{3-23}

where we have used the fact that \(B_T A = A_T B\) by definition of the transpose of a column vector and canceled terms on both sides of the equation. Introducing equation (3-21) and noting that
By definition of transpose, we obtain for (3-23)

$$A_T S_T A^T = A_T S A^T$$  \hspace{1cm} (3-25)

Because \( A' \) and \( A'' \) are arbitrary variables, (3-13) requires that

$$S_T = S$$  \hspace{1cm} (3-26)

The scattering matrix of a linear reciprocal system is symmetric, and in the two-port system

$$S_{12} = S_{21}$$  \hspace{1cm} (3-27)

Under certain conditions, Lorentz reciprocity doesn’t hold. The first and most commonly used is magneto-optical materials whose permittivity is an asymmetric tensor, which violates equation (3-18). Then the order by which \( E', \varepsilon \) and \( E'' \) are multiplied is nontrivial, and the right-hand side of equation (3-5) becomes nonzero. Another situation is nonlinear materials, \( \varepsilon \) is a function of the electric field strength and the right-hand side of equation (3-5) becomes \( E' \varepsilon (E') E' - E' \varepsilon (E') E' \), which is non-zero for different fields. Because nonreciprocity is equivalent to time-reversal symmetry breaking in lossless system, when the material properties are time dependent (doesn’t satisfy equation (3-18)), the system becomes nonreciprocal.

### 3.2.2 Breaking Reciprocity with Time Modulation

A conventional spatially modulated metasurface with a phase gradient (e.g. \( \phi = k_x x \)) on the surface is capable of imparting additional linear or orbital angular momentum to incident light. This breaks the inversion symmetry and enables full control over the photonic transitions in momentum space. However, this process is linear and time-reversal symmetric, and is inherently reciprocal (Fig. 3-2, b and c). In contrast, our space-time modulated metasurface has a permittivity modulation of the form

$$\varepsilon(x,t) = \Delta \varepsilon \cos(\Delta \omega t - k_M x)$$  \hspace{1cm} (3-28)
which leads to a phase modulation:

\[ \varphi(x, t) = \Delta \varphi \cos(\Delta \omega t - k_M x) + k_s x \]  

(3-29)

where \( \Delta \varphi \) is the temporal modulation depth, \( \Delta \omega \) is the modulation frequency, \( k_M \) is the modulation spatial frequency, and \( k_s \) is the static phase gradient introduced by the spatial distribution of nanoantennas. This modulation acting upon the incident light field gives an additional space-time varying phase factor expressed as \( \exp[i \varphi(x, t)] \). Applying the Jacobi-Anger expansion, this phase term can be rewritten as a series of Bessel functions of the first kind, which enables the reflected field to be expressed as

\[
\tilde{E}_r (\vec{r}, t) = \zeta J_0 (\Delta \varphi) \tilde{E}_i e^{i(\vec{k} \cdot \vec{r} + k_s x - \omega t)} \\
+ i \zeta J_1 (\Delta \varphi) \tilde{E}_i \left\{ e^{i\left( \vec{k} \cdot \vec{r} + k_M x + k_s x \right) - (\omega + \Delta \omega) t} \\
+ e^{i\left( \vec{k} \cdot \vec{r} - k_M x - k_s x \right) - (\omega - \Delta \omega) t} \right\} 
\]  

(3-30)

where \( \omega_i \) and \( \vec{k}_i \) are the frequency and free-space wavevector of the incident wave, \( \zeta = \sqrt{\eta} \), and \( \eta \) is the static diffraction efficiency of the metasurface. Note that only the zeroth- and first-order Bessel functions are retained since the phase modulation depth \( \Delta \varphi \) is small, which leads to negligible contributions from higher-order functions. It is evident from the second term in the right-hand side of equation (3-30) that a sinusoidal phase component will be decomposed into two photonic transitions (i.e. sidebands) in energy space with resulting frequencies \( \omega_r = \omega_i \pm \Delta \omega \), where `+' and `–' denotes an upward and downward transition, respectively. The dynamic phase modulation breaks reciprocity, and leads to time-reversal-asymmetric photonic transitions \(^{171}\). Different from the recently reported phonon-mediated nonreciprocal waveguides \(^{84,166}\) based on indirect interband photonic transitions \(^{171}\), our system is naturally phase matched for all free space modes and does not require complex design of the acoustic and photonic modes to fulfil stringent momentum/energy matching conditions. Furthermore, our space-time metasurfaces exhibit agile control over both momentum and energy conversions. In order to achieve unidirectional frequency conversion, the metasurface can be designed to either fulfil \( k_{ix} + k_s + k_M > k_i \) (\( k_{ix} \) is the projection of incident wavevector \( k_i \) along the x direction), \( 2k_M > k_s \), and \( -k_i < k_{ix} + k_s - k_M < k_i \) to ensure unidirectional down-
conversions (Fig. 3-2 d and e), or fulfill \( k_{ix} - k_s - k_M < -k_i \), \( 2k_M > k_i \) and \( -k_i < k_{ix} - k_s + k_M < k_i \) to ensure unidirectional up-conversions. As an example, we depict the case of unidirectional down-conversions in Fig. 3-2 d and e. The optical paths of forward and backward propagation are shown schematically in the top panel, and the photonic states represented by different color dots are shown in the bottom dispersion diagrams. The photonic transition from the blue dot state to green occurs in the forward propagation, whereas green to red occurs in the backward propagation. With a space-time modulated metasurface, the frequency transitions arise from the parametric processes caused by the temporal modulation, while the momentum transitions arise from both temporal and spatial modulation. As a result, the allowed transitions (i.e. downward transition) can be selected by pushing a given output state (i.e. upward transition) to the forbidden (i.e. non-propagative) region with a unidirectional momentum transfer, \( k_s \), provided by the spatial phase modulation of the metasurface. It is worthwhile to note that the reflection angle of the backward propagating light is not necessarily the same as that of the forward propagating light even though they have the same \( x \) component of the wavevector \( (k_s) \), because the frequency shift also changes the length of the wavevector. The paths of the incident and returning beam overlap only in the special case of normal incidence (i.e. \( k_{ix} = 0 \)), which is particularly useful for free-space optical isolators.
Figure 3-2 **Working principle of a nonreciprocal space-time phase modulated metasurface.** (a) An illustration showing the concept of a space-time phase modulated metasurface consisting of resonating dielectric nanoantennas operating in the reflection mode. A travelling phase modulation in a sinusoidal form is superposed on the designed phase gradient along the x direction. Light impinging on the metasurface with frequency $\omega$ is converted to a reflecting beam with frequency $\omega - \Delta \omega$ due to the parametric process arisen from dynamic phase modulation, while the back-propagating beam with frequency $\omega - \Delta \omega$ will be converted to $\omega - 2\Delta \omega$ instead of $\omega$, resulting in a nonreciprocal effect. (b), (c) and (d), (e). Comparison between a regular space modulated metasurface (b) and (c) and a space-time phase modulated metasurface (d) and (e). A regular space modulated metasurface only supports symmetric forward (b) and backward (c) reflections as shown in the dispersion diagrams. There is no frequency conversion (i.e. no transition in energy space). The process is reciprocal and the forward and backward beams share the same trajectory. In contrast, a space-time phase modulated metasurface supports asymmetric forward (b) and backward (c) reflections. It not only offers additional momentum along x direction to the reflected light but also converts its frequency. In either forward or backward case, the upward transition is forbidden because the resulting wavevector will be too large to be supported in free space, resulting in unidirectional photonic transitions in both energy and momentum spaces. Therefore, the trajectories of beams differ and reveal nonreciprocity effect.

$k_s$ is the linear momentum introduced by the spatial phase modulation and $k_M$ is the additional linear momentum introduced by the temporal phase modulation.

3.2.3 **Scattering Matrix Analysis**

We investigated the scattering matrix of our space-time phase modulated metasurface. As shown in Fig. 3-3, we considered our metasurface as a four-port system, where $s_1^+$ is the amplitude of the incident wave ($\omega_i, k_{ix}$), $s_2^-$ is the amplitude of the static diffraction ($\omega_i, k_{ix} + k_s$), $s_3^-$ is the amplitude of the down-conversion ($\omega_i - \Delta \omega, k_{ix} + k_s - k_M$) and $s_4^-$ is the amplitude of the up-conversion ($\omega_i + \Delta \omega, k_{ix} + k_s + k_M$).

Table 1 summarizes the frequency and wavevector of each port.

<table>
<thead>
<tr>
<th>Port number</th>
<th>Input $s_+$</th>
<th>Output $s_-$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>$\omega_i, k_{ix}, k_{iz}$</td>
<td>$\omega_i, -k_{ix}, -k_{iz}$</td>
</tr>
<tr>
<td>2</td>
<td>$\omega_i, k_{ix} + k_s, k_{2z}$</td>
<td>$\omega_i, -(k_{ix} + k_s), -k_{2z}$</td>
</tr>
<tr>
<td>3</td>
<td>$\omega_i - \Delta \omega, k_{ix} + k_s - k_M, k_{3z}$</td>
<td>$\omega_i - \Delta \omega, -(k_{ix} + k_s - k_M), -k_{3z}$</td>
</tr>
<tr>
<td>4</td>
<td>$\omega_i + \Delta \omega, k_{ix} + k_s + k_M, k_{4z}$</td>
<td>$\omega_i + \Delta \omega, -(k_{ix} + k_s + k_M), -k_{4z}$</td>
</tr>
</tbody>
</table>

Table 3-1 Frequencies and wavevectors of the waves related to the four ports.
Figure 3-3 (a) Illustration of a four-port system representing the metasurface, where port 1 is the incident port, port 2 relates to the static diffraction of the metasurface, and port 3 (4) couples to the down (up) converted mode. (b) A dispersion diagram depicting the energy/momentum relationships between modes \( a_0, a_-, \) and \( a_+ \). Mode \( a_0, a_- \) and \( a_+ \) will couple out to \( s_2, s_3, \) and \( s_4 \) due to the static phase gradient \( k_s \) of the metasurface.

High-order modes are not considered here since they are very weak with small phase modulation depth. We also omitted the specular reflection because the metasurface diffraction efficiency is close to unity around the designed wavelength.

According to equation (3-30), we can relate the \( s_i \) to \( s_{i+} \) by using the following equations:

\[
\begin{align*}
    s_{1-} &= \zeta J_0(\Delta \phi)e^{i k_s x} s_{2+} \\
    s_{2-} &= \zeta J_0(\Delta \phi)e^{i k_s x} s_{1+} \\
    s_{3-} &= i\zeta J_1(\Delta \phi)e^{i \Delta \omega} e^{-i k_M x} e^{i k_s x} s_{1+} \\
    s_{4-} &= i\zeta J_1(\Delta \phi)e^{-i \Delta \omega} e^{i k_M x} e^{i k_s x} s_{1+}
\end{align*}
\]

Therefore, the scattering matrix of the four-port system can be written as:

\[
S = \begin{pmatrix}
0 & \zeta J_0(\Delta \phi) & 0 & 0 \\
\zeta J_0(\Delta \phi) & 0 & 0 & 0 \\
i\zeta J_1(\Delta \phi)e^{i \Delta \omega} e^{-i k_M x} & 0 & 0 & 0 \\
i\zeta J_1(\Delta \phi)e^{-i \Delta \omega} e^{i k_M x} & 0 & 0 & 0
\end{pmatrix}
\]

The common factor \( e^{i k_s x} \) is omitted since it does not influence the form of the scattering matrix. It is evident that the scattering matrix is asymmetric (\( S_{13} \neq S_{31} \) and \( S_{14} \neq S_{41} \)) due to the asymmetric photonic transitions arisen from spatiotemporal phase modulation. Thus, the system breaks reciprocity. \( S_{12} (S_{21}) \) represents the
insertion loss to the nonreciprocal system. Under ideal condition, it could be reduced to zero when \( J_0(\Delta \varphi) = 0 \) (e.g. \( \Delta \varphi = 2.405 \) radians).

In order to validate the scattering matrix obtained directly from field calculation, we also studied the dynamic system based on the coupled mode theory. Considering the same four-port system shown in Fig. 3-3 b, the incident wave through port 1 will directly excite the resonant mode \( a_0 \):

\[
a_0(t) = \tilde{a}_0 e^{-i\omega_0 t}
\]

(3-33)

The dynamic phase modulation with form \( \Delta \varphi \cos(\Delta \omega t - k_M x) \) can be translated into effective permittivity modulation of the metasurface, which can be expressed as \( \Delta \varepsilon(x, t) = \Delta \varepsilon_{\text{eff}} \cos(\Delta \omega t - k_M x) \). The permittivity modulation induces the coupling among modes \( a_0, a_- \), and \( a_+ \). Therefore, we can obtain a set of master equations for the dynamics of the three excited modes as shown in Fig. 3-3 b:

\[
\begin{align*}
\dot{a}_0 &= (-i\omega_0 - \gamma_0)a_0 + i\kappa e^{-i\Delta \omega} e^{ik_M x} a_- + i\kappa e^{i\Delta \omega} e^{-ik_M x} a_+ + c_1s_{1+} \\
\dot{a}_- &= -i(\omega_0 - \Delta \omega - \gamma_-)a_- + i\kappa e^{i\Delta \omega} e^{-ik_M x} a_0 \\
\dot{a}_+ &= -i(\omega_0 + \Delta \omega - \gamma_+)a_+ + i\kappa e^{-i\Delta \omega} e^{ik_M x} a_0
\end{align*}
\]

(3-34)

where \( \gamma \) represents the decay rate of the modes, \( c_i \) is the coupling coefficient between the port \( i \) and the mode, \( \kappa \) represents the coupling strength between the modes induced by the permittivity modulation. The outgoing waves are expressed as:

\[
\begin{align*}
s_{1-} &= S_{12}s_{2+} \\
s_{2-} &= c_2 a_0 \\
s_{3-} &= c_3 a_- \\
s_{4-} &= c_4 a_+
\end{align*}
\]

(3-35)

where port 1 and port 2 are reciprocally connected because they are linked by the static phase gradient of metasurface which conserves reciprocity. Thus, \( S_{12} = S_{21} \). We consider that an incident wave \( (s_{1+}(t) = \tilde{s}_{1+} e^{-i\omega t}) \) is excited at frequency \( \omega \). Therefore \( a_0(t) = \tilde{a}_0 e^{-i\omega t} \), \( a_-(t) = \tilde{a}_- e^{-i\omega t} \), and
\[ a_+ (t) = \tilde{a}_+ e^{-i\omega t}, \] where \( \tilde{a}_0, \tilde{a}_-, \) and \( \tilde{a}_+ \) are the modal amplitudes for mode \( a_0, a_-, \) and \( a_+ \), respectively. By substituting \( A_0 (t) = a_0 (t), A_- (t) = a_- (t) e^{-i\Delta\omega t}, \) and \( A_+ (t) = a_+ (t) e^{i\Delta\omega t} \), we obtain the following solutions:

\[
A_0 (t) = \frac{c_1 (\gamma_+ + i(\omega_0 - \omega))}{D} s_{1+} (t) \\
A_- (t) = \frac{i \kappa e^{-i\omega_0 x} c_1 (\gamma_+ + i(\omega_0 - \omega))}{D} s_{1+} (t) \\
A_+ (t) = \frac{i \kappa e^{i\omega_0 x} c_1 (\gamma_+ + i(\omega_0 - \omega))}{D} s_{1+} (t)
\] (3-36)

where

\[
D = \left( \gamma_0 + i(\omega_0 - \omega) \right) \left( \gamma_- + i(\omega_0 - \omega) \right) \left( \gamma_+ + i(\omega_0 - \omega) \right) + \kappa^2 \left( \gamma_- + i(\omega_0 - \omega) \right) + \kappa^2 \left( \gamma_+ + i(\omega_0 - \omega) \right)
\]

Therefore, the scattering matrix of the system can be expressed as:

\[
S = \begin{pmatrix}
0 & \frac{c c_2 (\gamma_+ + i(\omega_0 - \omega))}{D} \\
\frac{c c_2 (\gamma_- + i(\omega_0 - \omega))}{D} & \frac{c c_2 (\gamma_+ + i(\omega_0 - \omega))}{D} \\
\frac{c \kappa e^{-i\omega_0 x} c c_3 (\gamma_+ + i(\omega_0 - \omega))}{D} & 0 \\
\frac{c \kappa e^{-i\omega_0 x} c c_3 (\gamma_- + i(\omega_0 - \omega))}{D} & 0 \\
\frac{c \kappa e^{-i\omega_0 x} c c_4 (\gamma_- + i(\omega_0 - \omega))}{D} & 0 \\
\frac{c \kappa e^{-i\omega_0 x} c c_4 (\gamma_+ + i(\omega_0 - \omega))}{D} & 0
\end{pmatrix}
\] (3-37)

Because \( \Delta\omega \approx 2.8 \text{ THz} \times 2\pi \) is much smaller than \( \omega_0 \) and the bandwidth of the metasurface resonance is much larger than \( \Delta\omega \), we can assume \( \gamma_0 = \gamma_- = \gamma_+ = \gamma \). In addition, we can also assume \( c_1 = c_2 = c_3 = c_4 = c \) because in a lossless system the coupling strength \( c \) between a port and a resonant mode is linked with the mode’s radiative decay rate \( c = \sqrt{2\gamma} \). Therefore, when the \( \omega = \omega_0 \), the scattering matrix can be further simplified as
\[
S = \begin{pmatrix}
0 & \frac{c^2 \left( \gamma^2 + \Delta \omega^2 \right)}{D} & 0 & 0 \\
\frac{c^2 \left( \gamma^2 + \Delta \omega^2 \right)}{D} & 0 & 0 & 0 \\
\frac{i \kappa c^2 \gamma e^{i \Delta \omega t} e^{-i k_{x} x}}{D} & 0 & 0 & 0 \\
\frac{i \kappa c^2 \gamma e^{-i \Delta \omega t} e^{i k_{x} x}}{D} & 0 & 0 & 0 \\
\end{pmatrix}
\] (3-38)

where \( D = \gamma \left( \gamma^2 + \Delta \omega^2 \right) + 2 \kappa^2 \gamma \).

By comparing equation (3-38) with equation (3-32), we found that the scattering matrices derived from two
difference methods are of the same form, in which \( S_{12} = S_{21}, |S_{31}| = |S_{41}|, S_{13} = S_{14} = 0 \), as well as \( S_{13} \neq S_{31} \) and
\( S_{14} \neq S_{41} \). Therefore, both derivations reach the same conclusion – the dynamically phase-modulated
metasurface possesses an asymmetric scattering matrix and is nonreciprocal.

### 3.2.4 Finite-difference time-domain (FDTD) Simulations of the Dynamic Phase Modulation Induced
Nonreciprocal Photonic Conversions

To verify the theoretical predictions of equation (3-30), we performed FDTD simulations of the
dynamically phase modulated metasurface which is simplified as a dipole array. A phase modulation of the
form \( k_{x} x_{n} + \Delta \phi \cos(\Delta \omega t - k_{x} x_{n}) \) was assigned to each dipole, where \( x_{n} \) is the \( x \) coordinate of the dipole
\( n \). This simplification provides insight into the nonreciprocal photonic transitions from space-time phase
modulation and allows us to simulate a large area of the metasurface with a reasonable amount of
computational resources.

In the first set of simulations, we simulated the case where both up and down-conversions are
allowed. The model consists of three supercells of metasurfaces with a period of 1200 nm (\( k_{s} = 0.72 k_{\text{probe}} \),
\( \lambda_{\text{probe}} = 860 \) nm), and three nanoantennas (represented by three dipoles) spanning a \( 2\pi \) phase shift range are
included in each supercell. As shown in Fig. 3-4 a, when we sent in a normally incident plane wave at \( \lambda = \)
860 nm, in consistency with the conversions shown in the dispersion diagram, the reflected plane waves are composed of three frequency/wavevector components: (1) The static diffraction is deflected to an angle of \( \theta = \sin^{-1} \left( \frac{k_s}{k_{\text{probe}}} \right) = 45.78^\circ \), (2) the up-conversion \((\lambda = 853 \text{ nm})\) is deflected to an angle of \( \theta = \sin^{-1} \left( \frac{(k_s - k_M)}{k_{\text{probe}}} \right) = 72.85^\circ \), and (3) the down-conversion signal \((\lambda = 867 \text{ nm})\) is deflected to an angle of \( \theta = \sin^{-1} \left( \frac{(k_s + k_M)}{k_{\text{probe}}} \right) = 28.54^\circ \).

In the backward propagation scenario, we sent back the two converted signals from the forward propagation case. Here, as we had two input fields, the field distribution of the outgoing waves would be a superposition of multiple plane waves. In order to clearly visualize the outgoing waves, we separated the converted waves arisen from the two backward inputs and presented their electric field distributions at modulated wavelengths in Fig. 3-4 b and c, respectively. The backward propagation of the down-converted signal \((\lambda = 867 \text{ nm})\) results in an upward converted wave at \(\lambda = 860 \text{ nm}\) with a reflection angle of \( \theta = \sin^{-1} \left( -\frac{(k_s - k_M) + k_M + k_s}{k_{\text{probe}}} \right) = -28.54^\circ \) and a downward converted wave of \(\lambda = 874 \text{ nm}\) with a reflection angle of \( \theta = \sin^{-1} \left( -\frac{(k_s - k_M) - k_M + k_s}{k_{\text{probe}}} \right) = 0^\circ \) (Fig. 3-4 b). The backward propagation of the up-converted signal \((\lambda = 853 \text{ nm})\) results in a down-conversion at \(\lambda = 860 \text{ nm}\) with a reflection angle of \( \theta = \sin^{-1} \left( -\frac{(k_s + k_M) - k_M + k_s}{k_{\text{probe}}} \right) = 28.54^\circ \) and an up-conversion of \(\lambda = 866 \text{ nm}\) with a reflection angle of \( \theta = \sin^{-1} \left( -\frac{(k_s + k_M) + k_M + k_s}{k_{\text{probe}}} \right) = 0^\circ \) (Fig. 3-4 c). None of the back-reflections return to the state of the original forward incidence, which proves that the spatiotemporal phase modulated metasurface is nonreciprocal.
Figure 3-4 FDTD simulation of the space-time phase modulated metasurface with both upward and downward photonic transitions using a simplified dipole array. Here we used $k_s = 0.72k_{probe}, k_M = 1/3 k_s = 0.24k_{probe}, \Delta \omega = 2.8$ THz, and $\Delta \phi = 0.2$ radians to allow both up and down-conversions. (a) The $y$ component of the electric field ($E_y$) distributions of the three outgoing waves from a normal incident plane wave at $\lambda = 860$ nm. From top to bottom, the field plots show the first-order up-conversion, the static diffraction, and the first-order down-conversion, respectively. The $k_x$-$\omega$ dispersion diagram shows the conversions which fulfil the momentum and energy conservations. (b) The incident wave (indicated by the black arrow) is switched to a plane-wave that is the backward of the down-conversion from (a). $E_y$ of the outgoing waves arisen from the first-order conversions and the static diffraction are shown and their frequencies (i.e. energy) and $k_x$ (i.e. momentum) agree with the transitions shown in the $k_x$-$\omega$ dispersion diagram. None of the outgoing waves follow the same path of the normal incidence of (a). (c) In the backward reflection of the up-conversion from (a) (indicated by the black arrow), three outgoing waves indicated by the $k_x$-$\omega$ dispersion diagram are shown to have different frequencies or reflection angles from the normal incidence in (a). In all $E_y$ distribution plots, the field strength of the static diffraction one order of magnitude stronger than the converted signals. This is consistent with the relation shown in equation (3-30) in the main text, where the static diffraction efficiency is about $|J_0(0.2)|^2 = 0.98$ and the first order conversion efficiency is about $|J_1(0.2)|^2 = 0.0099$. 
In the second set of simulations, we studied the case where only one direction of transition is allowed. Fig. 3-5 shows the simulation results of dipoles array with \( k_s = 0.78k_{\text{probe}} \), \( k_M = 2/3 \) \( k_s = 0.52k_{\text{probe}} \), \( \Delta \omega = 2.8 \text{ THz} \), and \( \Delta \phi = 0.2 \) radians. In the forward propagation, the normal incidence (\( \lambda = 860 \text{ nm} \)) is converted to two outgoing waves: the static diffraction and the first-order down-conversion. The up-conversion corresponds to a non-propagating mode that cannot radiate into free space. As shown in the field plot, this evanescent mode cannot carry energy away from the metasurface and no propagating field can be observed in free space. In the backward case, the incident wave is the backward of the down-conversion (Fig. 3-5 a). Only the down-conversion at \( \lambda = 874 \text{ nm} \) can be observed at the normal direction. Similarly, the up-conversion results in a non-propagating mode that cannot carry energy away.
Figure 3-5 FDTD simulation of the space-time phase modulated metasurface with only downward photonic transitions using a simplified dipole array. Here we used $k_s = 0.78k_{probe}$ and $k_M = 2/3 k_s = 0.52k_{probe}$ to allow only unidirectional down-conversions. (a) With a phase modulation depth $\Delta \varphi = 0.2$ radians, the forward normal incidence at $\lambda = 860$ nm is diffracted to a static diffraction and a first-order down-conversion. $E_y$ plot of the first-order up-conversion shows no propagating wave existing in free space, which is also confirmed by the dispersion diagram and demonstrated in the experiment. (b) The incident wave is switched to the backward of the down-conversion shown in (a). In response to this incidence, a down-conversion at $\lambda = 874$ nm reflects at the normal angle, while no propagating mode exists at $\lambda = 860$ nm. None of the outgoing waves overlaps with the forward incidence, and the process is nonreciprocal.
We also simulated the ideal case where the static diffraction vanishes under $J_0(\Delta \varphi) = 0$ ($\Delta \varphi = 2.405$ radians). In this case, the electric field strength of the static diffraction is negligible, and we can achieve near unitary conversion efficiency and ultra-high isolation ratio (Fig. 3-6)

Figure 3-6 Complete photonic transitions with ideal temporal phase modulation depth $\Delta \varphi = 2.405$ radians. The phase modulation parameters are the same as Figure 3-5 except that $\Delta \varphi$ is equal to 2.405 radians leading to $J_0(\Delta \varphi) = 0$. The static diffraction of the metasurface is almost completely eliminated.

### 3.3 Design

We used a set of nanobar antennas made of amorphous silicon ($\alpha$-Si), which has a large Kerr index and low optical loss, as the building blocks of the metasurface. With the adoption of a 50-nm-thick SiO$_2$ spacer layer and a silver back-reflector plate to create a gap resonance, the nanoantenna can induce a large phase shift (over $2\pi$) upon the incident light. The permittivity of the $\alpha$-Si nanoantennas can be changed by the resonance enhanced optical field due to the nonlinear Kerr effect (Fig. 3-7 a). This is an ultrafast process and is the key to obtaining the THz temporal phase modulation. Subsequently, the resonance response of nanoantennas will change and lead to shift in the phase (Fig. 3-7 b). Numerical simulations were carried out using a commercially available finite element method (FEM) solver package – COMSOL Multiphysics – with periodic boundary conditions for a single building block. Third-order finite elements and at least 10 mesh steps per wavelength were used to ensure the accuracy of the calculated results. The experimentally obtained optical constants of silver and amorphous silicon were used to model the back reflector and the nanoantennas. The refractive index of the spacer layer (SiO$_2$) was chosen to be 1.45. The scattered field formulation was used to calculate the reflected light at $\lambda = 860$ nm, and the nonlinear simulation method
introduced in chapter 2 was used to calculate the pump induced phase shift change. By sweeping the size of nanoantennas ($l_x$ and $l_y$), we mapped out the static phase shifts which cover over $2\pi$ range. Three nanoantennas were chosen to construct a unit supercell (marked by red diamonds Fig. 3-8 b), because they uniformly spanned the $2\pi$ phase shift range with a constant difference of $2\pi/3$, and they have the same pump induced phase shift change (Fig. 3-7 c). Fig. 3-8 c illustrates the simulated electric field distribution of a supercell at normal incidence ($\lambda = 860$ nm), showing a smoothly slanted wavefront. We also simulated the diffraction efficiency under different incident angles (Fig. 3-8 d), finding that a high diffraction efficiency above 84% can be achieved near normal incidence (within $\pm 10$ degrees).

Figure 3-7 (a) Simulation of the pump (800 nm) induced permittivity change of a nanoantenna. A quarter of the antenna is displayed for a better illustration of the distribution of relative permittivity. (b) Electrical field distribution of the probe beam (860 nm) reflected from the arrayed nanoantennas (left panel) without and (right panel) with a pump beam, respectively. The abrupt phase shift of the nanoantenna is changed by $\Delta \phi = 2\pi d/\lambda$, where $d$ is the shift in
The wavefront of the reflected light. (c) The simulated abrupt phase shifts of the three constituent nanoantennas vs. peak power intensity of the pump. (d) Reflection phase shift distribution across the modulated metasurface, with black solid line representing the static phase distribution, red solid line representing the spatiotemporally modulated phase at time instant $t$ and yellow dashed line representing the phase at $t+\Delta t$.

Figure 3-8 (a), A 3D illustration of a unit cell of the metasurface which consists of three $\alpha$-Si nanobar antennas. The thickness of the sliver ground plate, the SiO$_2$ spacer layer, and the $\alpha$-Si nanoantennas are 200 nm, 50 nm, and 150 nm, respectively. (b), Calculated phase shifts (surface plot) of reflected light in a 2D parameter space spanned by $l_x$ and $l_y$. It is overlaid by contour lines showing the pump-induced phase shift change of 0.32 radians (black dashed line) when illuminated with pump light at intensity 15 GW/cm$^2$. The white lines are the contours indicating the evenly spaced phase shifts in the static condition. Three different nanoantennas which cover $2\pi$ static phase shifts with an interval of $2\pi/3$ are chosen as the building blocks to construct the metasurface, as marked by the red diamonds intersecting the two types of contour lines. (c), A pseudo-color plot of the reflected electric field distribution from a designed metasurface with normal incidence. (d), Diffraction efficiencies of the metasurface at different incident angles (i.e. different wavevector along the $x$ axis).
3.4 Experiment

3.4.1 Sample Fabrication

The sample was fabricated following the process shown in Fig. 3-9. A 200 nm layer of silver was deposited onto a silicon substrate with a 5 nm Ge adhesion layer by electron-beam (e-beam) evaporator deposition (SEMICORE E-Gun Thermal Evaporator). A 50 nm SiO2 dielectric spacer layer and 150 nm amorphous silicon layer were then grown by plasma enhanced chemical vapor deposition (PECVD). The metasurface nanoantennas were created using a sequential process of electron-beam lithography (EBL), lift-off of chromium mask and inductively coupled plasma - reactive ion etching (ICP-RIE). In the EBL process, we employed a 1:1 diluted ZEP 520A e-beam resist to achieve high resolution. The total pattern size was 200 by 200 μm², written using a Vistec 5200 100 kV e-beam writer. A chlorine-based plasma RIE recipe involving Cl₂ and Ar gas was used to etch amorphous Si, creating the nanoantennas. The sample was finally immersed in a chromium etchant to remove the mask. Fig. 3-10 a shows a Field emission scanning electron microscopy (FESEM) image of the final sample.

Figure 3-9 Nonreciprocal metasurface fabrication process flow.
3.4.2 Linear Measurement

To characterize the sample’s static diffraction properties, we adopted the k-space imaging technique based on an inverted microscope (Nikon Eclipse TE 2000U). As shown in Fig. 3-10 b, the red dashed line represents the real space imaging light path, and the blue dashed line traces the k-space imaging light path. The sample was illuminated by a Ti:Sapphire femtosecond laser through an objective (Nikon Plan Fluor 60X/0.95 NA), and the reflected light was collected by the same objective. To acquire two-dimensional (2D) k-space images, the back focal plane of the objective was directly imaged by a Bertrand lens before a charge-coupled device (CCD) camera. Based on the k-space measurements, the diffraction efficiency of the metasurface was extracted and calibrated (Fig. 3-10 c). It exhibits a high efficiency of 84% near 860 nm. According to generalized Snell’s law $k_x = k_s + k_{inc}$, therefore the deflected angle fulfills $\sin(\theta) = \lambda / p_x$ for a normal incident light. We plotted the theoretical angles with the measured reflection angles at different wavelengths and periods, showing a good agreement (Fig. 3-10 d).

Figure 3-10  **Linear characterization of the metasurface.** (a) Field emission scanning electron microscopy (FESEM) image of a fabricated α-Si metasurface. Scale bars in the main figure and the inset are 1µm and 200nm, respectively.
(b) A schematic of the k-space imaging setup used to measure the reflection angle and static diffraction efficiency of the fabricated metasurface. (c) Experimentally measured static diffraction efficiencies of the sample with \( p_x = 1500 \) nm at different wavelengths. (d) Measured (red circles) and calculated (blue line) anomalous reflection angles of the metasurface at wavelengths ranging from \( \lambda = 680 \) nm to 1000 nm at normal incidence, with \( p_x = 1.5 \) μm. The inset shows the anomalous reflection angles for samples with \( p_x \) varying from 1.2 μm to 1.8 μm at \( \lambda = 860 \) nm. The experimental results agree well with the theory.

3.4.3 Measurement of Nonreciprocal Light Reflection

In order to create the spatiotemporal modulation, we used the method introduced in chapter 2. The experimental setup is illustrated in Fig. 3-11 a. Two frequency shifted pump beams were split from the output of a Ti:Sapphire pulsed laser radiation (140-fs pulse width, 80-MHz repetition rate) at \( \lambda = 800 \) nm. To create the probe beam, the other small portion of the Ti:Sapphire pulsed laser radiation was sent to a nonlinear photonic crystal fiber (PCF) to generate a supercontinuum. The probe light at 860 nm wavelength was selected using a monochromator. By adjusting delay line 2, the probe can be synchronized with the pumps. An aspheric lens with effective NA of 0.76 focused the three beams onto the metasurface which was mounted on a three-dimensional (3D) translation stage. Due to aberration of the aspheric lens, the focal spot of the pump beams is an ellipse with major and minor axis lengths of 50 μm and 45 μm, respectively. The reflected signal was directed to a fiber coupler by a D-shaped pickup mirror, of which the position was adjusted by a linear translation stage to collect the k-space information of the output signal. In our experiment, the center wavelength difference was around 6 nm and the frequency difference was around 2.8 THz (Fig. 3-11 b). In addition, \( k_M \) was adjusted by changing the angle between the two pump beams impinging onto the metasurfaces. Fourier transform analysis of the interference pattern shows a \( k_M \) equal to 0.54\( k_{\text{probe}} \) (Fig. 3-11 c), where \( k_{\text{probe}} \) is the length of the free-space wavevector of the probe beam.
Figure 3-11 Experimental demonstration of controllable space-time phase modulation. (a), A schematic of the experimental setup. The output of a Ti:Sapphire femtosecond pulsed laser at 800 nm is split into two beams: one is directed through a transmission grating to generate frequency-shifted pump beams; the other is sent to a photonic crystal fiber (PCF) to create a wavelength-tunable probe beam. Two delay lines are employed in order to achieve the temporal synchronization among the three beams. An aspheric lens focuses pump and probe beams onto the metasurface. The reflected signal is picked up by a D-shaped mirror and detected by a spectrometer. We map out the frequency and momentum of the reflected signal by monitoring the collected spectra across the Fourier plane of the aspheric lens. (b), Spectra of the two pump beams, showing a wavelength difference of 6 nm (corresponding to $\Delta f$ of 2.8 THz). (c), The 2D Fourier transform of the interference pattern of the two pump beams, revealing a $k_M$ equal to $0.54k_{\text{probe}}$, where $k_{\text{probe}}$ is the free space wavevector of the probe light at $\lambda = 860$ nm.

To demonstrate the nonreciprocal light propagation, the metasurface with $k_s = 0.72k_{\text{probe}}$ was imprinted by the interference pattern at a peak pump intensity around 1 GW/cm$^2$, which generated a dynamic phase modulation with $\Delta f = 2.8$ THz and $k_M = 0.54k_{\text{probe}}$. For the forward propagation experiment, the 860 nm probe light hit the metasurface at normal incidence and resulted in a reflected wave with a shift in the energy-momentum space. This shift was captured by collecting the reflected spectra from a fiber aperture scanning spatially on the Fourier plane (along the $k_s$ direction) of the focusing lens before the metasurface. Fig. 3-12 a displays a static diffraction at 348.6 THz and $0.72k_{\text{probe}}$ determined by $k_s$, and a
down-shifted signal at 345.8 THz and $0.18k_{\text{probe}}$ produced by the spatiotemporal modulation induced $\Delta f$, $k_M$, and $k_s$. The 0th-order reflection can also be detected, as the diffraction efficiency dropped due to edge effects and slight polarization misalignment. The aspheric lens used to focus the pump and probe beams and to collect converted signals has an effective NA of 0.76; $k_x/k_{\text{probe}}$ is therefore bounded by the limited collection angle. On the other hand, the accumulated $k_x/k_{\text{probe}} \sim 1.26$ of the upward transition is greater than unity. Therefore, it is evanescent and cannot carry energy away from the metasurface. For the backward propagation experiment, we sent in a probe beam ($f = 345.8$ THz and $k_x = -0.18k_{\text{probe}}$) with the same frequency as the previous down-converted signal but the opposite direction onto the metasurface. We observed another downward transition at 343.0 THz exit along the normal direction (Fig. 3-12 b). Similarly, the upward transition is nonexistent since its accumulative $k_x$ is greater than $k_{\text{probe}}$. Therefore, the backward propagating light cannot return to the initial state. We also did a fine scan over the $\omega$-$k_x$ regions where high order conversions may exist. But no converted signals were observed since these processes have much lower efficiency. In addition, we did a control experiment on amorphous silicon film (thickness ~ 150 nm) using similar pump intensity, but no converted signal was detected. The experimental results match with the FDTD simulation results in Fig. 3-5.
Experimental demonstration of nonreciprocal light reflection on the space-time phase modulated metasurface. (a) Under dynamic modulation with $\Delta f = 2.8$ THz and $k_M = 0.54k_{probe}$, the energy-momentum diagram of the normal-incident probe beam ($f = 348.6$ THz) on the metasurface ($k_s = 0.72k_{probe}$) shows a downward converted signal at $f = 345.8$ THz and $k_s = 0.18k_{probe}$. (b) In the backward case, a probe beam having the same frequency with previous signal ($f = 345.8$ THz) but the flipped direction ($k_s = -0.18k_{probe}$) was sent onto the metasurface. The energy-momentum diagram shows a further down-shifted signal at $f = 342.0$ THz exiting in the normal direction ($k_s = 0$). In both cases, the converted signals are magnified by $10^5$ for better illustration (the magnified regions are enclosed by the dashed white boxes). These results are perfectly matched with our theoretical prediction depicted in Fig. 1d and e. In the regions of interest (where all the possible photonic transitions exist), finer scanning steps and long spectrometer integration time were used to ensure to detect converted signals. In the regions with possible upper sidebands or higher-momentum sidebands, no signal was detected even with magnification. Therefore, here only the regions where the 1st-order conversions occur are magnified.
3.4.4 Analysis of the Operation Bandwidth of Nonreciprocal Metasurfaces

The bandwidth of time dependent nonreciprocal systems is usually determined by the phase matching conditions and modulation frequencies (more accurately, dynamic modulation induced frequency shift). For example, in ref. 84,174, bandwidths of a few hundred GHz were achieved by dispersion engineering at the expense of large device footprint, so that the phase matching condition was fulfilled over a larger bandwidth comparing with their counterparts 166. It should be noted that this bandwidth is valid under monochromatic excitation. For broadband pulses the bandwidth is limited by the modulation frequency since we need to avoid overlap between the main pulse and its temporal-modulation-induced sidebands, which will cause signal distortion 81,162,175. The space-time metasurface works with optical modes in the continuum (not discrete guided modes) and naturally fulfills phase matching conditions in a broad frequency range. Besides, the meta-atoms in our system has a low-quality factor (< 50) which allows the device to operate at a relatively broad bandwidth. In addition, the ultrafast modulation method features a modulation frequency $\Delta f \sim 2.8$ THz, which ensures at most 5.6 THz ($2 \times \Delta f$) bandwidth with broadband excitation (Fig. 3-15). To sum up, with either a narrowband or broadband excitation, our metasurfaces exhibit several THz bandwidth, which is at least one order of magnitude greater than the largest ones (a few hundred GHz) 84,174 reported on time dependent nonreciprocal systems.

In order to experimentally show the nonreciprocal operation wavelength range, we conducted additional measurements on the nonreciprocal processes by mapping out the dispersion diagrams (Fig. S3-12) with a narrowband probe of which the center wavelength is ranging from 854 to 914 nm for both forward and backward reflections. By extracting the conversion efficiencies at the tested wavelengths, a 3-dB bandwidth (full-width at half-maximum, FWHM) of approximately 5.77 THz was demonstrated (Fig. 3-14). It is worth noting that in contrast to the waveguide-based systems, the bandwidth of our space-time metasurface is not constrained by the phase-matching conditions. Our experimentally obtained bandwidth is at least one order of magnitude greater than the largest ones reported (a few hundred GHz) 84,174 on time-dependent nonreciprocal systems.
Figure 3-13 **Nonreciprocal light reflections at probe wavelength from 854 nm to 914 nm.** In the forward-propagating cases (figures in the left column), a normal incident light (with frequency $f$) was reflected by the metasurface, leading to a frequency down-shifted signal ($f - \Delta f$) with $k_x = k_{ms} - k_M$. In the backward-propagating cases (figures in the right column), the time-reversed counterpart of the signal was sent back and interacted with the metasurface again, leading to a reflected signal with frequency $f - 2\Delta f$ exiting at the normal direction. The dynamic modulation has $\Delta f = 2.8$ THz and $k_M = 0.35k_{probe}$ where $k_{probe} = 2\pi / (860 \text{ nm})$. The metasurface has a period of 1650 nm, corresponding to $k_{ms} = 2\pi / (1650 \text{ nm})$. The signals in the dashed white boxes are magnified by a factor of $4 \times 10^5$.

Figure 3-14 Conversion efficiencies and linear reflectance spectra of the space-time metasurface experimented in Fig. 3-12. The normalized conversion efficiencies (top panel) show a similar response as the reflectance (bottom panel), which exhibit a broad resonance around 885nm.
3.4.5 Tunable Nonreciprocal Reflection

Using our space-time metasurface, we are able to control independently the static phase gradient $k_s$, the dynamic spatial frequency $k_M$, and the temporal modulation frequency $\Delta \omega$, which provides unprecedented tunability in manipulating the photonic transitions. The modulation frequency can be changed by adjusting the frequency splitting between the two pumps (Fig. 3-16 a). Right now, it is limited by the pulse width of our laser source. However, we can realize a large and tunable $\Delta \omega$ with two optical parametric amplifiers (OPA) \(^{176}\), which can be used to build a tunable frequency-shifting optical isolator. Meanwhile, $k_M$ can be changed by adjusting the angle between the two pumps. Fig. 3-16 b displays the Fourier transform.

Figure 3-15 **Bandwidth of the nonreciprocal reflection of a broadband pulse incident on the space-time metasurface.** The dashed lines represent the incident light, and the solid lines represent the reflected signals in both forward- and backward-propagating cases. As long as $2\Delta f$ (frequency shift between the forward incidence and the back-reflection) is larger than $\Delta f_{\text{FWHM}}$ of the pulse, the back-reflection (solid red line) can be distinguished from the forward incident light (dashed blue line). Therefore, robust nonreciprocity is preserved with a bandwidth of at most $2\Delta f$. 

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\(^{176}\) Reference or notation for the two optical parametric amplifiers (OPA)
analysis of interference patterns (insets) with varying \( k_M \). It should be noted that the interference pattern is produced by the common frequency components in each pump, otherwise it would not be possible to observe the interference fringes because the pattern moves at high speed. By changing \( k_s \) we can selectively enable downward or upward transitions (Fig. 3-17).

In addition, we demonstrated nonreciprocal reflections with arbitrary transverse momenta by tuning both \( k_s \) and \( k_M \) along the metasurfaces (Fig. 3-18). We note that the conversion efficiency decreases from \( 10^{-4} \) to \( 10^{-5} \) as \( k_M \) increases. Since the two pumps are separated further apart to create increased \( k_M \), the focusing spot of the pumps is broadened as a result of the larger intersection angle between two pumps and distorted due to worse lens aberrations. Therefore, the peak pump intensity gradually decreases at the same input pump power, resulting in a reduced efficiency. Meanwhile, at larger \( k_M \), the period of dynamic modulation decreases and covers fewer nanoantennas in a single period, which further decreases the efficiency. In addition, with the same \( k_M \), the conversion efficiency increases as \( k_s \) increases. A larger \( k_s \) corresponds to a smaller supercell period; therefore more nanoantennas can be accommodated in a unit dynamic modulation period, which leads to a stronger modulation strength and higher efficiency. It is worthy to note that this level of controllability and flexibility has not been achieved in previous time-variant nonreciprocal systems.

![Figure 3-16](image)

Figure 3-16 (a) Pump beam spectra with central wavelength differences of 6 nm and 5 nm, respectively. (b) 2D Fourier transform of the interference patterns with various \( k_M \). Insets show the interference pattern generated by the common frequency components in two pumps.
Figure 3-17 **Experimental demonstration of direction selectivity of the photonic transitions.** (a) Only downward photonic transition occurs on a metasurface ($k_s = 0.72 k_{\text{probe}}$) modulated by $k_M = 0.32 k_{\text{probe}}$ and $\Delta f = 2.8$ THz. The converted signal is magnified by $10^4$ for better illustration. (b) On the contrary, with the same temporal phase modulation on a metasurface with $k_s = -0.72 k_{\text{probe}}$, only upward photonic transition takes place. The converted signal is magnified by $10^5$ for better illustration (the magnified regions are enclosed by the dashed white boxes).
Figure 3-18 (a-d) Dispersion diagrams of metasurface with $k_s$ changing from $0.48k_{\text{probe}}$ to $0.64k_{\text{probe}}$ at a fixed $k_M = 0.32k_{\text{probe}}$. This set of experiments were performed at the same total pump power of 320 mW. (e-h) At fixed $k_s = 0.72k_{\text{probe}}$, the dispersion diagrams of the metasurface modulated with $k_M$ changing from $0.32k_{\text{probe}}$ to $0.54k_{\text{probe}}$. This set of experiments were performed at a similar pump peak intensity of 1 GW/cm$^2$. The converted signals are magnified by $10^4$ or $10^5$ for better illustration.
3.4.6 Analysis of the Conversion Efficiency

As predicated by equation (3-30), the first order conversion efficiency is proportional to $J_1(\Delta \phi)^2$. Fig. 3-19 compares the experimentally acquired efficiencies at different peak pump intensities with the theory. In general, the efficiency was on the order of $10^{-4}$ with the pump intensity below the damage threshold of the sample. It started to deviate from the theoretical predication when the peak power intensity increases beyond 1 GW/cm$^2$. This is due to thermal damages resulting from the high repetition rate (80 MHz) laser. Nevertheless, the conversion efficiency (dashed line) increases super-linearly with increasing peak power intensity, which could lead to a large boost in efficiency if pumped with a low-repetition-rate and high energy laser. In addition, the pulse width of the probe is stretched to 2 ps due to a series of nonlinear effects in the PCF. The focal spot of the probe is about 5 times larger than that of pumps. Therefore, the converted signal only comes from a small fraction of the incident probe. By improving the temporal and spatial quality of probe, the conversion efficiency can be further increased. Despite being limited by the experimental conditions, the conversion efficiency we achieved is still two orders of magnitude greater than the efficiency of the third-order nonlinear generation reported to date in amorphous silicon nanostructures at comparable pump intensities$^{177}$.

In addition, as shown in our FDTD simulation results (Fig. 3-6), the conversion efficiency reaches up to 100% when $J_0(\Delta \phi = 2.405)^2$ equals to zero, which is attainable with optimized nonlinear materials and metasurface designs. Here we propose to increase modulation depth and conversion efficiency with a low pump power requirement using the following methods.
Figure 3-19 Conversion efficiency of the nonreciprocal metasurface. The calculated and measured down-conversion efficiency on a sample with $k_s = 0.57k_{\text{probe}}$ and $k_M = 0.32k_{\text{probe}}$. They agree well with each other below the damage threshold (1 GW/cm$^2$) of the sample. At higher peak power intensity, the conversion efficiency saturates as the thermal effect permanently damage the sample.

First, we can use materials with large nonlinearity to construct the meta-atoms. It has been reported$^{66}$ that ITO in the ENZ region has extremely large nonlinear Kerr index that is around two orders of magnitude greater than that of amorphous silicon. A recent study$^{73}$ combined ITO thin film with gold nanoantennas and demonstrated even higher effective Kerr index ($\sim 3.73$ cm$^2$/GW). To achieve a similar nonlinear phase shift ($\sim 0.03$ radians) as demonstrated in our experiment, the required pump power intensity is below 100 MW/cm$^2$ with the ITO-antenna system. In addition, this ITO-nanoantenna system achieves a maximum nonlinear phase shift of 0.68 radians at pump intensity of 3.27 GW/cm$^2$ (energy density $\sim 6.1$ pJ/µm$^2$). According to equation (2) in our paper, the conversion efficiency can be

$$\frac{J_1(\Delta \phi)^2}{\sum_{n \in \{\text{all existent orders}\}} J_n(\Delta \phi)^2} \times 100\% \approx 12\%,$$

which is about three orders of magnitude greater than that achieved in our experiment. Furthermore, by using high-quality-factor resonant nanoantennas and ENZ materials with low damping factors, the nonlinear phase shift can be further increased even at a moderate pump intensities$^{72,178}$. It is worth noting that this conversion efficiency is realized with a subwavelength
interaction length ~ 50nm (total thickness of gold nanoantenna and ITO film). In comparison, a recent demonstration on travelling modulation induced nonreciprocal system shows mode conversion efficiency of 1% at the cost of on-chip optical driving power of 90 mW and an interaction length of 2.39 cm.

On the other hand, interaction time/length can be increased by stacking two space-time modulated metasurfaces to form a cavity. Photons are trapped inside the cavity to enable much longer interaction time with the travelling-wave modulation, leading to a boosted nonreciprocal conversion efficiency. In addition, a metasurface integrated resonator system can be used to increase the effective interaction length. For example, we can integrate metasurfaces on top of a micro-ring resonator, and apply a travelling wave modulation across the metasurface. Photons circulating inside the resonator will interact with the travelling-wave modulation for a time determined by the quality factor (Q-factor ~ 10^3 ~ 10^6) of the ring resonator, which is much larger than that of our current resonant antennas (Q-factor ~ 40).

3.4.7 Pump-probe Study of the Ultrafast Optical Modulation

To study the temporal response of the nonreciprocal metasurface, we first characterized the temporal information of the pump and probe beams used in our experiment. We carried out the pumps correlation and probe-pump cross-correlation experiments using the setup shown in Fig. 3-11 a. The top panel in Fig. 3-20 a shows the correlation sum frequency generation (SFG) signal of the two frequency shifted pump beams at different time delay using a Beta Barium Borate (BBO) crystal. The pulse width of the Ti:Sapphire laser is 140 fs, and it is stretched to 400 fs after passing through dispersive optical components and split into two frequency-shifted components with half of the original bandwidth. The middle panel is the cross-correlation of the probe and the blue-shifted pump beam measured by monitoring the SFG signal at different time delay by adjusting the delay line 2. It can be seen that the pulse width of the probe is approximately 2 ps.
Next, the metasurface was pumped by the two frequency-shifted pump beams and probed by a weak beam of 869 nm. We monitored the down-conversion signal ($\lambda = 876$ nm) shown in Fig. 3-20 b at different probe-pumps delay by adjusting delay line 2. Due to the limitation of the probe pulse width (~2 ps), our experiment doesn’t have enough resolution to obtain the exact response time of the nonlinear Kerr effect of amorphous silicon metasurface. However, the down-conversion signal evolution follows the same envelope of the pump-probe cross correlation, proving that the Kerr nonlinear response time of the metasurface is at least within picosecond scale and thus ruling out the involvement of thermal effect in the phase modulation. According to the Ref. 179, the nonlinear Kerr effect response time of amorphous silicon wire waveguide is less than 100 fs. Thus, our pump-probe experiment confirmed that the dynamic phase changes is originated from the nonlinear Kerr effect.

Figure 3-20 (a) The cross-correlation signal of two frequency-shifted pump beams (top panel), the cross-correlation signal of the probe and the pump (middle panel), and the evolution of down-converted signal ($f = 342.4$ THz, $\lambda = 876$ nm) with different delay of the pumps. (b) $k$-space frequency distribution of the down conversion process of a normal incidence ($f = 345.2$ THz, $\lambda = 869$ nm) upon the metasurface ($k = 0.73k_{\text{probe}}$) and modulated by $\Delta\omega = 2.8$ THz and $k_{M} = 0.35k_{\text{probe}}$. The down-conversion signal is monitored while adjusting the delay line 2, and the results are plotted in the bottom figure of (a). The down-converted signal ($f = 342.4$ THz, $\lambda = 876$ nm, and $k_x = 0.38k_{\text{probe}}$) is scaled by $10^4$ for better illustration.
3.5 Conclusion

We experimentally demonstrated nonreciprocal light reflection based on a spatiotemporal phase gradient nonlinear metasurface. The heterodyne interference created by frequency-shifted pump beams provides robust and controllable spatiotemporal modulation, of which $\Delta \omega$ and $k_M$ can be readily tuned as desired. It is worth noting that more complex two-dimensional spatiotemporal modulation can be constructed from the heterodyne interference of three or more pump beams. The spatiotemporal phase modulation greatly expands the functions of conventional static phase gradient metasurfaces, providing an additional degree of freedom for manipulating the temporal properties of light and achieving nonreciprocal light propagation. Particularly, we achieved in our experiment a 2.8 THz modulation frequency, a huge step towards optical frequencies, and an approximately 5.77 THz 3 dB bandwidth, which is orders of magnitude greater than that of current time-variant nonreciprocal systems to the best of our knowledge.

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Chapter 4

Parametrically Excited Time Variant Metasurfaces for Electrically Tunable Second Harmonic Generation

Parametric resonance is an important physical effect in nonlinear wave-matter interactions. It can lead to generation and amplification of nonlinear signals, which enables many crucial applications such as parametric oscillators and parametric amplifiers. Lying at the heart of this effect is a time-variant perturbation to the properties of a resonant system, which is difficult to realize in a nanoscale optical resonator. In this chapter, I will introduce how to generate parametric resonance on optical metasurfaces and its application for second harmonic generation (SHG). Using a travelling-wave modulation created by a direct current (dc) electric field and an optical field, we parametrically drive large-amplitude resonance oscillations of amorphous silicon (α-Si) meta-atoms – nanoscale building blocks of a metasurface, and achieve a gigantic SHG. A Mathieu’s equation involving a time varying resonance captures the signatures of experimentally measured SHG, such as a super-quadratic dc field dependence, and provides insight into SHG enhancement through parametric excitation. We observed greatly enhanced SHG with a gain factor of 28 dB and an electric-field-controlled on/off enhancement factor over $10^4$. Our work provides a compact, electrically tunable and CMOS compatible approach to boosting and controlling nonlinear light generations. It holds great potential for applications in optical communications and signal processing.

4.1 Background and Motivation

Parametric resonances – originated from a time-varying property of a resonant system – has been an interesting topic explored in research areas ranging from mechanical engineering, quantum physics, plasma physics to electronics and radio-frequency engineering. Instead of directly driving the resonator with an external force, one applies a time-variant modulation on its physical
parameters, which in turn modulates the fundamental frequency of the resonator. For mechanical systems, these extraordinary behaviors can be modeled using a Mathieu’s equation, which involves a sinusoidal time-dependent modulation of the natural frequency of the resonator. A simple example is ‘children on a swing’, where the child moves in a up-and-down motion that effectively changes the length of the swing, and thereby imposing a time-varying modulation to the swing’s natural frequency $\omega_0$. According to the Mathieu’s equation, one can swing by pumping at particular frequencies $2\omega_0/n$, for any integer $n$. The most well-known case, namely parametric amplification, happens at $n = 1$, when the child moves the center of mass up-and-down at twice the swing’s natural frequency.

In optics, however, this high frequency pumping strategy requires an even-order nonlinear system, which is only possible in materials with inversion symmetry. Most conventional optical parametric oscillators (OPOs) and amplifiers (OPAs) utilize bulky second order nonlinear crystals and large optical cavities, which are too cumbersome to be integrated into modern photonic chips. In addition, integration of those second order nonlinear crystals is not compatible with the current complementary metal-oxide-semiconductor (CMOS) technology. Although on-chip parametric oscillation and amplification through four-wave mixing (FWM) have been demonstrated, yet they are limited by strict phase matching requirements, extremely long interaction lengths (millimeters to centimeters), and high pump power. It remains a challenge to create strong parametric resonances without the aforementioned limitations on a CMOS-compatible platform.

A new opportunity for miniaturized OPO has emerged with the development of time-variant metasurface – an artificially engineered surface consisting of subwavelength building blocks known as meta-atoms whose properties can be tailored both in space and time. In contrast to conventional nonlinear metasurfaces of which properties are static and difficult to change, the resonant property of time-variant metasurface can be modulated in time, leading to a strong parametric resonance and remarkable nonlinear properties. Here, we created such a time-variant optical metasurface consisting of amorphous silicon ($\alpha$-Si) meta-atoms that shows prominent time-dependent resonance shifts under travelling-wave
temporal modulation, and experimentally demonstrated tremendously enhanced second harmonic generation (SHG). Taking advantage of the large nonlinear Kerr effect of $\alpha$-Si meta-atoms, an ultrafast temporal permittivity modulation was achieved in response to the heterodyne interference between a control direct current (dc) electric field and an optical pump field. This in turn leads to a large temporal resonance shift accompanied by a dynamic phase shift change $\Delta \phi$. We used the Mathieu’s equation to model the dynamic process and found that the SHG intensity scales with $J_{12}(\Delta \phi)/J_{02}(\Delta \phi)$ and in addition has a super-quadratic dc electric field dependence, which agrees well with our experiment results. Compared with conventional electric field induced second harmonic (EFISH) generation which similarly uses a dc field on materials with third order nonlinearity to generate frequency doubled waves, our approach exhibits a gain factor of 28 dB at a mild dc field strength of $3.7 \times 10^4$ V/cm. We obtained a dc electric field controlled on/off enhancement factor ($\Delta I_{\text{SHG}} (V_{dc} = 37 \text{ V}) / I_{\text{SHG}} (0 \text{ V})$) over $10^4$, which are two to three orders of magnitude greater than those of conventional EFISH. A fast electric switching of SHG is also demonstrated, which opens opportunities for electrically controlled optical switching. Leveraging the time-varying phase/resonance frequency modulation of meta-atoms, our metasurfaces represent a new paradigm for electrically controlled efficient second harmonic generation. It holds great promise for constructing ultra-compact photonic/optoelectronic devices for on-chip coherent light generation, ultrafast optical switching, and information processing.

4.2 Theory

As illustrated in Fig. 4-1 a, a temporal modulation at frequency $\Omega$ shifts the resonance of meta-atom periodically, leading to a dynamic change of both the amplitude and the phase of meta-atoms’ optical response. These dynamic resonators constitute a time grating, from where the new frequency component will be diffracted (Fig. 4-1 a). This picture of nonlinear generation is commonly used in interpreting FWM which don’t have requirement on the modulation frequency, however, our case differs in that the meta-
atoms are parametrically modulated at specific frequencies $2\omega_0/n$ at which large-amplitude resonances can be excited.

![Diagram](image)

**Figure 4-1** Principle of parametric excitation enhanced second harmonic generation. (a) A time-varying modulation induces a time dependent resonant property of meta-atom. These parametrically excited resonators constitute a time grating, from which the new frequency component will be diffracted. (b) Schematic of the α-Si metasurface biased by interdigitated (IDT) electrodes array. The interference between dc electric field and pump optical field creates a travelling-wave modulation on the properties of meta-atoms, leading to efficient parametric modulation at a frequency equal to the resonance frequency of meta-atoms. The inset shows the resonance wavelength red-shifts under modulation.

In our case, the modulation is generated by the heterodyne interference of a dc electric field and an optical field with frequency $\omega_0$, leading to $\Omega = \omega_0$. This heterodyne intensity modulation induces a permittivity modulation of the meta-atom through nonlinear Kerr effect, which will shift the resonance and
subsequently induce a phase shift of $\Delta \varphi$. Assuming the metasurface as a collection of Lorentzian oscillators, their phase can be expressed as

$$\varphi = \arctan\left(\frac{\omega \gamma}{\omega_0^2 - \omega^2}\right) \quad (4-1)$$

where $\omega_0$ is the resonance frequency, $\gamma$ is the decay rate and can be expressed as $\gamma = \frac{\omega_0}{Q}$, the quality factor $Q$ of the metasurface can be calculated from Fig. 4-5 a, and it is around 50. The phase change $\Delta \varphi$ is related to the resonance frequency shift by

$$\Delta \varphi = \varphi'(\omega_0) - \varphi(\omega_0) = \arctan\left(\frac{\omega_0 \gamma}{(\omega_0 + \Delta \omega_0(t))^2 - \omega_0^2}\right) - \frac{\pi}{2} \approx \arctan\left(\frac{\gamma}{2 \Delta \omega_0(t)}\right) - \frac{\pi}{2} \quad (4-2)$$

Here, it is assumed that the change of decay rate $\gamma$ is negligible and the temporal modulation to $\omega_0$ is small.

After algebraic manipulation, we obtain

$$\Delta \omega_0(t) = -j\frac{\gamma}{2} \frac{e^{j\Delta \varphi(\omega_0)}}{e^{j\Delta \varphi(\omega_0)} - e^{-j\Delta \varphi(\omega_0)}} \quad (4-3)$$

Using Jacobi-Anger expansion $e^{\Delta \varphi(\omega_0)} = \sum_{n=-\infty}^{\infty} j^n J_n(\Delta \varphi)e^{jn\omega_0 t}$, equation (4-3) is further simplified to

$$\Delta \omega_0(t) = \frac{\gamma}{2} \sum_{k=0}^{\infty} (-1)^k J_{2k+1}(\Delta \varphi) \cos((2k+1)\omega_0 t) \approx \gamma \frac{J_1(\Delta \varphi) \cos(\omega_0 t)}{J_0(\Delta \varphi)} \quad (4-4)$$

In equation (4-4), the higher order Bessel functions ($k > 1$) are omitted, because they are much smaller than the zeroth and first order Bessel functions when $\Delta \varphi$ is small.

Next, we take the differential equation of motion of a parametrically excited oscillator to be:

$$m \ddot{x} + \frac{m\omega_0}{Q} \dot{x} + m\left(\omega_0 + \Delta \omega_0(t)\right)^2 \dot{x} + \tilde{\eta} x^2 \ddot{x} = 0 \quad (4-5)$$
where \( m \) is the effective mass, \( \omega_0 \) is the resonance frequency, \( Q \) is the quality factor of the resonance, and \( \tilde{\eta} \) is the coefficient for nonlinear damping which increases with amplitude of oscillation. We use the convention that physical parameters that immediately rescaled appear with twiddles. To scale away unnecessary parameters and leave only those that are physically significant, we do: (i) Measuring time in units of \( \omega_0^{-1} \) so that the dimensionless time variable is \( t = \omega_0 \tilde{t} \). (ii) Omitting the small term \( \Delta \omega_0(t)^2 \). (iii) Dividing the equation by an overall factor of \( \omega_0^2 \). This yields a scaled equation of the form

\[
\ddot{x} + Q^{-1} \dot{x} + (1 + H \cos(t))x + \eta \dot{x}^2 \dot{x} = 0
\]

(4-6)

where dots denote derivatives with respect to the dimensionless time \( t \), \( H = \frac{2J_1(\Delta \phi)}{QJ_0(\Delta \phi)} \) is proportional to the parametric drive, and \( \eta = \frac{\tilde{\eta}}{m \omega_0} \). It is noted that equation (4-6) assumes the form of a damped Mathieu’s equation.

Making use of secular perturbation theory and following the derivation used in paper, we shall use the damping \( Q^{-1} \) to define the small expansion parameter \( \epsilon \) (\( \epsilon \ll 1 \)). Considering the limit of weak oscillations, we require the deviation from equilibrium \( x \) to be on the order of \( \sqrt{\epsilon} \). We take the pump frequency to be an amount \( \epsilon \Omega_p \) away from the resonant frequency, and take the parametric drive amplitude to scale as the damping, \( \text{i.e.} \) we set \( H = \sqrt{\epsilon} \Delta \). The scaled equation of motion is therefore

\[
\ddot{x} + \epsilon \dot{x} + \left(1 + \sqrt{\epsilon} \Delta \right) \cos \left(\left(1 + \epsilon \Omega_p \right)t\right)x + \eta \dot{x}^2 \dot{x} = 0
\]

(4-7)

We try an expansion of the solution of the form

\[
x(t) = e^{\frac{i}{2} \int t} \left( A(T) e^{i \phi} + c.c. \right) + \epsilon x_{1/2}(t) + \epsilon^{3/2} x_1(t) + ... \]

(4-8)

The lowest order contribution to this solution is based on the solution to the linear equation of motion of a simple harmonic oscillator \( \ddot{x} + x = 0 \), where \( T = t \epsilon \) allows the complex amplitude \( A(T) \) to vary slowly in time. The slow temporal variation of \( A(T) \) also ensures that the perturbative correction \( x_1(t) \), as well as all
higher-order corrections to the linear equation, do not diverge. Substituting this expansion into equation (4-7) we obtain at order $\epsilon^{\frac{3}{2}}$ the linear equation. Using the relation $\dot{A} = \frac{dA}{dt} = \epsilon \frac{dA}{dT} \equiv \epsilon A'$, we calculate the time derivatives of the trial solution (4-8)

$$\ddot{x} = \frac{\epsilon^{\frac{3}{2}}}{2} \left([iA + \epsilon A']e^{it} + \text{c.c.}\right) + \epsilon \dot{x}_{\frac{3}{2}}(t) + \epsilon^{\frac{3}{2}} \ddot{x}(t) + ... \quad (4-9)$$

By substituting these expressions back into (1.9), and picking out all terms of order $\epsilon$

$$\ddot{x}_{\frac{3}{2}} + x_{\frac{3}{2}} = -\frac{h}{4} \left( A e^{i\alpha e} e^{i2t} + A^* e^{i\alpha e} + \text{c.c.} \right) \quad (4-10)$$

And at order $\epsilon^{\frac{3}{2}}$

$$\ddot{x}_{\frac{3}{2}} + x_{\frac{3}{2}} = \left(-iA' - \frac{i}{2} A - \frac{in}{8} |A|^2 A\right)e^{it} - \frac{h}{8} A e^{i3t} + \text{c.c.} - \frac{h}{2} \left(e^{i(t+\Omega e)} + \text{c.c.}\right) x_{\frac{3}{2}} \quad (4-11)$$

We can directly solve equation (4-10)

$$x_{\frac{3}{2}} = \frac{h}{4} \left( A e^{i\alpha e} e^{i2t} - A^* e^{i\alpha e} + \text{c.c.} \right) + O(\epsilon) \quad (4-12)$$

The emergence of the $e^{i2t}$ term signifies the second harmonic generation. Substituting the solution for $x_{\frac{3}{2}}(t)$ to equation (4-11) contributes the additional term from the parametric driving which has the form

$$\frac{\epsilon^{\frac{3}{2}}h^2}{8} \left( -\frac{A}{3} e^{i\alpha e} e^{i2t} + A^* e^{i\alpha e} + \text{c.c.} \right) \left(e^{i\alpha e} e^{it} + \text{c.c.} \right) \quad (4-13)$$

Taking all secular terms (proportional to $\epsilon^i$) into account, equation (4-11) becomes

$$\ddot{x}_{\frac{3}{2}} + x_{\frac{3}{2}} = \left(-iA' - \frac{i}{2} A - \frac{in}{8} |A|^2 A\right)e^{it} + \frac{h^2}{8} \left( \frac{2A}{3} + A^* e^{i2\alpha e} \right) \left(e^{i3t} + \text{c.c.} \right) + \text{non secular terms} \quad (4-14)$$
The collection of terms proportional to $e^{\beta t}$ must vanish so that the perturbative correction $x_1(t)$ will not diverge. So, the equation for determining the slowly varying amplitude $A(T)$ becomes

$$\frac{dA}{dT} + \frac{i}{8} \left( \frac{2}{3} A e^{2\Omega_p T} + \frac{A}{2} + \frac{\eta}{8} |A|^2 A \right) = 0$$  \hspace{1cm} (4-15)

Ignoring initial transients and assuming that the nonlinear terms in the equation are sufficient to saturate the growth of instability, we try a steady-state solution of the form

$$A(T) = a e^{\Omega_p T}$$  \hspace{1cm} (4-16)

Substituting this steady-state solution into equation (4-15), we obtain

$$\left[ -2\Omega_p - \frac{\hbar^2}{6} + i \left( 1 + \frac{\eta}{4} |a|^2 \right) \right] a = \frac{\hbar^2}{4} a^*$$  \hspace{1cm} (4-17)

By taking the magnitude squared of both sides, we obtain a non-trivial response given by

$$\left( 2\Omega_p + \frac{\hbar^2}{6} \right)^2 + \left( 1 + \frac{\eta}{4} |a|^2 \right)^2 = \frac{\hbar^4}{16}$$  \hspace{1cm} (4-18)

When the parametric pumping frequency is fixed at the resonance frequency, corresponding to $\Omega_p = 0$, the expression for the amplitude of the response simplifies to

$$|a|^2 = \frac{1}{\eta} \left( 4 - \frac{\sqrt{5}}{3} \frac{\hbar}{\Delta} \right)$$  \hspace{1cm} (4-19)

Using the solution of equation (4-19) and substituting it back to the trial solution (4-16), we obtain the expression for the second harmonic term

$$\epsilon |x|_2 = \frac{\epsilon \hbar}{12} \left( ae^{2i} + c.c. \right) = \frac{\epsilon^2}{6} J_1(\Delta \phi) \left( ae^{2i} + c.c. \right)$$  \hspace{1cm} (4-20)

Recalling that the polarization can be expressed as $P = Nq\chi$, where $N$ is charge density and $q$ is the effective dipolar charge, the second order nonlinear polarizability is
The intensity of SHG is therefore

\[ P_{20h} = Nq e x \frac{3}{2} = Nq e^{\frac{3}{2}} \sqrt{\frac{1}{\eta} \left( 4 - \frac{2\sqrt{5}}{3} e^{\frac{1}{2}} J_1(\Delta \varphi) \right)} \frac{J_1(\Delta \varphi)}{J_0(\Delta \varphi)} \cos(2\omega_0 t) \]  

(4-21)

The intensity of SHG is therefore

\[ I_{SHG} \propto e^3 \frac{1}{9 \eta} \left( 4 - \frac{2\sqrt{5}}{3} e^{\frac{1}{2}} J_1(\Delta \varphi) \right)^2 \frac{J_1(\Delta \varphi)}{J_0(\Delta \varphi)} \]  

(4-22)

It is evident that SHG arisen from the parametric excitation at the resonance frequency can be engineered through dynamic phase change \( \Delta \varphi \), rather than limited by the inherently weak second order nonlinear susceptibility.

### 4.3 Design

To implement the parametric excitation on the metasurface, we designed \( \alpha \)-Si meta-atoms biased by interdigitated (IDT) electrodes array (Fig. 4-1 b). \( \alpha \)-Si is chosen as the constituent material because it has high refractive index, large Kerr nonlinearity, and low optical loss in the telecommunication wavelengths. The travelling-wave intensity modulation creates a time dependent refractive index change of meta-atoms, leading to a dynamic change of the optical response. In order to maximize the resonance shift, meta-atoms are specially tailored to achieve large \( \Delta \varphi \). We simulated the phase shifts (\( \varphi_{\text{linear}} \) and \( \varphi_{\text{nonlinear}} \)) of meta-atoms with various sizes (\( l_x \) and \( l_y \)) under illumination of light (\( \lambda = 1550 \) nm) at low intensity (linear) and at high intensity (0.4 GW/cm\(^2\), nonlinear), respectively, and then calculated the dynamic phase shift change (\( \Delta \varphi = \varphi_{\text{nonlinear}} - \varphi_{\text{linear}} \)). The design of \( l_x = 600 \) nm and \( l_y = 650 \) nm was chosen to provide the largest \( \Delta \varphi \) (Fig. 4-2 b). \( \Delta \varphi \) shows a super-linear dependence on the modulation intensity (Fig. 4-2 c), leading to a large temporal modulation depth at a relatively low intensity. The reflectance spectra (Fig. 4-2 d) pumped by low intensity (linear) and high intensity (0.4 GW/cm\(^2\), nonlinear) light also shows an obvious resonance shift.
4.4 Experiment

4.4.1 Fabrication

The meta-device exhibiting dynamic phase modulation enhanced SHG was fabricated by the following procedure (Fig. 4-3). Fused silica substrate was cleaned in Nanostrip heated at 60 °C for 10 – 20
minutes. First, a 100 nm deep (70 by 80 µm) trench was patterned by Electron beam lithography (EBL) and inductively coupled plasma - reactive ion etch (ICP-RIE) on the fused silica substrate. Followed by E-beam evaporation of 100 nm gold into the pre-defined trench and stripping of the photoresist, the back-reflector plate was fabricated. Then, 50 nm of Al₂O₃ was deposited by atomic layer deposition (ALD) to provide good electrical insulation. Subsequently, the IDT electrodes and E-beam markers were created using a sequential process of EBL, e-beam evaporation of 3 nm chromium (adhesion layer) and 50 nm gold, and finally the lift-off process in MICROPOSIT 1165. Then, 350nm of α-Si was deposited by plasma enhanced chemical deposition (PECVD). Nanoantennas were defined by EBL with precise alignment to the IDT electrodes array, lift-off of chromium mask, ICP-RIE etch and mask removal in Cr etchant 1020. Finally, the electrical pads were wire bonded to a printed integrated circuit board for connection to external voltage supplier.

![Fabrication process of IDT electrodes biased α-Si metasurfaces.](image)

Figure 4-3 Fabrication process of IDT electrodes biased α-Si metasurfaces.
Figure 4-4 (a) Simulated dc ($V_{dc} = 37$ V) electric field distribution of the IDT electrodes biased nanoantenna. It shows a strong electric field along the edge of the nanoantenna. (b) Optical microscope image of the fabricated metasurface (c) Field emission scanning electron microscope (FESEM) image showing the nanoantennas array and electrodes.
4.4.2 Parametric Resonance Enhanced SHG

The linear response of the metasurface was characterized by measuring its reflectance spectra with and without a dc electric field, respectively. A broadband light source was coupled into an optical fiber and transformed into a collimated beam. It was focused through a long working distance, near infrared (NIR) objective (20X, NA = 0.40) from Mitutoyu on to the sample. The reflected beam was collected by the same objective and directed towards the spectrometer (Horiba iHR 320) equipped with an InGaAs infrared detector. Both reflectance spectra overlap with each other and show a prominent resonance around 1540 nm (Fig. 4-5 a). Evidently, dc field alone doesn’t affect the resonant property of the meta-atom.

Figure 4-5 Experimental demonstration of parametrically enhanced SHG. (a) Measured linear reflectance of white light source at dc voltage of 0 V and 30 V. They show the same resonance properties. (b) Comparison between parametrically enhanced SHG (asterisks) and conventional EFISH (diamonds), where a gain of 28 dB was achieved at 37 V. The fitting using equation (2) (red dotted line) agrees well with experiments. EFISH signals were collected under conditions where both the antennas array and the polarization of light were rotated by 90 degrees, so that the
same resonance enhancement effect was maintained while the effective dynamic modulation is reduced because $\chi^{(3)\text{yyyy}}$ ~ $1/3 \chi^{(3)\text{yxxy}}$. (c) Comparison between SHG signal collected when meta-atoms are on resonance at 1530 nm (asterisks) and off resonance at 1400 nm (diamonds). SHG of off-resonance was magnified by 100 for better illustration. (d) On/off enhancement factors at different wavelengths measured at fixed dc voltage of 30 V.

To characterize its nonlinear property, we first compared the parametrically enhanced SHG with conventional EFISH. Two sets of samples were fabricated: one with the designed parameter ($l_x = 600$ nm and $l_y = 650$ nm), the other was rotated by 90° ($l_x = 650$ nm and $l_y = 600$ nm), so that the same resonance enhanced local field effect can be kept the same by rotating the polarization of light to $x$ direction, but the effective parametric modulation will be different because $\chi^{(3)\text{yyyy}} = 3\chi^{(3)\text{yxxy}}$ \(^97\). The optical pump field was tuned to the resonant wavelength (1530 nm) of the meta-atom to leverage the field enhancement effect, creating strong parametric modulation. The dc electric field was aligned to the $y$ direction and increased gradually. SHG signals from both samples at different control voltages were collected using a spectrometer and scaled to the input optical pump power squared. Due the lack of inversion symmetry in $\alpha$-Si, only weak surface symmetry breaking induced SHG was detected, and it has been subtracted in all the following analysis. We extracted the gain as defined by $10\log((\chi^{(3)\text{yxxy}} / \chi^{(3)\text{yyyy}})^2 (I_{\text{SHG}}/I_{\text{EFISH}}))$ from Fig. 4-5 b and obtained a maximum gain of 28 dB at 37V. The fitting using equation (4-24) gives a good prediction on the growth of parametrically enhanced SHG.

Then we compare the SHG growth trend under different parametric modulation strength. The parametric modulation strength is proportional to $J_1(\Delta\phi) / J_0(\Delta\phi)$, indicating that a larger $\Delta\phi$ leads to stronger modulation. As shown by the simulation results in Fig. 4-2 c, when $\Delta\phi$ is small, it increases linearly with the modulation intensity, which is proportional to the dc control voltage. In addition, $\frac{J_1^2(\Delta\phi)}{J_0^2(\Delta\phi)} \approx \left(\frac{1}{2}\Delta\phi\right)^2$ under the small $\Delta\phi$ condition. Therefore, when the parametric modulation is weak, SHG intensity exhibits a quadratic dependence on dc control voltage, which is the case when the optical pump field is off-resonance (Fig. 4-5 c, red diamonds). However, when optical pump field is at the same wavelength as the resonance of meta-atoms, effective parametric resonance will be excited. Taking the whole equation (4-22) into account, SHG will have a super-quadratic dependence on dc control voltage.
(Fig. 4-5 c, blue asterisks). It also shows a maximum on/off enhancement factor of 15000 (\( I_{\text{SHG}} (37 \, \text{V}) - I_{\text{SHG}}(0 \, \text{V})/I_{\text{SHG}}(0 \, \text{V}) \)), which is three orders of magnitude greater compared with other EFISH nanophotonic devices\(^{194-196}\). On/off enhancement factors at different pump wavelengths were plotted in Fig. 4-5 d, confirming the importance of parametric resonance in boosting the SHG efficiency.

We estimated the effective second order sheet susceptibility \( \chi_{yyy}^{(2)} \) at 37 V using a z-cut quartz substrate as reference\(^{198-201}\). The nonlinear metasurface is placed on top of gold film and behaves as a sheet of nonlinear dipoles emitting fields at double frequencies into the free space. According to reference\(^{199}\), the second harmonic radiation field \( E(2\omega) \) driven by a sheet polarization \( P_{\text{dynamic}} \) located on top of a centrosymmetric substrate is:

\[
\vec{E}(2\omega) = i \left( \frac{\omega}{\varepsilon_0 c} \right) L \vec{P}_{\text{dynamic}} e^{i(2k_0 z - 2\omega t)}
\]

\[
\vec{P}_{\text{dynamic}} = \varepsilon_0 \chi_{\text{dynamic}}^{(2)} I^2 \left[ E(\omega) \right]^2 \hat{y}
\]

\[
\vec{E}(2\omega) = i \left( \frac{8\omega \chi_{\text{dynamic}}^{(2)}}{c} \right) \left[ E(\omega) \right]^2 e^{i(2k_0 z - 2\omega t)} \hat{y}
\]

where \( L \) is the Fresnel transmission factor and is equal to \( \frac{2k_0}{k_0 + k_{\text{Au}}} \approx 2 \) for normal incidence onto the gold substrate; \( k_0 \) is the free-space wavevector and \( E(\omega) \) is the amplitude of the fundamental excitation field.

Optical intensities are related to electric field amplitude by
\[
I(\omega) = 2\varepsilon_0 n c \left[ E(\omega) \right]^2.
\]
The second harmonic radiation intensity normalized by the square of the fundamental intensity is

\[
\frac{I(2\omega)}{I^2(\omega)} = \frac{32\omega^2}{\varepsilon_0 c^3} \left| \chi_{\text{dynamic}}^{(2)} \right|^2
\]

Next, we calculate the second harmonic intensity generated from \( \alpha \)-quartz under the same illumination conditions. The reflected SHG field in air is given by\(^{198,202}\),
\[ E_{\text{quartz}}(2\omega) = \frac{P_{\text{quartz}}^{(2)}(2\omega)}{2\varepsilon_0 n(n+1)} \]  
(4-25)

\[ P_{\text{quartz}}^{(2)} = \varepsilon_0 \chi_{\text{quartz}}^{(2)} [E(\omega)]^2 \]

where \( n \) is the refractive index of \( \alpha \)-quartz and \( P_{\text{dynamic}}^{(2)} \) is the nonlinear polarization. We then obtained the SHG intensity of \( \alpha \)-quartz normalized by the fundamental intensity.

\[ \frac{I_{\text{quartz}}(2\omega)}{I_{\text{quartz}}(\omega)} = \frac{|\chi_{\text{quartz}}^{(2)}|^2}{8\varepsilon_0 c n^2 (1+n)} \]  
(4-26)

\[
\frac{\chi_{\text{dynamic}}}{\chi_{\text{quartz}}^{(2)}} = \frac{1}{16k_0 n(1+n)} \frac{\sqrt{I(2\omega)/I(\omega)}}{\sqrt{I_{\text{quartz}}(2\omega)/I_{\text{quartz}}(\omega)}} \\
= \frac{1}{16} \frac{2\pi}{1.53 \times 10^{-6}} \frac{\sqrt{65535}}{150} = 3.735 \times 10^{-7} [m] \]  
(4-27)

\[
\chi_{\text{dynamic}} = 3.735 \times 10^{-7} \times \chi_{\text{quartz}}^{(2)} \\
= 3.735 \times 10^{-7} [m] \times 0.8 \times 10^{-12} [m/V] = 2.988 \times 10^{-19} [m^2/V] 
\]

Considering the filling factor of meta-atoms array of 0.156, the effective second order nonlinear susceptibility is

\[
\chi_{\text{dynamic}}^{(2)} = \frac{\chi_{\text{dynamic}}}{t \times \text{filling factor}} = \frac{2.988 \times 10^{-19} [m^2/V]}{3.5 \times 10^{-17} [m] \times 0.156} = 5.472 \times 10^{-12} [m/V] \]  
(4-28)

We can also calculate the conversion efficiency using eq. (22) and (26):

\[
\frac{I(2\omega)}{I(\omega)} = \frac{32\omega^2}{\varepsilon_0 c^3} |\chi_{\text{dynamic}}|^2 = 6\% [W^{-1}] \]  
(4-29)

The large \( \chi_{\text{yyy}}^{(2)} \) around \( 2.988 \times 10^{-19} \) m²/V is comparable to 2D materials\(^{203}\) with strong nonlinearity.
4.4.3 Electrical Tunable SHG

Fast electrical switching of SHG was also verified by measuring the time-resolved trace of SHG signal modulated by a square-wave voltage signal. We applied short pulses with 5V amplitudes across the IDT electrodes at 1MHz repetition frequency. The SHG signal was detected in a time-resolved measurement, which precisely follows the trace of the voltage pulses (Fig. 4-6). The rising and trailing edges were fitted to give a time constant of 5 ns which is limited by the edge time of the function generator (Fig. 4-7 a). The switching time of SHG is theoretically limited by the response time of nonlinear Kerr effect convoluted with the lifetime of the resonant meta-atom, which is in fact shorter than the pulse width of optical pump (~ 200fs). Therefore, our device can respond to the fastest electrical tuning method whose response time is in the range of picosecond to nanosecond.

![Diagram of experimental setup](image)

Figure 4-6 Time - resolved measurement setup. A modulated square wave voltage signal was applied on the sample with a function generator (Angilent 33220). The 1 MHz voltage signal changes between 0 V and 5 V with a duty cycle of 50%. In order to separate SHG from fundamental optical field and THG, the output signal passed through a transmission grating. An optical fiber was placed in the position for SHG ($\lambda = 765$ nm) collection and then directed the SHG towards a single photon avalanche detector (SPAD). The sync signal of the function generator was used as a start trigger, and the SPAD output was used as a stop in the photon counting module (Picoharp 300, Picoquant). The accumulated histogram of the electrical modulated SHG was obtained with accumulation time 300 s.

We also characterize the fundamental pump power dependence of SHG at different control voltage. As expected, the surface SHG intensity measured at 0 V scales quadratically with the power of the fundamental pump light. However, as the control voltage increased to 10V and 30V, it exhibited a sub-
quadratic power dependence (Fig. 4-7 b). This is due to the screening effect of the effective dc electric field experienced by the meta-atom. The effective band gap of $\alpha$-Si is $\sim 1.7$ eV, and there are exponential band tails with large density of states even below 1.6 eV due to the amorphous nature of silicon \textsuperscript{204}. Therefore, a large amount of carrier is generated through the absorption of enhanced SHG ($\sim 1.6$ eV) photons when biased by a dc voltage. These free carriers will shield the dc electric field imposed upon the meta-atoms, thus reducing the effective electric field. The shielding effect is stronger at higher dc voltage, which explains the observation that the SHG power dependence decreases as the control dc voltage increases. This non-quadratic fundamental power dependence is also universally observed in many EFISH demonstrations on semiconductor materials, in which an interfacial electric field induced by free carrier can modify the built-in electric field of the system \textsuperscript{205}. 

Figure 4-7 (a) Time-resolved trace of SHG (bottom panel) modulated by a square-wave electrical signal (top panel) at 1 MHz repetition rate. The SHG signal precisely follows the shape of the voltage pulses, indicating a 5 ns response time (extracted from the fittings of the rising and falling edges of SHG signal) only limited by the edge time of the function generator. (b) The fundamental pump power dependence at different dc voltages. The dashed lines are least-square fits to experimental data (asterisks). As the dc voltage increases, the measured SHG signal shows a decrease in the power dependence on the fundamental pump power. (c) Fourth order harmonic generation induced by the dc electric field.

More interestingly, the 4th order harmonic generation was also observed at a low pump intensity of 0.8 GW/cm² thanks to the strong parametric oscillation (Fig. 4-7 c), even though our device was not designed for high-order harmonic generation (HHG). Besides, the required pump intensity is two to three
orders of magnitude lower than a recent demonstration on HHG generation on dielectric metasurfaces\textsuperscript{206}. Therefore, our time-varying metasurface can be potentially used for enhancing and tuning high-order harmonic generations.

We also studied the polarization properties of the parametrically excited SHG. As expected, the surface symmetry breaking induced SHG has the same polarization state as that of the pump light (Fig. 4-8 a). With dc electric field, the polarization of SHG emission is predominantly along the direction of the dc electric field, whether the input polarization is parallel or perpendicular to the direction of the dc electric field (Fig. 4-8 b). This is because the fact that two nonzero effective second order susceptibilities $\chi^{(2)}_{yyy} = \chi^{(3)}_{yyyy} E_y^{dc}$ and $\chi^{(2)}_{yxx} = \chi^{(3)}_{yxxy} E_y^{dc}$ are generated under dc biasing. Regardless of the polarization of the optical field, the SHG signal is always in y polarization – that is along the dc field direction.

Figure 4-8 (a) Surface SHG polarization under zero dc voltage. It has the same polarization as that of the incident pump light. (b) Parametrically enhanced SHG under dc voltage of 30 V. Its polarization is aligned with the dc electric field, no matter what polarization state of the incident pump light is.
4.5 Conclusion

We demonstrated parametrically enhanced and electrically tunable SHG on a time-varying metasurface, which provides a compact and CMOS compatible platform for tuning and boosting nonlinear generations. It paves exciting ways for constructing electrically tunable nonlinear optical devices, such as optical parametric sources, ultrafast optical switches and modulators, for applications in optical communication, imaging, sensing and laser technology.
Chapter 5

Controlling Free-space Light with Guided-wave-driven Metasurfaces

Metasurfaces with unparalleled controllability of light have shown great potential to revolutionize conventional optics. However, they mainly work with free-space light input, which makes it difficult to fully integrate them on-chip with light sources. On the other hand, integrated photonics enables packing optical components densely on a chip, but it only provides limited free-space light controllability. In this chapter, I introduced a new type of metasurfaces that are driven directly by guided waves. By dressing those metasurfaces on top of waveguides, we molded the guided waves into any desired free-space modes to achieve complex free-space functions, such as out-of-plane beam deflection and focusing on a photonic integrated chip. Our study shows a viable route towards complete control of light across integrated photonics and free-space platforms, and a pave a new exciting way for creating multifunctional photonic integrated devices with agile access to free space which could enable a plethora of applications in communications, remote sensing, displays, etc.

5.1 Background and Motivation

With the fast-growing demands for big data, electronic chips and interconnects with insufficient bandwidth can hardly meet the requirements on data transmission speed and energy efficiency of future computing and storage systems. Wiring light on a chip like electronic circuits, integrated photonics provides a promising long-term solution \textsuperscript{207,208}. A photonic integrated circuit (PIC) combines many light-controlling components into a single chip, with the ultimate aim of creating miniature optical circuits similar to CMOS (complementary metal oxide semiconductor).
chips that have revolutionized the electronics industry. It offers great advantages in terms of speed, bandwidth, reliability, scalability, power consumption, and etc. In order to fully exploit the benefits of PICs in free-space applications, it is crucial to have an interface that can flexibly control light when it converts between guided and free-space modes. However, two conventional coupling techniques – edge couplers and surface gratings – have limited functionalities and lack complete control over light. Although arrays of gratings can achieve more advanced functions, such as off-chip beam steering, focusing, and holographic image construction, they have large footprints and suffer from loss due to the existence of high-order diffractions. Subwavelength gratings have compact footprints but they mainly works in controlling the guided waves rather than the manipulation of light across PICs and free space. Recently, optical nanoantennas are integrated on top of waveguides, which provides a new way for interfacing guided and free-space optical modes and adds more functionalities to PICs. Nevertheless, most of them rely on a singular property of nanoantennas to achieve a specific purpose, such as photo-detection or modulation based on plasmonic field enhancement and directional routing from spin–orbit coupling. A unified approach leveraging the collective free-space functions of nanoantennas on PICs has not been demonstrated.

Here, we combined synergically two powerful, complimentary technologies: integrated photonics and metasurfaces, and developed a hybrid architecture where metasurfaces are directly driven by guided waves to realize complex free-space functions. We placed subwavelength-sized meta-atoms on top of photonic integrated components (Fig. 5-1 a). In contrast to existing metasurfaces that operate with both input and output light in free space, our integrated metasurface bridges guided waves inside a waveguide with free-space ones. Through it the guided light is tapered into free space and molded into desired light fields. The subwavelength spacing of the meta-atoms eliminates diffraction loss and also allows denser on-chip integration. Meanwhile, multiple metasurfaces can be connected via waveguides to achieve different free-space functions.
simultaneously. Although phase-gradient plasmonic metasurfaces have been used for guided mode conversion \(^{21,219}\), their limited phase controllability (~\(\pi\)) makes it inadequate for wavefront shaping. Our design overcomes this limitation by utilizing metal-dielectric-metal nanoantennas to expand the phase tuning range to over \(2\pi\). Such guided-wave-driven metasurfaces go beyond the existing technology of leaky-wave antennas made of periodic structures \(^{220}\) and extend the functionalities of microwave waveguide-fed metasurface \(^{221}\) to new spectral regions.

The developed technology will potentially be a huge step towards full control of light across integrated photonics and free-space platforms, and will pave new exciting ways for building multifunctional PIC devices with flexible access to free space as well as guided-wave-driven metasurfaces with full on-chip integration capability. It could enable a plethora of applications in optical communications, optical remote sensing (e.g. light detection and ranging (LiDAR) \(^{27}\)), free-space optical interconnects (FSOIs) \(^{222}\), and displays \(^{223}\). In addition, a library of those functional hybrid components can be established for reusing and creating consistency across various devices or systems.
Figure 5-1 The working principle of guided-wave-driven metasurfaces. (a) A schematic of a guided-wave-driven metasurface. The phase of the extracted light from a guided wave by each meta-atom can be tuned individually. An array of meta-atoms on the waveguide work collaboratively to form certain wave fronts and fulfill different functions, such as beam deflection and focusing. (b) An illustration of the wave front formation of the extracted wave. The total phase shift of the extracted wave at coordinate $x$ is contributed from two parts: the phase accumulation $\beta x$ from the guided wave propagation and the abrupt phase change $\Delta \phi(x)$ induced by the meta-atom. As a result, the phase of the extracted wave can be expressed as $\phi_0 + \beta x + \Delta \phi(x)$, where $\phi_0$ is the initial phase of the incidence.

5.2 Design

In contrast to the free-space metasurfaces where the spatial phase profile is solely provided by the meta-atoms, the total phase shift of the extracted wave from our guided-wave-driven metasurface is contributed from two parts: (i) the phase accumulation from the propagation of the guided wave $\beta x$ (where $\beta$ is the propagation constant of the guided mode and we assumed that the waveguide lays straight along the $x$ direction) and (ii) the abrupt and spatially variant phase shift $\Delta \phi(x)$ induced by each meta-atom at coordinate $x$ (Fig. 5-1 b). The essence of such metasurfaces is to use subwavelength-sized meta-atoms to form spatially varying optical response, which extracts
and molds guided waves into any desired free space optical modes. This distinguishes such metasurfaces from leaky-wave antennas which typically do not have spatial-variant electromagnetic responses. As a result, the phase distribution of the extracted wave along the $x$ direction can be expressed as

$$\phi(x) = \beta x + \Delta\phi(x)$$  \hspace{1cm} (5-1)

To achieve complete control of the wavefront, at least $2\pi$ phase shift range induced by meta-atoms is required since we do not have much control over the propagation induced phase once the waveguide design is fixed. Placing the meta-atoms on the waveguide, it is challenging to use a mirror resonance or a geometrical phase in order to achieve $2\pi$ phase shift. Therefore, here we designed a metal-dielectric-metal sandwiched nano-bar antenna (Fig. 5-2 a) – which supports two resonant modes and is evanescently coupled with the guided waves inside the waveguide – as the meta-atom. The fundamental transverse electric mode (TE$_{00}$) in a dielectric rectangular waveguide (Fig. 5-2 b) was used to excite resonant modes of meta-atoms as its field distribution has a good spatial overlap with the electric dipolar mode in a nano-bar antenna. The thickness, width and length of the sandwiched nanoantennas were carefully chosen so that when the bottom gold cuboid is excited by the evanescent tail of a guided wave and induced an electric dipole, an antiparallel one can be induced in the top cuboids, therefore, an effective magnetic dipolar resonance is excited (Fig. 5-2 b). Simultaneously, the electric dipoles induced in the top and bottom gold cuboids do not completely cancel out, resulting in a net electric dipolar resonance (Fig. 5-2 b). The magnetic resonance combined with the electric one creates a directional radiation that extracts the guided wave to free space, and it also provides an abrupt phase shift range $\sim 2\pi$ to the extracted wave (Fig. 5-2 c). This abrupt phase shift can be tuned by varying the geometrical parameters of the meta-atoms. In addition, by controlling the amount of spatial mode overlap between the antenna mode and the guided mode, we are able to flexibly adjust amplitude of the extracted wave. With the light
extraction and control capabilities of meta-atoms, various free-space optical functions can be realized by distributing them strategically along the waveguide.

Figure 5-2 (a) A schematic of a metal/dielectric/metal sandwich-structured meta-atom on top of a photonic integrated waveguide. The bottom left inset shows the simulated electric field distribution of the TE_{00} guided mode propagating inside the waveguide. The bottom right inset is the simulated magnetic field distribution of the sandwich-structured nanoantenna, which indicates an effective magnetic dipole. (b) Simulated field distribution of silicon waveguide, Au/SiO_{2}/Au sandwiched nanoantennas (l_x = 150nm, l_y = 280nm) and the integrated systems. The antenna exhibits electric dipole (ED) and magnetic dipole (MD) eigenmodes at 1400nm and 1820nm, respectively. At 1510 nm, the integrated system exhibits electric field and magnetic field distributions characteristic of the ED and MD, validating that the overlap of two resonances create the phase shift range over 2π. (c) Calculated scattering phase induced by Au/SiO_{2}/Au sandwiched nanoantennas (l_x = 150nm, l_y = 280nm) at different wavelengths, showing a phase shift range greater than 2π. (d) A pseudocolor map of the simulated abrupt phase shifts in a parameter space spanned by the meta-atom width (l_x) and length (l_y). A thickness of 30 nm was used for each layer. The meta-atom was placed on top of a silicon ridge waveguide (height 220 nm). The three white stars indicate the meta-atom designs covering 2π phase range with an even interval. We also ensured that the extracted waves from the chosen meta-atoms have roughly the same amplitude of $1.5 \times 10^5$ V/m. (e) Simulated electric field distribution ($E_y$) of the extracted waves from the three selected meta-atoms, showing abrupt phase shifts of 2π/3, 0, and −2π/3, respectively.

In order to show the capability of the guided-wave-driven metasurfaces, we designed off-chip beam deflection and light focusing directly from a photonic integrated waveguide. Numerical simulations were carried out using a commercially available finite element method (FEM) solver package – COMSOL Multiphysics. Third-order finite elements and at least 10 mesh steps per
wavelength were used to ensure the accuracy of the calculated results. We used an eigenmode solver to find the $\text{TE}_{00}$ mode of the silicon waveguide as well as its modal index at 1550 nm wavelength. Then this modal index was used in the model to further calculate the phase and amplitude of the extracted light by monitoring the field at a few wavelengths over the waveguide. The antenna is centered on top of the waveguide. We swept the geometrical parameters of the meta-atoms and monitored the phase and amplitude of the scattered wave into free space, and obtained the phase and amplitude maps/contours as shown in Fig. 5-2 d. The trapezoidal shape of meta-atoms resulted from our nanofabrication was also taken into account in our model to get accurate design parameters. In order to simulate the beam steering, a full device model that consists of an array of meta-atoms placed on top of Si waveguide was established. The meta-atoms were distributed along the waveguide so that they formed a linear phase gradient (Fig. 5-2 e). As shown in Fig. 5-3 a, light is extracted by the phase-gradient metasurfaces into free space and forms well-aligned wavefront. In a similar fashion, an array of meta-atoms fulfilling the spatial phase distribution of a lens were placed on top of Si waveguide to simulate the light focusing effect (Fig. 5-3 b).
Figure 5-3 (a) Simulated electric field ($E_y$) distribution of the guided-wave-driven metasurface (20 sets of supercells, 60 nanoantennas) for beam deflection. (b) Simulated normalized electric field ($|E|$) distribution of the guided-wave-driven metasurface (65 nanoantennas) for focusing.

5.3 Experiment

5.3.1 Fabrication

The fabrication process is shown in Fig. 5-4. Samples were fabricated on a commercially available silicon-on-insulator wafer with 220-nm-thick (for beam steering experiments) and 500-nm-thick (for light focusing experiments) Si device layer and 3-μm buried silicon dioxide. The wafer was cleaned by sonication in acetone and IPA for 3 minutes, respectively. Alignment marker was defined by electron beam lithography with 100 kV beam (Vistec EBPG5200) followed by evaporation of 50 nm Au with a 5-nm-thick Ti adhesion layer (Kurt J. Lesker Lab-18) and lift-off.
Then negative resist Fox-16 (Dow Corning Corp.) was spin-coated and prebaked at 100 °C for 4 minutes. The waveguide pattern was written followed by development in CD-26 developer (MicroChem) for 25 minutes to reduce proximity effect. Chlorine-based inductively coupled reactive ion etching (ICP-RIE) was used to etch crystalline Si with FOX-16 resist as mask (Plasma-Therm Versalock 700). The sample was immersed in buffered oxide etchant for 20 seconds followed by water rinse to remove the remaining mask. ZEP 520A (Zeon) was spin-coated on the sample and soft-baked at 180 °C for 3 minutes. A second-step electron beam lithography was conducted to define the metasurface layer on top of the waveguide with precise alignment. The exposed sample was developed in N-amyl-acetate for 3 minutes followed by MIBK:IPA immersion for 1 minute. Au/SiO2/Au films were subsequently deposited using an electron beam evaporation system (Semicore). The pattern was then lifted off in 1165 remover (MicroChem) at 85 °C in water bath for 2 hours. The sample was finally diced along the input port of waveguide for measurement.

Figure 5-4 Process flow of making guided-wave-driven metasurfaces.
5.3.2 Off-Chip Beam Deflection

According to equation (5-1), the linear momentum of extracted light along \( x \) direction is \( k_x = \beta + \partial \Delta \phi(x)/\partial x \). If \( \partial \Delta \phi(x)/\partial x \) is a constant, the extracted beam has a well-defined angle \( \theta \) given by \( \theta = \sin^{-1}(k_x/k_0) \), where \( k_0 \) is the free-space wave number. In contrast to grating couplers on PICs, our metasurface approach introduces abrupt and large phase shifts with a subwavelength spacing, which eliminates high-order diffractions and offers a much large beam deflection angle range.

We used Au/SiO\(_2\)/Au sandwich-structured nanoantennas as the meta-atoms for beam deflection. The meta-atoms are periodically distributed on a silicon waveguide to provide a phase gradient \( \partial \Delta \phi(x)/\partial x = -2\pi/\Lambda \), where \( \Lambda \) is the length of a supercell which consists of three meta-atoms with abrupt phase shifts \(-2\pi/3, 0, \text{ and } 2\pi/3\). Therefore, the output angle of the extracted beam is \( \theta = \sin^{-1} \left( 1/k_0 \left( \beta - 2\pi/\Lambda \right) \right) \) (Fig. 5-3 a).

Different lengths of supercells were chosen to demonstrate flexible control of the beam deflection angles. The propagation constant \( \beta \) was numerically calculated for the fundamental TE modes at different wavelengths. Fourier-space imaging system (Fig. 5-5 b) was employed to measure the scattering angles. A free-space laser beam output from a Ti:Sapphire laser pumped optical parametric oscillator (OPO) was coupled into a commercially available tapered lensed single-mode fiber. The focused laser beam from the tapered fiber was coupled into the input port of our fabricated ridge waveguide sample in end-fire manner by using a three dimensional (3D) translational stage. The coupled-in light propagated through a triangle taper linking the input port and the single-mode waveguide, during which the high-order modes vanished, and only fundamental transverse electrical mode survived. The light scattered into free space by metasurfaces on top of the single-mode waveguide was collected by an objective (N.A. = 0.95) and then transmitted through a tube lens. Part of the light was reflected by a beamsplitter for real-space imaging. And the light transmitted through the beamsplitter was focused by a Bertrand lens to form
Fourier-space image. The laser wavelength was tuned using the OPO to acquire wavelength-dependent beam deflection angles. In addition, Fourier-space images were taken by coupling 1550nm laser beam into samples with different supercell periods. After that, the scattering angles were extracted from the Fourier-space images calibrated by a ruled reflective grating (grooves density of 600/mm).

We experimentally measured the output angles with different wavelengths and supercell periods, respectively, and the results agree well with those from our theoretical calculations (Fig. 5-5 c and d). The slight discrepancy originates from the fabrication error. The line-shaped intensity profile in Fourier space reveals the in-plane wavevector of the extracted light, where $k_x$ is determined by the metasurface and $k_y$ spans the whole Fourier plane because no phase modulation is applied in y direction. The divergence of the steering angle, which is depicted by the width of the line, is inversely related to the length of the metasurface region. The bright ends of the lines (near the cut-off lines in Fourier space images limited by the numerical aperture (NA = 0.95) of the objective) are originated from the internal reflection in the objective.
Figure 5-5 Demonstration of off-chip beam deflection with guided-wave-driven metasurfaces. (a) Field emission scanning electron microscope (FESEM) images of a guided-wave-driven metasurface on a silicon waveguide (220 nm thick and 600 nm wide). Each supercell consists of three meta-atoms as depicted in Fig. 2B. (b) A schematic of the experimental setup for the off-chip beam steering and focusing measurements. A free-space laser beam emitted from a Ti:Sapphire laser pumped OPO was coupled into a tapered lensed single-mode fiber and then to the input port of the sample. The extracted light in free space was collected by an objective (NA = 0.95) and then transmitted through a tube lens. The light was partially reflected by a non-polarizing beam splitter for real-space imaging. And the rest light was transmitted through the beam splitter and was focused by a Bertrand lens to form Fourier-space images. (c) Output beam angle versus the incident guided wave wavelength with supercell size $\Lambda = 575$ nm measured by our Fourier-space imaging system (Fig. S5). The blue dots and the red dashed line depicts the experimentally measured and the simulated data, respectively. Three typical Fourier-space images of the extracted free-space light corresponding to the circled data points are shown on the right. The horizontal and vertical axes represent $k_x$ and $k_y$ respectively. An objective with numerical aperture (NA) of 0.95 was used in the measurements. (d) Output beam angle versus the supercell size at 1550 nm wavelength. The blue dots and the red dashed line depict the experimentally measured and the simulated data, respectively. Similar to (B), three typical Fourier-space images are shown on the right.
The efficiency of the device was estimated using full-wave numerical simulations. Extraction efficiency was calculated by dividing the surface integrated power flow from a plane above the waveguide to the input power. And ohmic loss was estimated by dividing the volume integration of the power dissipation in nanoantennas to the input power. For beam deflection, the power loss due to back reflection \( R \) is small and most of the loss is induced by the metallic antenna absorption. From the simulation, we extracted the up-extraction efficiency per supercell \( \eta_e \approx 0.1\% \) and the total dissipation per supercell \( \eta_t \approx 1.1\% \). According to the following equation:

\[
\text{efficiency} = (1-R) \frac{\eta_e}{\eta_t} \left(1-e^{-\eta_t l} \right)
\]

where \( l \) is the number of supercells. As a rough estimation, the maximum up-extraction efficiency can be approximated as \( \frac{\eta_e}{\eta_t} \approx 9\% \) (Fig. 5-6 c). However, this can be alleviated by using pure dielectric SiO\(_2\)/Si/SiO\(_2\) sandwiched nanoantennas (Fig. 5-6 a) which have no material loss in telecommunication wavelengths range. The efficiency can be controlled by the number of supercells integrated on the waveguides, and a maximum value as high as 80\% can be expected with sufficient numbers of supercells (Fig. 5-6 c).
Figure 5-6 (a) A pseudo-color map of the simulated phase shifts generated by SiO$_2$/Si/SiO$_2$ nanoantennas with different lengths ($l_x$) and widths ($l_y$). Three meta-atom designs (marked by the red stars) with a constant phase shift difference of $2\pi/3$ were selected to construct the supercell used in simulation of beam deflector shown in (b). (b) Simulated electric field ($E_y$) distribution of the guided-wave-driven metasurface beam deflector using designs of (a) (20 sets of supercells, $\Lambda = 680$ nm). (c) Calculated up-extraction efficiency for Au/SiO$_2$/Au nanoantennas (left) and for SiO$_2$/Si/SiO$_2$ nanoantennas (right).
5.3.3 Off-Chip Beam Focusing

In addition, spatially arranging the meta-atoms along a waveguide to fulfill a lens phase function \( \phi(x) = -k_0 \sqrt{x^2 + f^2} \), we can focus the wave in free space with a designated focal length \( f \). Therefore, considering Eq. (5-1) the abrupt phase shifts provided by the meta-atoms should be

\[
\Delta \phi(x) = -k_0 \sqrt{x^2 + f^2} - \beta x
\]  

(5-3)

As a proof of concept, we simulated such a metalens on a silicon waveguide with a focal length \( f = 5 \) µm (we chose a short focal length in order to reduce the demand for computational resources) at 1550 nm (Fig. 5-7). Evidently, light is extracted and focused into free space by the metalens. We designed and fabricated a larger guided-wave-driven metalens with a focal length of 225 µm. The intensity distribution at different heights above the waveguide was measured and reconstructed in the \( xz \) plane, which shows clear focusing effect matching our simulation results.

In addition, the light focusing effect was also observed at different wavelengths both by simulations and experiments (Fig. 5-8). According to equation (5-3), as wavelengths increase, both \( k_0 (2\pi/\lambda) \) and \( \beta \) will decrease, so that the designed phase shift at 1550 nm is smaller than the required phase shift at longer wavelengths. The opposite argument is true when the wavelength decreases. Therefore, the focal point shifts towards the input port as the wavelength increases to compensate for the phase difference.
Figure 5-7 **Demonstration of off-chip light focusing with a guided-wave-driven metalens.** (a) Simulated electric field distribution above a guided-wave-driven metalens on a silicon waveguide (500 nm thick and 1.5 µm wide). The extracted light converged at the designed focal point (5 µm above the waveguide) at 1550 nm wavelength. (b) Experimentally measured intensity profile of the focusing effect of a fabricated device. The inset shows an FESEM image of the metasurface region. The designed focal length is 225 µm.
Figure 5-8 (a) Simulated and (b) experimentally measured guided-wave-driven off-chip focusing at different wavelengths. The focal distance of simulated device is 3 µm, and the focal distance of the sample is 225 µm.
5.4 Conclusion

The guided-wave-driven metasurface, consisting of subwavelength-spaced meta-atoms placed on top of photonic integrated waveguides, provides a highly versatile and compact platform for bridging the gap between guided waves in PICs and free-space waves. The developed technology not only empowers the photonic integrated devices with agile free-space light controllability in the subwavelength scale, but also enables metasurfaces to be directly driven by guided waves which makes possible a denser and higher level of on-chip integration.

We have experimentally demonstrated off-chip beam deflection and focusing using the guided-wave-driven metasurfaces on silicon waveguides, which could enable a wide spectrum of applications ranging from optical communications to LiDAR, as well as miniaturized display technology for virtual reality (VR) and augmented reality (AR) devices. In addition, due to reciprocity, free-space modes can be selectively coupled into the metasurface-dressed waveguides. The metasurface region can be engineered to couple light with a tilted or even distorted wave front into a waveguide, which is especially useful for optical sensing and detections. Moreover, dynamic control of the coupling between guided modes and free-space ones can be realized by incorporating tunable elements \(^{34,47}\), which further empowers the PICs with the capability of tuning the optical functionalities dynamically.

This chapter is adapted from the publication “Xuexue Guo, et. al. "Molding free-space light with guided wave driven metasurfaces." Science Advances In press, (2020)".
Chapter 6

Phase Gradient Metasurface Integrated Orbital Angular Momentum Microring Laser

Due to the asymmetry of phase gradient metasurfaces, the spatial inversion symmetry of the system is broken, leading to unidirectional beam deflection. Leveraging this unique property of metasurface in an active microring resonator, we break the degeneracy of clockwise- and counterclockwise-propagating whispering gallery modes, and hence realize direct orbital angular momentum (OAM) lasing on a chip. Our approach opens up a way to directly generate controlled OAM light source for applications in quantum information processing and optical communications.

6.1 Background and Motivation

Integrated photonics offers potential solution to the bottleneck in scaling and data transport for semiconductor microchips \(^{225,226}\) by integrating energy-efficient, high-bandwidth photonic devices to electronic chips. However, challenges still exist and hinder the large-scale photonic integration, such as device miniaturization, increasing operation bandwidth and stability, and reducing insertion loss and heat dissipation \(^{225,226}\). Thanks to the introduction of metasurface \(^{227–229}\), the properties of light can be manipulated efficiently on the wavelength scale, which provides a powerful toolkit in addressing some of the challenges in integrated photonics \(^{21}\). One of the key component in integrated photonic devices is light source, and a prominent class is based on microring \(^{230,231}\) lasers. Due to the high refractive index contrast to surrounding media, microring resonator supports high quality factor whispering gallery modes (WGMs) that circulates within the waveguide and inherently carries orbital angular momentum (OAM) \(^{232,233}\).
OAM – characterized by helical wavefront with a spatially dependent phase of $\exp(ilm\phi)$ – is a property of light like the frequency, polarization and phase, where $l$ is an integer known as the topological charge, and $\phi$ is the azimuthal angle with respect to the propagation direction. In contrast to its counterpart - spin angular momentum of light, OAM can assume infinite number of states $l\hbar$. This unique property makes it a excellent candidate for encoding information in quantum information and optical communication. In addition, the vanishing field in the vortex center enables the imaging beyond Rayleigh limit. And the helical wavefronts have been used to expand the capabilities of optical tweezers by creating toroidal torque-exerting traps.

Traditional methods in generating OAM light are based on combining the active component lasers and passive phase modulation components such as spatial light modulator, spiral phase plates, q-plate, and more recently homogeneous anisotropic media, microrings with angular grating and metasurface. Even though with compact passive component like microrings and metasurface, the overall device size is bulky, which prevents its wide application in integrated photonics. Recently, researchers have theoretically and experimentally demonstrated direct vortex laser based on molecular chromophore array, chiral nanoemitter array, photonic crystal with topological defect, intracavity mode selection and non-Hermitian optical system, which mark a giant advancement towards integratable and scalable OAM light source for on-chip applications.

Inspired by the spatial phase tunability of metasurface, we designed an OAM microring laser based on the asymmetric phase modulation of counter-propagating WGMs. It is known that the degenerate nature of clockwise (CW) and counter clockwise (CCW) WGMs cancels out the OAM of each other and the OAM order carried by WGM modes are normally very high. Therefore, it is difficult to extract laser radiations with controllable OAM orders. However, this degeneracy of scattered light is lifted as WGM modes interact with the metasurface that imposes a unidirectional $k_{ms}$ (Fig. 6-1 a). Leveraging the asymmetric coupling induced by the guided-wave-
driven metasurface, we have created a photonic integrated micro-ring OAM laser.

6.2 Design

The micro-ring resonator intrinsically supports two degenerate whispering gallery modes (WGMs) – a clockwise (CW) and a counter-clockwise (CCW) mode. These modes by themselves carry high-order OAM. But due to the inversion symmetry of the micro-ring, the OAMs of the CW and CCW modes have opposite signs and the net OAM is zero. In order to obtain controllable OAM emission, our metasurface accomplished three functions: (1) Extract light from the micro-ring without destroying the guided modes; (2) Break the degeneracy of the two WGM modes to get non-zero net OAM emission; (3) Control the topological charge of the OAM.

Due to the asymmetric coupling effect of the guided-wave-driven metasurface, only one of the two counter-propagating WGMs can couple to the free-space emission (Fig. 6-1 a), and therefore we are able to break the degeneracy of the WGMs and achieve a controllable OAM emission. As the degenerate WGMs interact with the metasurface on a micro-ring that introduces a unidirectional phase gradient $\partial \phi / \partial \varphi$ ($\varphi$ is the azimuth angle), the radiated light of CW and CCW mode will gain additional but opposite momenta. One radiation mode will gain too large $k$ to propagate in free space, while the other one can be successfully launched into free space with a well-defined OAM order.

Let us suppose we want the CCW mode to be extracted and form OAM emission in free space. The propagation constant of the $M^{th}$-order CCW WGM is given by $\beta_{CCW} = 2\pi n_m / \lambda = M / R$, where $n_m$ is the modal index and $R$ is the micro-ring radius. The guided-wave-driven metasurface is placed on the micro-ring so that it induces a phase gradient that is equivalent to a wave number $k_{ms} = -2\pi / \Lambda$, where the phase shift provided by the meta-atoms decreases linearly
along the CCW direction. The azimuthal phase dependence of OAM emission can be expressed as 
\[ \phi_{OAM}(\varphi) = l\varphi. \] Due to momentum conservation, the following condition should be satisfied

\[ l\varphi = \phi_{OAM} = \phi_{CCW} + \phi_{w} = \beta_{CCW}R\varphi - 2\pi R/\Lambda \] (6-1)

Assuming the total number of metasurface supercells on the micro-ring is \[ N = \frac{2\pi R}{\Lambda}, \] we can obtain from equation (6-1) a well-defined topological charge \[ l = M - N, \] which can be easily engineered either by tuning the order of the WGM mode or by placing different numbers of supercells on the micro-ring.

Figure 6-1 (a) Electric field distribution of the extracted light from a phase-gradient metasurface driven by forward- (top panel) and backward- (bottom panel) propagating guided waves. The metasurface consists of an array of meta-atoms that form a phase gradient \( \partial \Delta \varphi(x)/\partial x \) (which is along the -x direction in this example). The extracted light from a forward-propagating guided wave carries a transverse wavevector \( k_x = \)
where $\beta$ is the propagation constant of the guided wave. It is launched into free space with a well-defined angle $\theta = \sin^{-1}(k_x/k_0)$. In contrast, light extracted from the backward-propagating wave gains a transverse wavevector so large that it exceeds the maximum supportable wavenumber in free space, and therefore it bounds to the metasurface and eventually dies out due to ohmic loss from the materials. (b) A schematic of a micro-ring OAM laser enabled by the guided-wave-driven metasurface. Unidirectional phase modulation provided by the metasurface breaks the degeneracy of the CCW and CW WGMs inside the micro-ring resonator, leading to a selective OAM radiation.

We designed our OAM laser based on an InGaAsP/InP multi-quantum-well (MQW) micro-ring resonator. Four Au/Si/Au sandwich-structured meta-atoms covering $2\pi$ abrupt phase shift range (Fig. 6-2 a) were used to construct one metasurface supercell and patterned periodically on top of the micro-ring (Fig. 6-1 b). We showed using full-wave FEM eigen-mode simulations that the emitted light is radially polarized and exhibits the characteristics of OAM emission. With $M = 59$ and $N = 58$, the electric field $E_r$ forms a spiral pattern, and its phase changes by $2\pi$ upon one full circle around the center of the vortex, indicating $l = 1$ (Fig. 6-2 c, left panel). We also showed the simulation results of $l = 2$ with $M = 59$ and $N = 57$ (Fig. 6-2 c, right panel). The phase profile depicts a $4\pi$ winding around the center of the vortex.
Figure 6-2 (a) A pseudo-color map of the simulated abrupt phase shifts overlaid with amplitude contours generated by the Au/Si/Au meta-atoms with different lengths ($l_x$) and widths ($l_y$). Four meta-atom designs (marked by the yellow stars) with a constant phase shift difference of $\pi/2$ were selected to construct the metasurface supercell. The black dashed line is the contour of the extracted electric field amplitude of $2 \times 10^5$ V/m. (b) The simulated electric field distribution of the micro-ring resonator (diameter = 9 µm, width = 1.1 µm and height = 1.5 µm) with WGM order $M = 59$. A close-up view of one segment of the micro-ring shows a good spatial overlap between the waveguide mode and the meta-atoms. (c) Simulated electric field (radial component) and phase distribution of emitted wave with different numbers of metasurface supercells $N$. The azimuthal order of the WGM is $M = 59$ at the resonant wavelength of 1550 nm, and the number of supercells is $N = 58$ (top row) and $N = 57$ (bottom row). The resulting topological charge of the OAM radiation can be seen by the number of $2\pi$ phase evolution along the circumference, which is +1 (top row) and +2 (bottom row), respectively.

6.3 Experiment

We fabricated the micro-ring OAM laser (Fig. 6-3) using similar process introduced in chapter 5. It was fabricated on InGaAsP (500 nm, multi-quantum-well layer) / InP substrate. First
the micro-ring resonator and electron-beam markers were defined by electron beam lithography with FOX-16 negative resist. The resist acted as an etch mask in the BCl₃ based ICP-RIE process. Then the sample was immersed in buffered oxide etchant to remove the mask. A second-step electron beam lithography using ZEP 520A resist was performed with precise alignment to define the metasurface layer on top of the micro-ring resonator. A sequential electron beam evaporation was done to deposit Au/Si/Au films, followed by a standard lift-off process in 1165 remover at 85 °C in water bath for 2 hours.

Figure 6-3 Field-emission scanning electron microscope (FESEM) images of an OAM microring laser. The diameter of the microring is 9 µm and the width is 1.1 µm, and it consists of a 500 nm InGaAsP MQW layer and a 1 µm InP layer. A supercell of the metasurface consists of four Au/Si/Au meta-atoms, which provides the extracted wave with abrupt phase shifts from 0 to 2π. The total number of supercell on the micro-ring is N = 58.

Fig. 6-4 shows the experimental setup to measure the laser emission and characterize its OAM properties. The micro-ring was pumped by 900-nm femtosecond pulses (~140 fs) from a
Ti:Sapphire laser. The lasing emission was collected by the objective and then transmitted through the dichroic mirror and detected by a spectrometer (Horiba) and a far-field imaging system. With a flip mirror to switch the paths, the laser emission was either sent into the spectrometer/imaging system or the interferometry setup.

Figure 6-4 A schematic of the experimental setup for characterizing the OAM laser emission. A femtosecond pulsed pump laser (~140 fs, repetition rate 80 MHz) at 900 nm wavelength was reflected by a dichroic mirror and then focused by a Newport 20X objective (NA = 0.40) onto the micro-ring resonator. The lasing emission was collected by the same objective and then transmitted through the dichroic mirror to be detected by a spectrometer, a far-field imaging system and a Michelson interferometry setup. A flip mirror was used to switch the paths.

The spectra gradually transitioned from spontaneous emission (SE) to amplified spontaneous emission (ASE) and finally to lasing as the pump intensity increased (Fig. 6-5 a). We observed a kink at around 0.47 GW/cm² in the light-light curve, which indicates the transition from ASE to lasing. The spectral full width at half maximum (FWHM) is as narrow as 2 nm, indicating a quality factor around 800. Because the microring resonator posseses the rotational symmetry, the polarization of the emission is expected to carry the same polarization state as the light inside the ring cavity. In our microring resonator, the fundamental TE mode is designed to the lasing mode because it has a better spatial overlap with metasurface elements (Fig. 6-2 b). Therefore, the radiated OAM light is predominantly azimuthally polarized (Fig. 6-6), indicated by the
orthogonality between polarizer axis and the measured OAM intensity pattern after the polarizer.

Figure 6-5 (a) Light-light curve of the micro-ring laser (top row), which shows a lasing threshold of about 0.47 GW/cm² at 1555 nm wavelength. Three emission spectrum corresponding to different stages – photoluminescence, amplified spontaneous emission, and lasing – of the laser are shown from 2nd to the last row. (b) Far-field intensity distribution of the OAM laser radiation captured by an infrared camera (right panel), which matches well with the simulated one (left panel). Both figures show an annular shape. (c) and (d) Calculated (left panels) and measured (right panels) self-interference patterns of OAM laser radiation. The double-fork (F) and triple-fork (G) in the fringe patterns confirmed that the resulting OAM emission has a topological charge of +1 (F) and +2 (G), respectively.
Figure 6-6 Measured intensity distributions after a polarizer in the directions indicated by the white arrow, where a two-lobe intensity pattern is orthogonal to the polarizer axis.

The OAM characteristics were characterized by analyzing both the spatial intensity profile of the emission using a near-infrared camera and its self-interference pattern using Michelson interferometry (Fig. 6-4). The laser emission was split into two beams by a pellicle beam splitter, and then recombined with an off-center beam overlap to form an interference pattern recorded by an infrared camera. A delay line was used in order to balance the optical path lengths of the two arms. We observed the intensity of lasing emission spatially distributed in a doughnut shape with a dark core in the center (Fig. 6-5 b), which is due to the phase singularity at the beam axis where the phase becomes discontinuous. The presence of the OAM was also validated by the self-interference patterns (Fig. 6-5 c and d). We split equally the beam emitted from the micro-ring into the two arms of a homebuilt Michelson interferometer. Because in an OAM beam, the phase varies drastically (helically) close to the central singularity, whereas it is relatively uniform (quasi-planar) at the outer rim, we intentionally created a horizontal offset between the two split OAM beams at the observation plane, so that the dark center of one beam overlapped with the bright outer rim of the other, and vice versa. The interference between helical and quasi-planar phase distributions revealed two inverted forks in the resulting fringes (Fig. 6-5 c). In each fork, a single fringe split into two, which evidently confirmed that the emission from the laser carries OAM with topological charge \( l = 1 \). Similarly, OAM laser emission with topological charge of 2 was also observed experimentally in another design (Fig. 6-5 d), which matches perfectly with our theory.
6.4 Conclusion

Taking advantage of the intrinsic asymmetric coupling originated from unidirectional phase distribution provided by the metasurface, we also demonstrated an on-chip micro-ring OAM laser which directly emits beam that carries OAM with a designable order. This technique holds great promise for achieving compact on-chip OAM light sources (or detectors) for large-scale photonic integration. Especially, it can be used for free-space optical communications with an additional degree of freedom provided by the OAM states.

This chapter is adapted from the publication “Xuexue Guo, et. al. "Molding free-space light with guided wave driven metasurfaces." Science Advances In press, (2020)".
Chapter 7

Summary and Future Work

This dissertation has investigated the important optical phenomena and applications brought forth by spatially and/or temporally phase-modulated metasurfaces. It is in general categorized into two parts: the first is the study on novel optical effects originated from time-varying metasurfaces; and the second is the integration of the multi-functional and versatile metasurfaces into photonic integrated circuits (PICs). Both are frontier areas of metasurfaces researches, which offers immense opportunities as well as challenges.

An ultrafast heterodyne optical modulation method has been developed based on strong nonlinear Kerr effect and optimized metasurface design. Different material platforms are evaluated on their capability for ultrafast and efficient optical modulation (Chapter 2). Taking advantage of the ultrafast modulation, a nonlinear metasurface breaking time-reversal symmetry and Lorentz reciprocity was theoretically proposed and experimentally demonstrated. I have discussed the relation between time-reversal symmetry and Lorentz reciprocity, and validated the nonreciprocal property of our system using both scattering matrix analysis and FDTD simulations. In particular, I achieved a 2.8 THz modulation frequency, a huge step towards optical frequencies, and an approximately 5.77 THz 3-dB bandwidth, which is orders of magnitude greater than that of current time-variant nonreciprocal systems to the best of our knowledge. I also demonstrated the flexibility of the spatiotemporal phase gradient metasurfaces (Chapter 3). In addition, inspired by the extraordinary nonlinear properties of time-varying oscillators, I demonstrated a parametric optical resonator with compact footprint, CMOS compatibility and electrical controllability. Despite the difficulty in creating an effective nanoscale optical parametric resonator, I managed to realize an efficient temporal resonance modulation based on the inherently strong third order nonlinearity of the specially designed amorphous silicon nano-resonator. The parametric enhanced SHG agrees
with the theoretical prediction derived from Mathieu’s equation, which governs the motion of a
temporally modulated oscillator. A large gain factor of 28 dB and an electric-field-controlled on/off
enhancement factor over $10^4$ were achieved in a subwavelength ($\sim 350$ nm) optical resonator using
a weak optical pump power of less than 1 mW (Chapter 4).

I have proposed a unified design methodology for integrating free-space metasurfaces into
PICs, making breakthrough in expanding the available phase tuning range by judicious
nanoantennas design. Leveraging the excellent light controllability offered by metasurfaces and the
mature PICs technology, we push the application of metasurfaces to a broader range, including
optical communications, optical remote sensing (e.g. light detection and ranging (LiDAR)), free-
space optical interconnects (FSOIs), and holographic displays. We demonstrated beam steering and
focusing for guided waves (Chapter 5) and spatial inversion symmetry broken enabled orbital
angular momentum lasing (Chapter 6).

Looking forward, spatiotemporal metasurfaces promise a lot of new physics and
applications. For example, taking advantage of the broken time-reversal symmetry, photonic
topological phenomena $^{253}$, such as one-way edge states and synthetic dimensions $^{254}$, can be
explored. Reciprocity in a broader spectrum, including thermal radiation $^{255}$, microwave $^{80}$ and
acoustic wave $^{256,257}$, can be manipulated for applications in photovoltaics, wireless
communications $^{155}$ and sound engineering. Doppler frequency shift induced/enhanced by temporal
modulation also opens new door to nonlinear optics. These interesting physical effects are mostly
built upon strong and ultrafast optical modulation, making it important to develop new materials,
structures and strategies for boosting light-matter interactions. On the other hand, one of the
ultimate goals of metasurfaces is to create miniaturized spatial light modulator $^{258}$. Tunable and
reconfigurable metasurfaces actuated by mechanical $^{36}$ and electrical stimulations $^{47}$ hold great
promise for this purpose. However, massive challenges need to be tackled towards achieving the
full technological potential of dynamic metasurfaces. First, individual meta-atoms should respond
to external stimulus with large amplitude and phase tuning capabilities. Second, individually addressing and activating massive number of subwavelength meta-atoms poses huge integration and power consumption challenges \(^3\).
Appendix A

Details of InGaAsP/InP Microring Laser Fabrication

InGaAsP multi-quantum wells (MQW) have found wide applications in photodetectors, lasers\(^{259}\), modulators and switchers. III-V MQW structures have several advantages over conventional semiconductors such as silicon and germanium. They have direct bandgap that can be tuned by varying the allow composition. They have large gain values in the range of several thousand cm\(^{-1}\)\(^{259}\) and high carrier mobility. The undoped gain layer was grown on an InP wafer in the InGaAsP quaternary system using metal organic chemical vapor deposition (MOCVD) (OEpic Inc.). The MQW consisted of 29 5 nm thick wells embedded in 29 12 nm thick barriers, with a photoluminescence (PL) emission peak at 1546 nm (Table. A-1). For the fabrication of optically pumped microring laser, we used fabrication processes that consist of a mixture of standard recipes, such as wet etch and electron-beam evaporation, and optimized recipes, such as ebeam lithography of hydrogen silsesquioxane (HSQ) resist and ICP-RIE etch.

<table>
<thead>
<tr>
<th>Layer</th>
<th>Material</th>
<th>Thickness (nm)</th>
<th>Dopant</th>
<th>Loop</th>
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<tr>
<td>7</td>
<td>InP</td>
<td>10</td>
<td>undoped</td>
<td>1</td>
</tr>
<tr>
<td>6</td>
<td>1.2Q InGaAsP</td>
<td>12</td>
<td>undoped</td>
<td>29</td>
</tr>
<tr>
<td>5</td>
<td>1.6Q InGaAsP</td>
<td>5</td>
<td>undoped</td>
<td>29</td>
</tr>
<tr>
<td>4</td>
<td>1.2Q InGaAsP</td>
<td>12</td>
<td>undoped</td>
<td>1</td>
</tr>
<tr>
<td>3</td>
<td>InP</td>
<td>1000</td>
<td>undoped</td>
<td>1</td>
</tr>
<tr>
<td>2</td>
<td>InGaAs</td>
<td>100</td>
<td>undoped</td>
<td>1</td>
</tr>
<tr>
<td>1</td>
<td>InP</td>
<td>300</td>
<td>undoped</td>
<td>1</td>
</tr>
<tr>
<td>0</td>
<td>2&quot; InP undoped</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Total thickness (micron) 1.915

Table A-1 Multiple-quantum-well InGaAsP/InP epitaxial structure.

A.1 Ebeam Lithography

Ebeam patterning process needs to be optimized to faithfully produce the designed pattern, which is influenced by factors such as pattern fracturing method, electron beam dosages, ebeam
resist, proximity effect correction, develop conditions and so on. We used negative tone hydrogen silsesquioxane (HSQ) ebeam resist with high resolution (< 10 nm)\textsuperscript{260,261}, thermal stability and high etch resistance \textsuperscript{260}. Under ebeam exposure, HSQ has an amorphous structure similar to SiO\textsubscript{2}, making it exceptionally suitable for dry etch pattern transfer process. It is very sensitive to the surface condition of the InGaAsP wafer and so correct surface preparation needs to be done, otherwise features would be just washing away in developer. We have found that performing an oxygen plasma clean can be useful in helping to prepare a good surface. We used flowable oxide FOx-16 (16\% of HSQ in methyl isobutylketone (MIBK)) by Dow Corning, and to form a 900nm thick film, it was spun at 4000 rpm. Then the substrate was baked at 100 °C for 3min. Ebeam writing was performed on Raith 5200 ebeam writer with acceleration voltage of 100 kV and beam current of 1 nA. We fractured the microring pattern with a resolution of 5 nm to achieve a smooth curvature, although the width of ring is a lot larger (~ 1 μm). By doing a dose test, we located the optimum dose range 1100 – 1300 μC/cm\textsuperscript{2} (Fig. A-1). HSQ is normally developed by solution containing Tetramethylammonium hydroxide (TMAH). We immersed exposed sample in MF-CD-26 (1.0 - 5.0 \% TMAH) for 20 mins at room temperature. Due to the low concentration developer and thick resist layer, a long development time was used to fully remove weakly crosslinked HSQ and mitigate the footing effect.
Figure A-1 Field emission scanning electron microscopy images of 1 μm microring patterns exposed using electron beam doses of 1200 and 1440 μC/cm². By comparison, dose 1200 µC/cm² has less footing effect. Scale bar (left): 1 μm; scale bar (right): 200 nm.

A.2 Inductively Coupled Plasma – Reactive Ion Etching

Dry etching of InGaAsP/InP requires careful development and optimization to get vertical and smooth sidewalls. Conventionally, the plasma etching of InP-based materials has been predominantly achieved using CH₄/H₂/Ar ICP-RIE, where the CH₄ produces volatile by-products Ga(CH₃) and In(CH₃), and H₂ forms AsH₃ and PH₃. The addition of Ar can enhance the ionization and dissociation of gas molecules and provide ion-assisted desorption of etch by-products. However, it has the disadvantage of polymer deposition on the etched surfaces and reactor sidewalls, resulting in slow etch rate and chamber contamination. The alternative approach is to use Cl₂-based plasma chemistry that is uniquely suitable for Ga-containing material etching, because the Ga by-product is volatile. However, there are challenges involved in dry etching the InP-based compounds in Cl environments due to the involatility of In(Clₓ). It tends to sediment on the etched surfaces and impede the further etch process, resulting in sloped sidewalls. This can be alleviated by raising the substrated temperature (> 200 °C) or using high physical
sputtering of the surface with high-energy ions. Limited by our facility’s dry etching capabilities, we are unable to raise the substrate temperature above 100 °C. We used BCl₃ as the etching chemistry which has been demonstrated for InP etching at room temperature, because In(Clₓ) was removed by ion-enhanced (BCl₂⁺, BCl₃⁺) etching bombardment before a thick nonvolatile layer can build up. As shown in Fig. A-2 b, with optimized parameters (gas: BCl₃ 30 sccm, pressure: 2 mT, chuck power: 200W, coil power: 400W, chuck temperature: 80 °C, no helium cooling), we created very smooth and anisotropic InP etch. It is evident that temperature has an important impact on the etch anisotropy, where samples without helium backside cooling exhibits more vertical sidewalls even though we didn’t purposefully set the chuck temperature above 200 °C. It is probable that the dense and energetic ions bombard the materials and raise the temperature.

By the same token, sample etched with low coil power (a) showed large roughness, because the plasma energy is not strong enough to fully remove the nonvolatile In(Clₓ) while Ga(Clₓ) can be easily removed at room temperature, resulting in an imbalanced removing of different elements and a rougher sidewall. This can also be seen from the different slope of the top InGaAsP MQW layer and bottom InP support layer. Different gas chemistries also have an impact on the etching roughness. It has been reported that the addition of N₂ helps to passivate the etched surface and therefore reduce surface roughness and lateral etching. However, we observed opposite effect in our system. Comparing (b) with (d), we saw obvious roughness increase with the introduction of N₂. This might be due to the fact that the dilution of heavy positive ions BCl₂⁺, BCl₃⁺ reduces the ion energy.
Figure A-2 Field emission scanning electron microscopy images of microrings under different dry etch conditions (the top dark layer is HSQ mask). Common parameters: chuck bias power: 200 W; pressure: 2 mT; chuck temperature: 80 °C. All except (e) don’t have back helium cooling. (a) and (b) comparison of the sidewall straightness and smoothness using different coil power. (b) and (c) comparison of the etch anisotropy under different etching temperatures, without helium cooling (b), the plasma bombardment creates high heat and raise the local temperature, leading to a better etch anisotropy. (b) and (d) comparison of the etch roughness using different gases. Scale bar: 1 μm.
Appendix B

Fabrication of Nanophotonic Devices Based on Plasma-enhanced Chemical Vapor Deposition (PECVD) Silicon Nitride

Silicon nitride is a CMOS compatible material with high refractive index and low loss in a broad wavelength range (visible to near infrared), providing a suitable platform for making various high efficiency nanophotonic devices such as metasurfaces\textsuperscript{270} and photonic integrated circuits\textsuperscript{271,272}. Silicon nitride also has good nonlinear properties: its TPA is negligible in the NIR compared with silicon despite its smaller Kerr nonlinear coefficient\textsuperscript{273}. This has enabled many on-chip non-linear applications, such as supercontinuum and frequency comb generation\textsuperscript{274}. Silicon nitride can be deposited by low pressure chemical vapor deposition (LPCVD) at high temperature (\textgreater{} 700 °C), which produces stoichiometric Si$_3$N$_4$ with excellent control over homogeneity. But the film has very high strain and the operating temperature put severe limitation on multilayer process (with metallization layers). PECVD provides an alternative way to deposit silicon nitride using NH$_3$/SiH$_4$ gases at low a temperature (\textless{} 400 °C). The film composition depends strongly on the gas ratio and can be silicon-rich (high refractive index and loss at shorter wavelengths) or nitrogen-rich (lower refractive index and loss at shorter wavelengths). We will investigate the influence of the PECVD conditions on silicon nitride’s optical properties and the optimized dry etch conditions to produce smooth and anisotropic etch results.

B.1 Study on the Effect of the Precursor Gas Ratio on Optical Properties of Silicon Nitride in the Visible

PECVD growth of Si$_x$N$_y$ involves gas mixture of SiH$_4$/NH$_3$/N$_2$. The chemical composition of the film is controlled by varying the ratio of NH$_3$ to SiH$_4$, which in turn affects the refractive index and absorption of the deposited Si$_x$N$_y$ film. In the visible range, it has been reported that large
absorption in the blue to UV range is attributed to silicon incorporation into the film \(^{275,276}\).

Therefore, in order to minimize the optical loss while keeping the high refractive index, we have studied the influence of NH\(_3\) to SiH\(_4\) ratio on the optical properties of Si\(_x\)N\(_y\) film. We used ellipsometer (J.A. Woollam Co.) to measure the optical properties of Si\(_x\)N\(_y\) film deposited on silicon substrate with different NH\(_3\) to SiH\(_4\) ratios. Other PECVD parameters are: temperature 300 °C, pressure 3.5 T, power 300W and N\(_2\) 2000 sccm. We used Tauc-Lorentz model \(^{277}\) suitable for amorphous materials to fit the refractive index. As shown in Table B-1, compared with LPCVD, our PECVD Si\(_x\)N\(_y\) film has a much lower absorption loss at the blue wavelength (\(\lambda = 473\) nm). Fig. B-1 shows the general trend of the real part and imaginary part of the refractive index change at different wavelengths and gas ratios. Depending on the requirement on the photonic devices and the working wavelength, we can choose the best gas combinations to fulfill both high refractive index and low absorption loss.

<table>
<thead>
<tr>
<th>NH(_3):SiH(_4)</th>
<th>n(473nm)</th>
<th>k(473nm)</th>
<th>n(633nm)</th>
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<tr>
<td>100:75</td>
<td>2.01</td>
<td>0.0039</td>
<td>1.96</td>
<td>0.0026</td>
</tr>
<tr>
<td>110:65</td>
<td>1.97</td>
<td>0.0033</td>
<td>1.92</td>
<td>0.0020</td>
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<tr>
<td>115:60</td>
<td>1.95</td>
<td>0.0028</td>
<td>1.90</td>
<td>0.0017</td>
</tr>
<tr>
<td>120:55</td>
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<td>0.0025</td>
<td>1.89</td>
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<tr>
<td>130:45</td>
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<td>1.88</td>
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<td>0.0223</td>
<td>2.23</td>
<td>0.00059</td>
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</table>

Table B-1. Refractive indices and absorptions of Si\(_x\)N\(_y\) film deposited with different NH\(_3\) to SiH\(_4\) ratios.
B.2 Inductively Coupled Plasma – Reactive Ion Etching

One conventional approach for etching silicon nitride uses a mixture of methyl fluoride (CH$_3$F), tetrafluoromethane (CF$_4$) and oxygen (O$_2$) \textsuperscript{278}. To enable an anisotropic etch profile, sidewall passivation from hydrofluorocarbon polymer residues is necessary. Similar to the silicon etch process, the hydrofluorocarbon film is easily removed in the presence of ion bombardment, but is also able to protect the sidewalls from the impact of sputtered ions, neutrals, and \textit{etc}. \textsuperscript{279}. The involvement of oxygen is to remove hydrofluorocarbon polymer film and create a smooth etched
surface. In order to achieve balanced anisotropy and smoothness, we investigated the role of O₂ and power in the etching of silicon nitride. We etched silicon nitride post (~ 200 nm) using chromium hard mask, and took the FESEM images of different samples (Fig. B-1). Keeping the other two gases constant (CF₄/CH₃F: 30/10 sccm), the comparison between Fig. B-1 a and b shows that an increase in the oxygen flow results in a smoother etched surface but a more isotropic etch. With the same etching gases, by increasing the chuck power we observed more polymer residues on the sidewall and a slight improvement on the anisotropy (Fig. B-1 a and c) due to larger ion bombarding energy. In addition, a higher coil power means higher concentration of reactive species, and we observed more roughness but straighter sidewall (Fig. B-1 a and d). We are especially interested in making high aspect ratio silicon nitride metalens, so it is important to have an optimum etch anisotropy. Based on the etch test, we chose the recipe with minimum oxygen (2 sccm) that produces vertical sidewall while still maintaining a tolerable roughness (Fig. B-1 e). For applications in integrated photonics, it is preferential to minimize the surface roughness which will lead to severe scattering loss. In this case, we will increase the oxygen flow rate to remove sidewall roughness.
Figure B-2 (a) – (d) Dry etch results under different conditions (common parameters: etch time 120s, temperature 25 °C, pressure 2.5 mT, CF4/CHF3: 30/10 sccm). (e) 600 nm thick silicon nitride metalens etched using recipe: CF4/CH3F/O2: 30/10/2 sccm, temperature 25 °C, pressure 2.5 mT, chuck power 50W, coil power 500W. Scale bar: 200 nm.
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VITA

Xuexue Guo

Education:
Pennsylvania State University, University Park, PA           May. 2014 – May. 2020
Doctor of Philosophy in Electrical Engineering
Fudan University, Shanghai, China        Sept. 2009 - Jun. 2013
Bachelor of Science in Optical Science and Engineering

Peer-reviewed Journal Publications:

Conference Presentations or Proceedings:
5. Yimin Ding, Yao Duan, Xi Chen, Xuexue Guo, Xingjie Ni. "A compact photonic integrated spectrometer based on metasurface-decorated waveguides” Proc. SPIE 11080, Metamaterials, Metadevices, and Metasystems 2019, 11080-77