Electrically Reconfigurable Optical Metasurfaces for Universal Wavefront Shaping

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ABSTRACT

The ability to control the properties of light in a compact, reconfigurable platform is essential for advancing nanophotonic technologies. Active metasurfaces — flat optical components with tunable subwavelength elements — enable real-time manipulation of wavefronts and thus offer a path toward versatile optical systems. This thesis furthers the development of electrically programmable metasurfaces as a step toward a universal platform for independent and comprehensive wavefront control. By integrating system-level optimization strategies, novel operation modes, and advanced material platforms, we establish a framework for next-generation, on-demand optical processing components.

First, we introduce an array-level inverse design approach for beam steering metasurfaces, that co-optimizes the spatial amplitude and phase responses to enhance target functionalities. Using the platform of a plasmonic, indium tin oxide (ITO)-based active metasurfaces, we demonstrate non-intuitive configurations that achieve high-directivity, continuous-angle beam steering up to 70°. Experimental validation confirms the effectiveness of this approach, which we further extend to advanced applications including flat-top beams, tunable beam widths, and multi-beam steering.

To expand the functional channel capacity of active metasurfaces, we then explore space-time modulation as a means of enabling multi-frequency operation. By modulating ITO-based metasurfaces operating at near-infrared wavelengths with tailored waveforms at frequencies up to 10 MHz, we experimentally generate desired frequency harmonics, which appear as sidebands offset from the incident laser frequency. Introducing phase offsets between the driving waveforms enables tunable diffraction of frequency-shifted light. Theoretical extensions of this work highlight the potential of space-time metasurfaces to realize active multitasking components capable of dynamically performing multiple independent functions.

For improved efficiency and broadband operation, we investigate electro-optically tunable metasurfaces based on the Pockels effect in barium titanate (BTO). We develop a scalable fabrication technique to obtain high-quality, thin-film BTO via stress-induced exfoliation from single-crystal substrates, preserving its bulk electro-optic properties. The experimentally measured Pockels coefficient r_{33} exceeds that of commercially available thin-film lithium niobate, demonstrating the potential of

this material platform for integration into high-speed, low-loss optical metasurfaces. Leveraging these properties, we design transmissive BTO-based metasurfaces for high efficiency beam steering at visible wavelengths.

The results presented in this thesis lay the foundation for next-generation programmable metasurfaces by addressing key challenges in materials, design methodologies, and system-level control architectures. We conclude with a discussion of future directions, including the discovery of high-performance tunable materials, the development of advanced unit cell designs for independent control over multiple optical properties, and the miniaturization of control networks for large-scale metasurfaces. Ultimately, this work advances the development of reconfigurable and intelligent optical systems capable of adapting to diverse technological demands in a broad range of imaging, communication, and computing applications.

PUBLISHED CONTENT AND CONTRIBUTIONS

1. **Thureja**, P., Nyholm, A. W., Thomaschewski, M., Jahelka, P. R., Belleville, J. & Atwater, H. A. Spalled barium titanate single crystal thin films for functional device applications. *arXiv:2505.04045 [physics.app-ph]*. `https://doi.org/10.48550/arXiv.2505.04045` (2025).

P.T. participated in the conception of the original idea, performed spalls and thin film characterization, developed the thin film transfer technique, prepared samples for electro-optic testing and performed the corresponding measurements, and wrote the manuscript.

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P.T. conceived the idea for this project, fabricated and characterized the active metasurfaces, performed quasi-static and time-modulated measurements, provided inputs for the real-time waveform optimization algorithm, designed the electrical PCB, and contributed to writing the manuscript.

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Chapter 1

INTRODUCTION

1.1 Optical wavefront engineering: Micro- and nanoscale platforms

The first reported optical lens, dating back to the Ancient Egyptian era (2620 BC), employed convex polished rock crystals to form the eyes of ornate statues [1]. The unique function of these lenses enabled the eyes to appear to track the viewer's movement around the statue [2]. Ever since, scientists have dedicated efforts to comprehend and manipulate the properties of light, leading to a variety of advanced technologies that are now integral to society. Notable examples include the invention of the telescope, enabling distant objects to be observed as if they were nearby, and the development of the first photographic camera, capable of capturing the visual information from a scene. In these constructs, optical wavefronts are controlled using bulky optical elements which have a thickness of several millimeters. However, with significant advances in micro- and nanofabrication in recent decades, researchers are now pioneering the development of next-generation micro- and nanoscale optical components. These components allow for precise manipulation of optical wavefronts, giving rise to versatile and unprecedented control over light. In the following, we provide an overview of three micro- and nanoscale technologies that have recently emerged as promising pathways for optical wavefront engineering: optical phased arrays (OPAs), spatial light modulators (SLMs), and (active) metasurfaces.

Optical phased arrays

Optical phased arrays (OPAs) function as the optical analog of radio-wave-based phased arrays and were first realized in the early 1970s [3]. In an OPA, a light source is divided into an array of individual optical emitters, typically facilitated by waveguides connected to antennas. The integration of independently addressable phase shifters enables precise manipulation of the emitted phase at each individual element, enabling applications such as dynamic beam steering. The primary methods employed for implementing phase shifters in OPAs include utilizing thermo-optic effects [4, 5] or p-i-n junctions [6]. Thermo-optic phase shifters offer minimal loss, but typically rely on higher power consumptions and operate at lower speeds. By contrast, p-i-n junctions induce phase delays through refractive index change

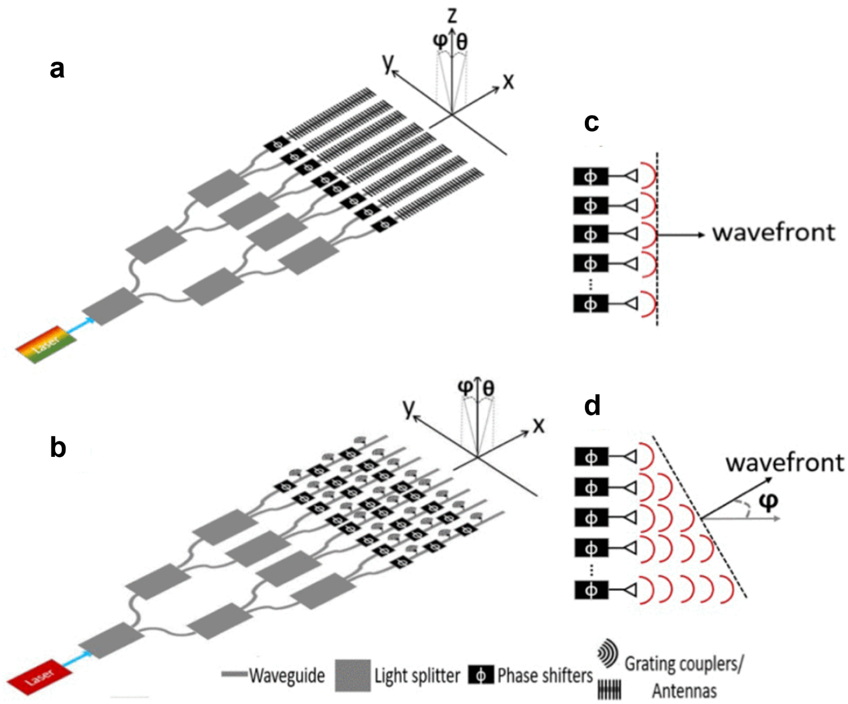

Figure 1.1: **Schematic representation of optical phased array (OPA) for two-dimensional beam steering.** (a) One-dimensional OPA with laser wavelength tuning for steering along longitudinal direction, (b) two-dimensional grating coupler array. (c) Emitted wavefront with no phase delay between emitters and (d) incremental phase delay for steering along transversal direction. © 2021 IEEE [8]

via dopants. Although electro-optic p-i-n phase shifters provide higher modulation frequencies (up to 200 MHz [7]) and lower power consumption compared to their thermo-optic counterparts, dopants typically introduce waveguide losses [8].

The performance and application scope of OPAs further depend on the type of antennas they employ. Typically, antennas in OPAs are constructed using either grating couplers or edge and end fire couplers [9, 10]. Edge emitters are primarily employed for one-dimensional beam steering, while two-dimensional steering has been achieved using grating couplers. One common strategy for achieving two-dimensional beam steering using this platform involves a one-dimensional array of grating couplers, where steering in the longitudinal direction, θ, is accomplished through laser wavelength tuning and steering along the transversal direction, ϕ, is achieved using phase shifters (Fig. 1.1a) [4]. In this configuration, researchers have demonstrated pixel sizes as small as 0.8 μm [11]. Alternatively, a two-dimensional array of individually addressable grating couplers can be employed (Fig. 1.1b) [5]. However, the spatial arrangement of waveguides in arrays typically necessitates large inter-antenna spacings to prevent crosstalk between adjacent emitters. This spacing requirement is further compounded by the need for two-dimensional phase shifter arrays, leading to larger pixel sizes of up to 9 μm [5] that ultimately limit the grating lobe-free steering range. Notably, researchers have demonstrated beam steering with large field of view (FOV) using non-uniform antenna arrangements in sparse arrays [12, 13]. Nonetheless, considerable amounts of background noise are reported in the measured far-field radiation patterns.

In summary, given the operation principle of OPAs, they excel primarily in beam steering and other applications reliant on spatial manipulation of the optical phase front, as schematically illustrated in Fig. 1.1c and d. However, their utility in controlling the properties of an electromagnetic wave beyond amplitude and phase is limited.

Spatial light modulators

Spatial light modulators (SLMs) enable precise control over the amplitude, phase, and polarization of optical wavefronts in response to electrical or optical stimuli [14]. Unlike OPAs, which comprise individual emitters within each unit cell, SLMs function by modulating light transmitted or reflected through a medium, with an individual unit cell, or *pixel*, defined by the control architecture employed in its implementation.

Among the diverse range of SLMs realized to date, the predominant platforms used in the commercial landscape are based on microelectromechanical systems (MEMS) [15] or liquid crystals [16]. MEMS-based SLMs, exemplified by digital micromirror devices (DMDs), utilize miniaturized mirrors that can be tilted to selectively reflect light, and therefore alter the scattered light amplitude. An individual pixel in this configuration is defined by the size of a micromirror and typically ranges around 10 μm or larger [17]. Operating frequencies in such DMDs can reach several tens of kHz [14].

On the other hand, Liquid Crystal on Silicon (LCoS) technology offers smaller pixel sizes and thus precise phase mapping over a larger FOV. In LCoS SLMs, a liquid crystal cell containing electrodes controls the orientation of the naturally birefringent liquid crystal molecules, thus altering the refractive index of the medium. Typical pixel sizes in this configuration range between $3 - 4$ μm [18]. Further reduction in pixel size is limited by crosstalk resulting from the high driving voltages required to modulate a thick liquid crystal cell necessary for full phase control [19]. However, recent advances in nanophotonic device design and microfabrication have allowed a reduction of the pixel size to $\sim$1 μm by modulating phase *via* resonance tuning rather than accumulation across the layer thickness [20]. While this advancement significantly enhances FOV capabilities, the pixel size remains a limiting factor for achieving diffraction-free steering within the visible spectrum. Furthermore, the limited response time of liquid crystal-based SLMs, ranging from hundreds of microseconds to milliseconds, hinders their application in high-frequency beam manipulation [21].

Metasurfaces

Metasurfaces represent a cutting-edge approach to optical manipulation utilizing two-dimensional (2D) arrays of resonant scatterers, typically on the nanoscale. These scatterers have the ability to locally and abruptly alter various properties of light impinging on an interface [22], including its amplitude, phase, polarization, spectrum, and momentum. The collective response from an array of scatterers results in constructive far-field effects that lead to to a variety of phenomena like anomalous reflection [23], polarization control [24], or beam focusing [25]. Due to the subwavelength scale of each metasurface unit cell, metasurfaces enable higher diffraction angles and numerical apertures compared to conventional diffractive optical elements [21] and provide a promising alternative to optical phased arrays and spatial light modulators.

Metasurfaces have garnered significant interest from industries that greatly benefit from the availability of miniaturized and efficient optical components, which can easily be integrated in optoelectronic systems at low cost. Initial efforts focused on passive metasurfaces, where individual scatterers are geometrically designed to induce desired local changes in the electric field (*e.g.*, a phase offset) at the operating wavelength [22, 26–28]. Using this approach, researchers have demonstrated multifunctional passive metasurfaces capable of performing several tasks simultaneously [29], which would traditionally require a combination of multiple bulk optical components.

However, a fundamental limitation of passive metasurfaces is their static nature, lacking post-fabrication tunability. This restriction confines their use to specific, pre-defined functions. In contrast, many modern applications in the realms of optical imaging, communication, and computation require dynamically tunable components for on-demand wavefront shaping. This demand has led to the development of active metasurfaces that can be reconfigured at frequencies up to several GHz, depending on the choice of the material platform. The subsequent section provides a summary of the current state-of-the-art on active metasurfaces.

1.2 Active metasurfaces

Active metasurfaces, in contrast to their passive counterparts, consist of an array of geometrically periodic, often identical, unit cells. Dynamic control over the optical response of the metasurface is obtained through application of an external stimulus. This external stimulus can modify the resonant properties of the subwavelength scatterers *via* actively inducing a refractive index change in an active material layer integrated into the metasurface. Alternatively, the application of an external stimulus can alter the metasurface unit cell dimensions or the relative position of individual scatterers, thereby enabling dynamic control of the wavefront of light reflected or transmitted from the array.

To date, active tuning of metasurfaces has leveraged mechanical deformation of nanophotonic structures [37, 38], field-effect tuning [30, 39], electro-optic [31], thermo-optic [32], electrochemical [33] and chemical effects [40], structural changes in phase change materials [34, 41] and liquid crystals [35], as well as all-optical modulation schemes [36] (Fig. 1.2). Moreover, recent experimental demonstrations have showcased multifunctional active metasurfaces capable of switching between multiple continuously tunable functions. The basis for such devices lies in implementing an active metasurface with individually addressable unit cells. Using this concept, Kafaie Shirmanesh *et al.* [42] demonstrated switching between diverse optically tunable functions, such as dynamic beam steering and varifocal lensing, by adjusting the spatial configuration of voltages applied to a single field-effect tunable metasurface. By using specifically tailored metasurface designs and varying the external stimulus applied onto each metasurface unit cell, researchers have further managed to dynamically generate desired changes in the amplitude, phase, and polarization of the scattered electric field.

The timescale of active reconfiguration of a metasurface is an important characteristic that determines its mode of operation. When the temporal rate of reconfiguration is significantly smaller than the optical frequency, the metasurface operates within what we term the *quasi-static* regime (Figure 1.3a). Conversely, when the metasurface elements are actuated with frequencies higher than the incident laser linewidth, typically a few tens of kHz for state-of-the-art lasers at optical wavelengths, the *time-*

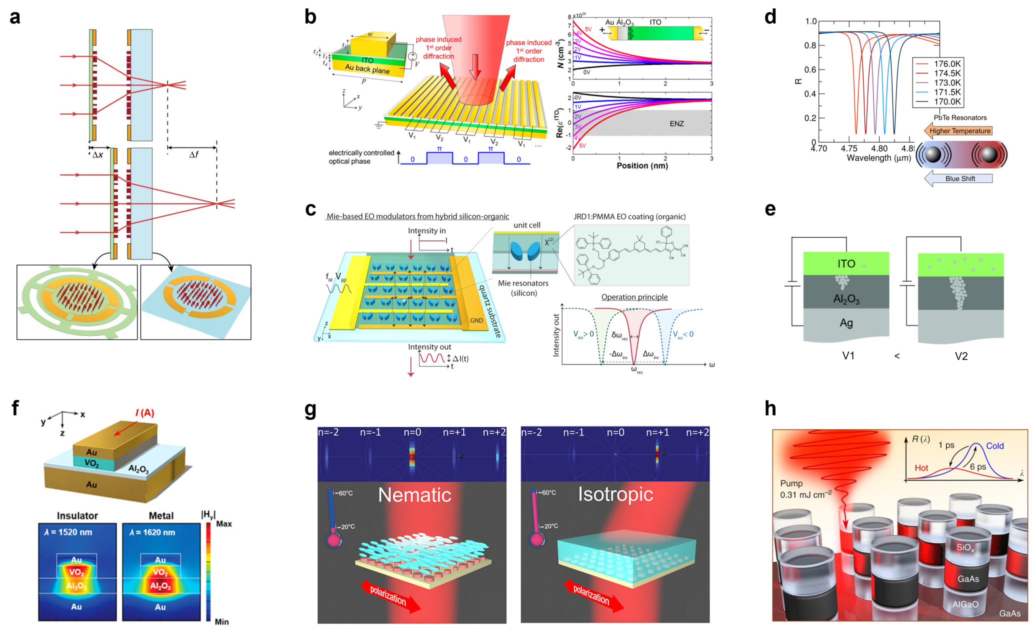

Figure 1.2: **Active metasurface modulation platforms**. (a) Schematic of MEMS-tunable metalens consisting of a moving lens on a membrane (left) and a stationary lens on a substrate; reprinted from [29]. (b) Schematic of field-effect tunable metasurface for beam switching. The tunable semiconductor layer (indium tin oxide, ITO) undergoes an epsilon-near-zero (ENZ) crossing upon carrier modulation. Reprinted with permission from [30]. Copyright 2016 American Chemical Society. (c) Schematic of Mie-resonance based electro-optic metasurface using JRD1 molecules and operation principle. The transmitted intensity undergoes a resonance shift upon voltage application; reprinted from [31]. (d) Thermo-optic tuning of the resonance in lead tellurium (PbTe) Mie resonators. Adapted with permission from [32]. Copyright 2017 American Chemical Society. (e) Schematic of electro-chemical modulation through formation of silver filaments in dielectric layer upon bias application; adapted from [33]. (f) Schematic of metasurface unit cell based on vanadium dioxide (VO_2) phase change material. VO_2 undergoes an insulator-to-metal phase transition in vanadium dioxide (VO_2) upon resistive heating. Reprinted with permission from [34]. Copyright 2019 American Chemical Society. (g) Schematic of beam switching metasurface infiltrated with liquid crystals. The liquid crystals transition from the nematic to isotropic phase upon heating. Adapted with permission from [35]. Copyright 2018 American Chemical Society. (h) Schematic illustration of all-optical resonance tuning in a gallium arsenide (GaAs) metasurface using femtosecond laser pulses. Free-carrier injection and subsequent recombination allow modulation on picosecond timescales; reprinted from [36].

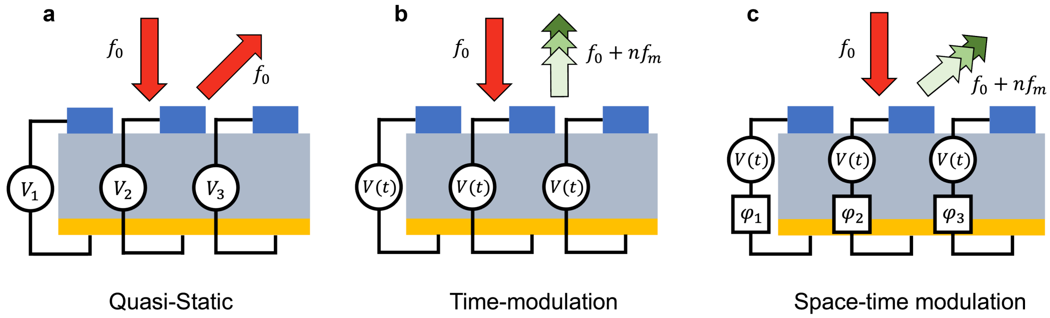

Figure 1.3: **Quasi-static and time-modulated metasurfaces**. Incident light of frequency f_0 is reflected from an electrically programmable metasurface. (a) In the quasi-static operation regime, the frequency of the reflected light remains unchanged and only the propagation direction is altered. (b) In the time-modulated regime, metasurface elements are collectively driven at a high frequency, f_m, allowing the generation of higher order harmonics in the reflected light with frequencies $f_0 + nf_m$. (c) In a space-time modulated metasurface, a phase offset φ_i is added between the time-modulated signals of individual metasurface elements using external phase shifters. This enables shaping of frequency-modulated reflected waves.

modulated operation regime is reached. In this regime, the metasurface gives rise to additional frequency harmonics that appear as sidebands, displaced in frequency relative to the incident laser frequency, as depicted in Figure 1.3b. These individual frequency harmonics can be independently controlled using *space-time modulated* metasurfaces (Figure 1.3c). Here, an additional phase offset is introduced between the high-frequency driving waveform of each metasurface element using external non-resonant phase shifters [43, 44]. This concept holds significant promise for optical communications applications, where high-frequency time modulation enables an increase in the number of communication channels, similar to wavelength division multiplexing (WDM) in optical fiber communications [45]. Furthermore, the ability to simultaneously control multiple frequency sidebands opens the door to increased processing speeds and parallelization capabilities in the realms of optical imaging and computation.

1.3 Goals of this thesis

The ability to dynamically control the optical response in both quasi-static and time-modulated regimes unlocks a vast design space that can be fully exploited by engineering suitable structures for the arbitrary control of light. Currently, researchers are increasingly working toward maximizing the information capacity that can be controlled with an individual nanophotonic element by leveraging control over various input variables, such as modulation amplitude and frequency, to simultaneously alter multiple properties of the scattered light, *e.g.*, its propagation direction, frequency, and polarization. The practical implementation of such devices ultimately involves designing highly efficient optical elements capable of operating at specific optical wavelengths and modulation frequencies. Addressing these challenges requires a holistic approach that encompasses system-level problems, nanophotonic design optimization, and understanding of fundamental materials science. This thesis addresses multiple aspects of this broader objective.

Chapter 2 of this thesis focuses on the optimization of spatially varying wavefronts scattered from an active metasurface using an array-level inverse design approach. The optimization is numerically and experimentally demonstrated for a field-effect tunable indium tin oxide (ITO)-based metasurface with independently addressable metasurface unit cells. The tools developed in this chapter are applicable to a variety of active metasurfaces that are based on localized modes and provide a pathway for minimizing system-level losses in metasurfaces. We study the impact of nonideal phase and amplitude response and analyze the requirements for high-directivity beam steering metasurfaces.

In Chapter 3, we experimentally realize space-time modulated metasurfaces at optical frequencies, by driving ITO-based metasurfaces operating at telecommunication wavelengths with MHz voltage waveforms. By tailoring the phase offsets of applied waveforms, we engineer spatiotemporally varying configurations for targeted steering of frequency-shifted beams. This concept allows us to extend the information capacity of an individual metasurface, ultimately forming a path toward active *multitasking* metasurfaces, that allow simultaneous and independent dynamic control over multiple frequency channels.

Chapter 4 delves into the aspects of increased efficiency and operation at target wavelengths across the visible spectrum using the electro-optic response of barium titanate ($BaTiO_3$, BTO). We demonstrate a novel synthesis approach for single-crystalline thin films that involves exfoliation from a stressed single crystal bulk substrate. We furthermore characterize the BTO thin films in terms of their physical properties as well as their electro-optic response. Finally, we design electrically tunable transmissive metasurfaces based on BTO for operation in the visible spectrum.

In Chapter 5, we provide an outlook toward the development of *universal* active metasurfaces, discussing the requirements to achieve this goal and identifying key challenges that remain. This chapter synthesizes insights from the preceding chapters to outline the future of active metasurface research. We additionally delve into prospective use cases of universal active metasurfaces in the realms of optical imaging, communication, and computation. The dissertation is concluded with a summary of the key findings, highlighting significant advances and potential impacts on the field of nanophotonics, in Chapter 6.