Chapter 5

TOWARD UNIVERSAL ACTIVE METASURFACES FOR OPTICAL IMAGING, COMMUNICATION AND COMPUTATION

The material in this chapter was in part presented in [21, 96].

5.1 Universal active metasurfaces

The ability to dynamically control the optical response in the quasi-static and time-modulated regimes opens a multidimensional design space that can be fully harnessed by developing appropriate nanophotonic structures for arbitrary manipulation of light. In the previous chapters, we explored how these capabilities can enhance the information density of metasurfaces while utilizing high-performance materials that can be seamlessly integrated into designs for both reflection and transmission. Building on this foundation, we now address the fundamental question: What does it take to realize a universal optical element (Fig. 5.1) which enables dynamic, independent, and comprehensive control over all constitutive properties of light in both reflection and transmission? State-of-the-art wavefront shaping metasurfaces generally encompass active and continuous control over the amplitude and phase of light scattered from each nanostructured element. A universal active metasurface, in comparison, should additionally provide complete control over the polarization, spectrum and momentum, the orbital angular momentum as well as the shape of optical pulses. Such a universal active metasurface would have the potential to serve as a programmable transfer element that can encode arbitrary functions and perform a variety of complex tasks using a single dynamically tunable component. As such, it could be integrated into a wide range of applications, including free-space communications, analog computing, and holographic displays to name a few.

In this chapter, we focus on the pathways toward achieving a universal active metasurface for independent and comprehensive, real-time control over all properties of light, including its amplitude, phase, polarization, momentum, spectrum, orbital angular momentum, and pulse shape. We identify key challenges and highlight the respective recent developments in three areas that will drive the realization and unlock the potential of a universal active metasurface: metasurface design, control architecture, and advanced operation modes. We outline the target performance

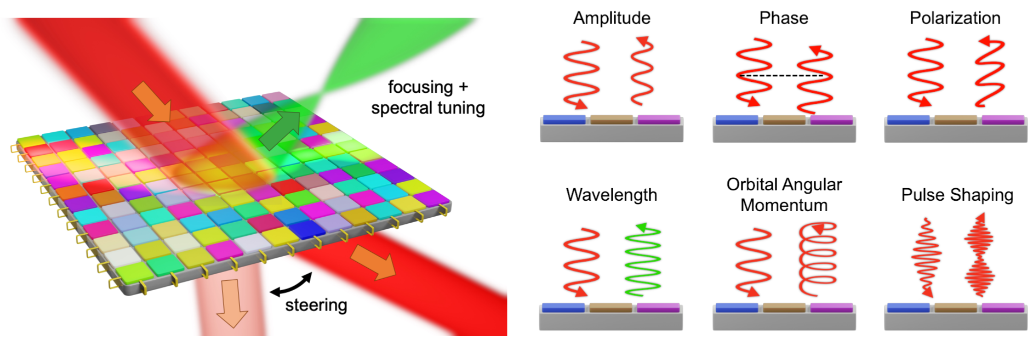

Figure 5.1: **Universal active metasurface.** Schematic of a universal active metasurface enabling dynamic wavefront shaping in reflection and transmission. This metasurface should allow for dynamic and independent control of the amplitude, phase, polarization, wavelength, the orbital angular momentum, and pulse shape of an electromagnetic wave.

characteristics with regards to a high-performance active metasurface design and discuss potential pathways for their realization. We further examine challenges toward realizing a two-dimensional control architecture and discuss strategies to overcome currently existing limitations. We then explore uncharted operation modes that could be attained with a universal metasurface, providing inspiration for future applications in the realms of optical imaging, communication, and computation. Finally, we give a perspective and point toward key technological problems that will require further research from the nanophotonics community to bring us closer to the realization of a truly universal active metasurface.

5.2 Metasurface design and promising material platforms

The design of a universal active metasurface forms a convoluted problem involving several parameters. Passive metasurfaces generally rely on shape- and orientation-dependent phase retardation introduced by resonant scatterers to create desired changes in the scattered electric field. In comparison, the performance of an active structure is determined by its geometrical parameters as well as the choice of the active material and/or the corresponding modulation scheme. In the following, we review active metasurface design strategies that hold the potential to realize a universal active metasurface. The desired objectives can be summarized as follows: First, a large dynamic range of tuning is desired for deterministic wavefront shaping. Here, the objective is to obtain complete and independent control over the characteristic properties of light. This includes a 0-to-2π phase shift upon actuation (while maintaining constant amplitude) and 100% intensity modulation efficiency (with constant phase). We define the latter as $\eta = 1 - I_{\text{min}}/I_{\text{max}}$, where I_{min} and I_{max} are the minimal and maximal electric field intensity at a given wavelength, respectively. In addition, low loss structures are required to ensure high output efficiencies. To access the time-modulated operation regime, active metasurfaces further need to support large modulation frequencies. Additional performance metrics include broadband operation, allowing for a tunable excitation wavelength and achromaticity. While these criteria do not form an exhaustive list of requirements for a high-performance universal metasurface, they determine critical design choices for the architecture and materials employed in a metasurface unit cell as well as its modulation mechanism.

Field-effect tuning based on carrier index modulation (discussed in detail in Chapters 2 and 3) has attracted significant attention over the past decade, as it has been used to experimentally demonstrate a large dynamic range of tuning of amplitude, phase, and polarization. Individual studies have reported up to 96% reflectance modulation efficiency [215], greater than 1.5π phase modulation [42, 53, 54, 216], as well as a linear to circular and cross-polarization conversion [217]. Field effect tuning relies on the charge carrier dependent optical properties in semiconductors, often transparent conductive oxides (TCOs), or in graphene. In semiconductor-based tuning, a metal (or semiconductor)-insulator-semiconductor heterostructure [30, 69] is integrated into a resonant unit cell. Upon gating, a local charge carrier accumulation or depletion zone is created at the insulator-semiconductor interface (Fig. 5.2a), causing a change in the complex dielectric permittivity, ε [218, 219]. Additionally tuning the dielectric permittivity into the epsilon-near-zero (ENZ) regime leads to an extreme localization of the electric field in this zone, which perturbs the optical resonant mode of the unit cell [220]. This ability to transition the active material through the ENZ region, and hence dynamically modify the scattering of a resonant unit cell has led to the development of electronically programmable active metasurfaces with independently addressable metasurface elements [42, 221]. Moreover, modulation frequencies of up to 10 MHz have been demonstrated using TCO-based metasurfaces [30, 95], with the potential of accessing GHz frequencies with optimized design of device electrical interconnects and driver circuits.

The operating wavelength of TCO metasurfaces is chosen based on the bulk carrier density of the active semiconductor during fabrication. Indium tin oxide (ITO) is a commonly used active material for operation in the near-infrared. A recent study, however, showed that over 70% of the incident light is absorbed in the ITO layer at its ENZ transition and only 2.7% of the light is reflected [74]. Cadmium oxide (CdO) was proposed as an alternative transparent conducting oxide with enhanced optical properties, leading to reflectance values above 22% [74]. For operation in the mid-infrared (3 – 10 μm), several materials including graphene, and doped semiconductors such as indium arsenide (InAs) [222] have been proposed. Phononic materials, such as silicon carbide (SiC) [223], indium phosphide (InP) [224] and gallium arsenide (GaAs) [225], may provide a promising pathway to realize field-effect tunable metasurfaces operating beyond 10 μm [226]. This class of materials supports a phonon-polariton mode that undergoes an ENZ transition in the far-infrared while maintaining a low extinction coefficient. For operation in the visible, transition metal nitrides stand out as an emerging class of materials that undergo an ENZ transition between 400 nm – 800 nm [226, 227].

Atomically thin polar van der Waals (vdW) materials and transition metal dichalcogenides (TMDCs) offer an alternative pathway to extend the operation wavelength of active metasurfaces to the visible spectrum. These materials support strong excitonic resonances at wavelengths determined by the dielectric properties of the TMDC material. By integrating the 2D semiconductor material into a capacitive structure and biasing the gate electrode, charges can be injected into the active layer. Due to the exciton-charge interaction, a modification of the excitonic resonance is observed [228–230]. As a result, a large change in refractive index can be achieved [231, 232]. The index changes at excitonic resonances manifest themselves in phase and amplitude modulation even without explicit optical resonators. An integration into planar heterostructures forming resonant cavities further enhances the observed dynamic range of tuning [233–235]. Recently, black phosphorus (BP) was proposed as an excitonic material suitable for active metasurfaces. The intrinsic anisotropy along the crystal axes [236] of BP allows for dynamic polarization control [237]. Currently, patterning of 2D materials and integration into metasurface building blocks is being pursued to further enhance light-matter interactions and enable advanced optical functions [83]. Recent experimental demonstrations of this include a tunable zone plate lens allowing for a modulation of the focal intensity [238] as well as a nanostructured array for dynamic control of the scattering pattern, supporting modulation frequencies of up to 625 MHz [132].

Despite the progress on the dynamic modulation achieved using field-effect tunable metasurfaces, there are several limitations that need to be overcome for use in applications. One of the challenges with metasurfaces based on thin films of excitonic materials is the scalability of devices to large areas (greater than 500 μm^2). The highest quality 2D semiconductor films are obtained through mechanical exfoliation. This approach becomes especially challenging when multiple layers need to be stacked on top of each other. Recent advances with robotically assisted thin film transfer [239, 240] may provide a potential pathway toward realizing scalable active heterostructures. Nevertheless, current TMDC-based active structures require cryogenic temperatures to realize their full dynamic range of tuning [132]. Additionally, while the operating wavelength of TCO-based metasurfaces can be changed to some degree *via* an appropriate choice of material doping, the operating wavelength range for TMDC-based structures is intrinsically fixed by the excitonic transition of the material. Field-effect tunable metasurfaces further require a high electric field confinement and mode overlap of their active media with resonant nanocavity structures to obtain strong light-matter interactions. This can be achieved *via* plasmonic resonators and cavities, which, however, contribute to high absorption and thus lower device efficiencies. Alternatively, a double distributed Bragg reflector cavity was proposed to enhance light-matter interactions in a BP-based planar heterostructure [241]. Using this design, a phase modulation of 300° was predicted along with a minimum reflectance above 50% at an operating wavelength of 2.9 μm, where BP exhibits lower losses. Further research is required to explore a potential extension of this platform to an active metasurface with independently addressable unit cells.

Electro-optic tuning of metasurfaces offers a promising approach to low-loss metasurfaces. As discussed in Chapter 4, the Pockels effect relies on a linear variation of refractive index in response to an applied electric field (Fig. 5.2b) [242]. This is a broadband phenomenon which appears in non-centrosymmetric crystals, such as lithium niobate (LN) [243], barium titanate (BTO) [173], and aluminum nitride (AlN) [157] amongst others. Electro-optically tunable materials further support ultrafast modulation frequencies (>100 GHz in electro-optic modulators [244]). While polycrystalline or amorphous thin film materials typically exhibit smaller electro-optic coefficients compared to single crystal materials (see Table 4.2), commercially available high-quality LN thin films have recently propelled the integration of electro-optic materials into active metasurfaces [105, 245]. Another significant advance entailed a recent demonstration of epitaxially grown thin-film BTO with an approximately 30× larger electro-optic coefficient compared to LN [165]. Reports of active metasurfaces integrating BTO as a tunable layer, however, have thus far been limited to lower electro-optic coefficients [139]. In Chapter 4, we propose spalling

BTO thin films from bulk single crystals to overcome this challenge. While our measurements indicate that spalled thin films retain bulk electro-optic properties, further research is needed to realize seamless integration of spalled films into active structures. Organic electro-optic (OEO) chromophores, in comparison, provide a promising pathway for low-cost, high-throughput integration using methods such as spin-coating, micro-dispensing or ink-jet printing [161, 246, 247]. They have additionally enabled a strong modulation of the transmitted intensity at frequencies up to 5 GHz [31]. However, we note that high electric field intensities may cause photo degradation of the chromophores due to the presence of oxygen in the active material [248]. Besides the Pockels effect, the quantum confined Stark effect offers an alternative route to obtain a fast, electro-optically tunable response. In this case, a change in refractive index is induced upon an applied electric field, which causes a shift in the interband transition energy [153–155]. Based on this principle, Wu *et al.* [75] recently proposed an all-dielectric active metasurface based on multiple quantum wells (MQWs). In this work, active beam switching was realized with a phase shift of 70° and a simultaneous reflectance modulation efficiency of $\eta \sim 73\%$.

We refer the reader to Table 4.1 for a summary of the electro-optic refractive index change obtained in various nanophotonic devices using the active dielectric materials highlighted here. Due to a modest index change, the active electro-optic layer generally needs to be coupled to a high-quality (Q) resonant mode to obtain a strong modulation of the scattered wavefront using active metasurfaces. To this end, several metasurface designs relying on guided mode resonances have been realized [75, 152]. However, in this case, the extended mode along one dimension complicates the design of compact resonant elements for two-dimensional metasurface arrays with independently addressable unit cells. Active metasurfaces governed by optical bound states in the continuum (BICs) or quasi-BICs [249–251] may offer another promising pathway to incorporate electro-optic materials into high-Q resonant structures [252] and thus represent one approach to the design of non-local active metasurfaces [77]. Non-local metasurfaces rely on a collective mode generated by an array or subset of metasurface elements. Similarly, Weiss *et al.* [245] proposed a design strategy relying on the mode overlap of a Fabry-Pérot resonance, a localized surface plasmon resonance, and a surface lattice resonance. Notably, the surface lattice resonance only appears when the array periodicity coincides with the localized surface plasmon resonance of the metasurface elements. Therefore, further investigations are required to analyze whether this modulation scheme can be extended to gain active control on an individual unit cell level.

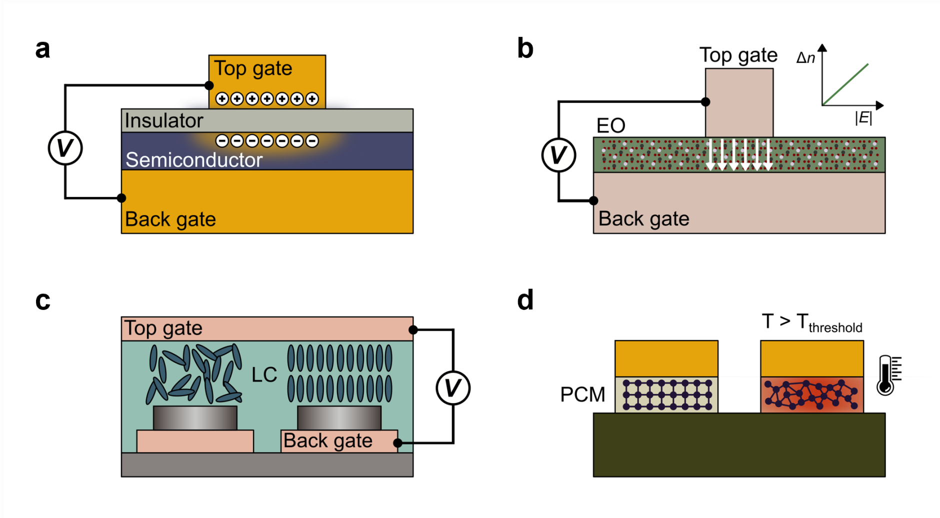

Figure 5.2: **Promising material platforms for universal active metasurface.** (a) Field-effect tunable metasurface. When the active semiconductor is integrated into a metal (or semiconductor)-oxide-semiconductor layer, a charge accumulation layer is formed upon application of a gate voltage. (b) Electro-optically tunable metasurface relying on Pockels effect. An electric field E (white arrows) applied across an electro-optic (EO) thin film of a non-centrosymmetric crystal leads to a linear change in refractive index Δn. (c) Liquid crystal-based metasurface. The liquid crystal molecules (LC) reorient in an external electric field. (d) Active metasurface based on phase change materials (PCM). The active material undergoes a phase transition from crystalline to amorphous when heated above a threshold temperature $T_{\text{threshold}}$.

Liquid crystals are well understood materials that have been widely deployed in display applications, and thus offer another active medium for realizing active metasurfaces with individually addressable unit cells. Here, a tunable optical response is obtained due to the optical birefringence of the liquid crystal molecules, which reorient upon application of a thermal [35, 253] or electrical stimulus (Fig. 5.2c) [20, 254]. The corresponding large refractive index change ranges from $\Delta n = 0.15 - 0.4$ across the visible and infrared spectrum ($\sim$ 460 nm – 80 μm) [20, 255]. A commonly used active metasurface design approach relies on Huygens' scatterers which are based on the spectral overlap of electric and magnetic dipoles. By actuating individual unit cells using an electrical bias, the liquid crystal orientation can be locally modified, as shown by Li *et al.* [108] in a transmissive liquid crystal-based metasurface. Alternatively, an active reflective liquid-crystal structure was recently realized based on Fabry-Pérot nanocavities that support multiple resonances across the visible spectrum [19]. Here, a continuously tunable phase shift across 2π is obtained while maintaining a high reflectance above 40%. It is worth noting that as the metasurface pitch is reduced to values below 1 μm, crosstalk between unit cells arises due to the elastic motion of liquid crystals [20, 256]. Furthermore, due to the response time of the molecules, the modulation frequencies are limited to several kHz, with currently fastest rates of 40 kHz reported in commercial light detection and ranging (LiDAR) devices [131].

Phase change or phase transition materials, by contrast, produce structural changes in the active layer upon Joule heating (Fig. 5.2d), and take advantage of the large difference in complex refractive index achievable in different phases of the same material. Phase change materials, such as germanium-antimony-telluride (GST) and its derivatives [41, 257], undergo a non-volatile transition from an amorphous state to a crystalline state. Phase transition materials, on the other hand, rely on a volatile insulator-to-metal transition in materials such as vanadium dioxide (VO_2) [34, 258]. Both material categories have been integrated into active structures exhibiting multi-level phase tuning. Experimental realizations of active metasurfaces using an electrical stimulus, however, have thus far been limited to modulation frequencies of 3 kHz [258]. Optical control has enabled significantly faster modulation rates, with amorphization in GST induced upon 50 fs long laser pulses [259]. (Notably, the recrystallization required repetitive pulsing of the fs laser for 1 s at 960 Hz.) Furthermore, several metasurface designs realizing tunable functions, such as active beam switching [258, 260] and bifocal lensing [260], have been demonstrated using phase change and phase transition materials. Nevertheless, the implementation of a metasurface with independently addressable elements requires further investigations on thermal heat management. In a preliminary study, Kim *et al.* [261] theoretically showed that thermal crosstalk between adjacent elements in a VO_2-based metasurface could potentially be mitigated by incorporating heat conduction layers into the design.

High-performance active metasurfaces comprise structures that allow independent and comprehensive control over all properties of light. A full 2π phase coverage with close to unity amplitude has previously been obtained through strategies such as coupling of a resonator to a back reflector [152]. Additional independent control over the amplitude was attained using dual-gated structures [53], which can cover the entire complex amplitude space based on distinct voltage configurations [54, 216]. Notably, however, the design of compact and efficient active metasurfaces becomes increasingly challenging with the addition of each degree of freedom. At each step, the design problem constitutes of a thorough co-optimization of the active material, the geometrical parameters of the metasurface element, as well as the external control variable. Furthermore, it becomes crucial to weigh trade-offs between different objectives for a desired application. As such, the task of finding an optimally functioning universal active metasurface is ideally formulated as an inverse design problem.

While inverse design has been widely explored for the geometrical optimization of unit cells in passive metasurfaces [57, 60, 62], active metasurfaces present a unique challenge in which the performance of a device must be optimized at multiple states simultaneously [262]. This concept of multi-state optimization has been theoretically demonstrated to optimize the function of active metasurfaces based on phase change materials [262, 263] or liquid crystals [64], which can switch between two states. In these studies, a shape or topology optimization was conducted on a single unit cell level using multi-objective optimization algorithms. For the design of a continuously tunable metasurface, however, the number of operation states of a metasurface dramatically increases. This challenge is further amplified with the independent addressability of metasurface elements. Several groups have thus employed a so-called array-level inverse design (see Chapter 2) [46, 264]. Here, the value of the external bias is optimized at each element to overcome the limitations posed by a forward-designed active unit cell, which exhibits co-varying phase and amplitude and smaller than 2π phase shift upon actuation. We would like to note here that a common challenge in obtaining high-performance active metasurfaces is the degraded array performance despite working with structures that exhibit a large phase modulation and high reflectance. While some of these discrepancies can be attributed to fabrication imperfections, an important aspect that needs to be addressed is mutual coupling between neighboring metasurface elements [265].

Table 5.1 provides an overview of the performance of different modulation mechanisms in terms of the initially defined objectives. Field-effect tunable metasurfaces relying on carrier modulation and liquid crystal-based active structures stand out in terms of the achievable intensity modulation efficiency and the phase modulation. While field-effect tunable metasurfaces outperform in terms of the modulation frequency, liquid crystal-based architectures enable higher efficiency designs in both reflection and transmission. However, a drawback of current liquid-crystal based structures is the lack of subwavelength control in the visible due to mutual crosstalk. Meanwhile, metasurfaces relying on electro-optic tuning seem promising in terms of their efficiency and the accessible modulation frequencies. The operation wavelength can additionally be chosen from a broadband regime. However, the small index changes in the active material pose additional challenges to the realization of high-Q resonant metasurface unit cells at small dimensions needed for full two-dimensional control. Metasurfaces based on 2D materials or phase change/phase transition materials may provide an effective alternative to satisfy the desired objectives, however, several challenges need to be surmounted before these technologies can be used in applications, including the design of low-loss broadband structures with individual unit cell control. Ultimately, the realization of an optimal universal metasurface will rely on inverse design which is part of an overall hierarchical co-design of the active metasurface components: from the desired dielectric function of an active layer to an optimization of the unit cell shape as well as the configuration of the external stimuli. Multi-objective optimization algorithms could further support the design of metasurface unit cells that are robust to fabrication imperfections. To this end, an active area of research will consist of the development of computationally efficient algorithms. Supplementing the algorithms with physics-based models and constraints will enable efficient search and optimization of experimentally feasible design spaces [266].

5.3 Two-dimensional metasurface control

To shape arbitrary wavefronts in space, a universal active metasurface needs to be fully reconfigurable in two dimensions. This is of particular interest for many imaging and communication applications, which may require directional scanning of beams across a scene. Two-dimensional control further enhances the information processing capability of an optical computing metasurface by dramatically increasing the number of independently addressable unit cells. However, this increased number demands sophisticated control architectures that allow for compact chip packaging with subwavelength unit cell spacings. Additionally, the control architecture should have minimal interference with the optical response of an active metasurface, *i.e.*, ideally the control system should not cause any degradation in the dynamic range of tuning or the attainable efficiency. Most demonstrations of active metasurfaces to date have consisted of a collective modulation of an entire array of scatterers [77, 244, 258] or addressing of individual unit cells along one dimension of the array (while connecting the scatterers along the perpendicular direction) [42, 216]. In the following, we highlight the active modulation schemes most suitable for achieving two-dimensional control based on the photonic modes and the nature of the external stimulus. We then discuss several pathways of designing an appropriate control architecture and evaluate the impact the respective approaches may have on the performance of an active metasurface.

To achieve an active metasurface that can be controlled in two dimensions, the resonant photonic mode needs to be confined along the lateral, longitudinal, and vertical dimension of the metasurface. This has been achieved in various active metasurface platforms using geometrically resonant scatterers in field-effect tunable metasurfaces [42] or liquid crystal-based structures [20]. In the case of a guided mode resonance [75], an extended mode is formed. Thus, independent control of metasurface elements can only be performed along the direction perpendicular to the guided mode, limiting this approach to one-dimensional spatial modulation. Similarly, active non-local metasurfaces [77] rely on phase modulation over length scales that are larger than the wavelength (and thus an individual metasurface element), but potentially smaller than the metasurface aperture. While this does not necessarily exclude two-dimensional control, additional research efforts are required to evaluate how non-local metasurfaces can be used for arbitrary wavefront shaping.

Additional constraints toward the realization of a two-dimensional control architecture are imposed by the nature of the external stimulus, which can be electrical, thermal, optical, chemical, or mechanical. Mechanical deformations, that are caused by, for example, stretching of an elastic substrate [274, 275], have previously been used to demonstrate two-dimensional beam focusing. However, prior research relied on changes that are introduced over the entire array configuration. As a result, the realized devices are restricted in terms of achieving different functions using a single chip. Similarly, it is challenging to confine changes induced using external chemical sources (such as the hydrogen or oxygen flow in hydrogenation metasurfaces [40]) or thermal sources [35, 276] to subwavelength spaces. Advanced schemes to inhibit the interference of neighboring unit cells would be necessary, requiring extensive multiphysics analysis and design. In comparison to these concepts, all-optical modulation relies on a pump-probe experiment, in which an intense pump pulse generates free carriers in the active medium and thus alters the properties of the scattered probe pulse [277]. Alternatively, interference of the two beams [278] or optical pumping of metasurface elements [279] can introduce structural and refractive index changes in the active medium. In either case, the resolution of the spatial pattern generated on the metasurface is given by the diffraction-limited spot of the pump laser. The requirement of additional pump lasers and beam scanners to write individual elements, however, inhibits a compact integration.

Over recent years, electrical biasing of metasurface elements has emerged as a promising pathway to enable two-dimensional control on a single unit cell level. The introduction of localized effects using an electrical stimulus has led to the development of several technologies ranging from electrically induced structural changes in phase change materials [118, 257] or liquid crystals [20] to mechanical deformations of scattering elements [37]. Due to its versatility in being able to be integrated with several modulation schemes, the following discussion aims to highlight potential pathways toward realizing a two-dimensional electrical biasing network and their respective challenges.

One route to designing an interconnect architecture for a two-dimensional active metasurface is to create individual biasing lines for each metasurface element. Kim *et al.* [221] recently experimentally demonstrated two-dimensional wavefront control using a plasmonic, field-effect tunable metasurface. In this work, fan-outs were used to electrically address each individual unit cell (Fig. 5.3a). The gate electrodes were designed orthogonal to the scatterer orientation, as illustrated in Fig. 5.3a. This configuration results in minimal perturbation of the resonance when the scatterers are excited with linearly polarized light in the y-direction. A common challenge with a biasing architecture like this, however, is its scalability. For metasurfaces with thousands or even millions of scattering elements, larger spacings between unit cells are required due to routing complexities. To overcome this limitation without sacrificing the metasurface aperture, multiple scatterers can be connected to one electrode, as shown in [221]. Notably, as the unit cell size approaches the incident wavelength, the field-of-view of the metasurface is strongly reduced. Additionally, an increased amount of undesired scattering is expected for coarsely resolved phase profiles, as previously shown for gradient beam steering metasurfaces [42].

An alternative strategy relies on row-column or perimeter tuning of individual unit cells. This approach involves connecting the metasurface elements in individual rows and columns (Fig. 5.3b), respectively, allowing for a dramatic reduction of biasing lines from N^2 (for individual unit cell control) to $2N$ for an array consisting of $N \times N$ unit cells. Row-column tuning is commonly used in commercial spatial light modulators consisting of large arrays of pixels (more than 1000×1000 elements). Spatial light modulators comprise non-resonant pixels relying on a phase accumulation in thick liquid crystal layers [280]. Arbitrary wavefront shaping is achieved using either a dynamic random-access memory (DRAM) or a static random-access