Opinionated article

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The road to atomically thin metasurface optics

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Abstract: The development of flat optics has taken the world by storm. The initial mission was to try and replace conventional optical elements by thinner, lightweight equivalents. However, while developing this technology and learning about its strengths and limitations, researchers have identified a myriad of exciting new opportunities. It is therefore a great moment to explore where flat optics can really make a difference and what materials and building blocks are needed to make further progress. Building on its strengths, flat optics is bound to impact computational imaging, active wavefront manipulation, ultrafast spatiotemporal control of light, quantum communications, thermal emission management, novel display technologies, and sensing. In parallel with the development of flat optics, we have witnessed an incredible progress in the large-area synthesis and physical understanding of atomically thin, two-dimensional (2D) quantum materials. Given that these materials bring a wealth of unique physical properties and feature the same dimensionality as planar optical elements, they appear to have exactly what it takes to develop the next generation of high-performance flat optics.

Keywords: 2D materials; flat optics; metasurfaces.

1 A brief historical perspective of passive metasurfaces

In optics, we traditionally control and measure the behavior of light using bulky optical components. The field of flat optics aims to manipulate the flow of light with more compact elements and we are currently seeing an explosion of research on this topic. To appreciate the somewhat daunting number of directions this field is currently moving into, it is important to briefly look back at its historical development. Many reviews already provide valuable, different perspectives on the topic [1–6] and in this opinionated article I just highlight some of the key developments. During the second world war, early work by Kock already aimed to create compact diffractive lenses in the microwave range [7, 8]. However, only in the last five decades have researchers attempted to replace bulky refractive optical elements operating in the visible and infrared spectral ranges by razor-thin, planar components. This journey started with the development of diffractive optical elements (DOEs), which came with the exciting promise to reduce the size and weight of complex optical systems. Early work includes research on metallic artificial index gratings that could serve as beam deflectors and polarizers [9]. The first dielectric échelette-type DOEs, such as the Fresnel lens, shaped the phase front of light by manipulating its propagation phase. They were created by judiciously structuring the surface of transparent materials to create wavelength-scale height variations. These designer topographic features directly translate into spatially variant changes of the local phase pickup as light propagates through the element. Binary blazed gratings evolved from échelette components and they manipulate the propagation phase by nanostructuring thin films into dense forests of pillars that feature a binary height-profile [10]. If the refractive index of the pillar material is sufficiently high, they can serve as tiny, truncated waveguides whose physical dimensions determine the locally incurred phase lag upon propagation from one end to the other. By carefully choosing all of the pillar cross-sections in an array, it is again possible to imprint spatially varying phase changes onto an incident light wave. These elements are easier to fabricate and offer higher diffraction efficiencies for significant deviation angles (high-NA) by avoiding undesired shadowing effects. For very dense arrays of high-index pillars or ridges, optical coupling cannot be ignored and a physical picture involving coupled Bloch-modes [11] becomes the most insightful to explain some of the unique properties that are achievable at higher areal densities, such as broadband reflectance ($\Delta\lambda/\lambda \approx 0.3$) and resonances with very narrow linewidths ($\Delta\lambda/\lambda < 10^{-3}$). This kicked off the field of high-contrast gratings with many integrated optics applications [12]. It was shown that similar types of

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wavefront manipulation can also be attained by engineering the light transmission through nanoscale holes [13, 14] or engineered gap plasmonic waveguides [15] carved into thin metallic films.

In 2001 geometric-phase, flat optical elements entered the scene and provided a fundamentally new way to shape wavefronts by varying the orientation as opposed to the size and shape of subwavelength structures [16–19]. They offer complete 0 to 2π phase control by simply rotating structures, whose geometry and spacing is optimized to achieve high diffraction efficiencies [20]. Geometric phase elements have fabrication advantages as all scattering elements can be the same size and shape. By having the structure's orientation control the phase, it was for the first time possible to realize high-negative-permittivity metallic optical elements that are deep-subwavelength in thickness [21]. A unique and sometimes challenging feature, is that the local phase pickup depends on the polarization handedness of the incident light. This can be used to one's advantage in e.g. polarimetry applications [22, 23] or avoided through clever design [24].

Another important conceptual step in the development of flat optics was the realization that optical resonances in first metallic [25, 26] and later high-index semiconductor [27, 28] nanostructures could also be harnessed to create flat optical elements. Resonances can notably enhance light scattering cross sections of small particles and effectively control the phase [29]. Their design closely follows that of radiofrequency antennas and were consequently termed optical antennas [30]. As these structures resonate guided optical modes with strongly reduced wavelengths, their size can be much smaller than the free-space wavelength [31–34]. For this reason, optical antennas can be used to further thin optical elements that rely on manipulating propagation phase, which includes dielectric geometric-phase-elements [27]. The resulting flat optical element can be treated as 2D and appear to cause a discontinuity in the phase of transmitted and reflected light waves. This brought about the notion that the optical properties of a nanopatterned surface of a material can be as important as their bulk optical properties (i.e. the refractive index) and led to the formulation of generalized laws of refraction [25, 35] and the term metasurface [1, 5]. Taxonomy is a dynamic science and nowadays, the concept of metasurface-based optics and flat optics are often used interchangeably. Independent of the nomenclature, it is important to realize that the transmission, reflection, absorption, and polarization conversion coefficients of ultrathin metasurfaces satisfy a set of well defined fundamental relations [36, 37] that can assist in metasurface design thinking. One challenge with simple resonant antennas is that their Lorentzian polarizability only offers a limited 0 to π phase pickup. This spurred new ideas on Kerker resonators with two degenerate resonances [28, 38] and overcoupled gap plasmon resonators on a reflective substrate [39–41] capable of producing phase pickups across a full 2π range. Another challenge with antennas is that the light scattering properties (amplitude and phase) are strongly wavelength dependent. However, with new computational inverse and topological design techniques [42–44] the dispersive nature of nanoscale antennas can also be an invaluable asset in creating high-efficiency and virtually aberration-free metasurfaces [45].

The various concepts above have been instrumental in realizing small-form-factor optics delivering multi-functionality [46–49], very high-numerical apertures [50, 51], meaningful integration with conventional optics [52], minimal aberrations [53–56], nonlinear optics [57, 58], 3D and holograms [59–62], and control over the light field [63–65]. Given the extreme complexity of the current elements, it is good to note that there are still some very basic needs and challenges with even the simplest flat optical elements such as a lens. For example, while it is possible to supply a lens with a broadband antireflection coating, it is very challenging to avoid reflections from high numerical (N.A.) flat optics. This makes it hard to cascade them into multicomponent systems while avoiding ghost images and most likely a blend of flat and conventional optics can provide the highest performance.

2 Metasurfaces in optoelectronic devices

Concurrent with the development of flat optics for passive wavefront manipulation, it has also been realized that the layers in thin film optoelectronic devices can be nanostructured to enhance, manipulate, and control light absorption and emission functions. In such devices, metasurfaces and metafilms are serving as sources or terminals for light and are not used to transform light waves by manipulating scattering and interference phenomena. The typical questions about imaging aberrations are shifted to new questions pertaining to the best ways to facilitate and control light emission or absorption. In current semiconductor devices, the critical dimensions are already at the nanoscale and it is important to co-optimize the size and shape of the nanostructures to achieve the best electronic and (resonant) optical properties. For example, in today's optoelectronic devices the metallic electrodes typically only serve as electrical leads. However, with a simple patterning step they can also increase light absorption in solar cells and detectors [66, 67], enable polarimetry [22, 23, 68] and spectral filtering functions [69–71], or prevent radiative decay of quantum emitters into lossy plasmonic modes in solid state light sources [72]. The semiconductor layers in photodetection systems can also be structured at a subwavelength scale to create metafilms with reengineered absorption spectra [73–77], structural color [78–80], valuable color sorting functions at the nanoscale [81], or achieve an angle-sensitive response [82]. Metasurfaces of Mie and plasmonic resonators also enabled the design of conceptually new types of high-performance antireflection coatings [66, 83–85]. New research further shows that active light emitting layers can be patterned at the subwavelength scale to enhance and direct light emission [86–89]. This is an area where commercialization of metasurface concepts can be possibly fastest as the device infrastructure for realizing nanostructured optoelectronic devices is already very mature. Existing, commercially viable technologies may be re-envisioned by exploring whether minor, extra patterning steps in a process can yield tremendous optical performance benefits.

3 Open challenges and opportunities for metasurfaces

Despite the many impressive advances, there remain a great number of exciting open challenges and opportunities for further development and research. To a large extent, these opportunities are brought to us by emerging application areas for metasurfaces. These include computational imaging, dynamic wavefront manipulation, ultrafast spatiotemporal control of light, quantum communication, thermal emission management, novel display technologies, and sensing. It is of value to analyze these areas and pinpoint the specific new functionalities that they require.

It is the focus of the computational imaging field to extract high-dimensional data from images and transform it into useful numerical or symbolic information that can be further processed, interpreted and used in decision-making [90]. With advances in deep learning with artificial neural networks, digital computers are now able to analyze images with a logic structure that is similar to how humans think [91, 92]. However, there are many imaging applications that require high-throughput, real-time, and low power image processing for which digital electronics is not ideally suited. To enable such applications, it is possible to off-load certain critical and computationally intensive tasks to passive, nonenergy consuming, and ultrafast flat and deep optics [93–101]. Next-generation computational imaging systems may therefore include a series of deep and flat optical layers that need to be easily stackable and have a series of very demanding linear, nonlocal and nonlinear optical properties. When properly designed, basic image processing tasks can be performed simply as light flows through these stacks.

A number of emerging metasurface applications need to deliver dynamic wavefront control, such as light detection and ranging (LIDAR) and dynamic holography [102, 103]. This requires the design of optical antennas capable producing very large changes in their resonant light scattering response. Effective ways to manipulate resonances are currently applying mechanical motion [104–109], electrical gating to modify carrier concentrations [110–116] or the Stark effect [117], electrochemistry [118–122], liquid crystals [123–125], and phase change materials [126–130].

Metasurfaces with ultrafast responses also show great promise as it is becoming eminently clear that such surfaces are not bound by the fundamental limits of static elements [131–133] and novel incarnations of effective medium theories are required to describe their behavior [134]. They can be applied to break Lorentz reciprocity [135], attain frequency conversion [136, 137], and new opportunities for wavefront manipulation [138]. For this reason, new materials need to be identified that are easily incorporated in metasurfaces and can offer very strong, tunable, and ultrafast light-matter interactions.

Nanophotonics and quantum have been a natural match as one of the key strengths of nanophotonic elements is to concentrate light and enhance light–matter interaction for quantum objects [139–141] and overcome quantum decoherence [142]. Now, new types of metasurfaces are being developed that can control quantum properties of emitted, transmitted, and reflected light [143–148]. Further development requires emitting materials with large oscillator strengths and related radiative decay rates that well exceed nonradiative and pure dephasing decay rates.

Thermal emission from objects tends to be spectrally broadband, unpolarized, and temporally invariant. These common notions are now challenged with the emergence of new nanophotonic structures and metasurface concepts that afford on-demand, active manipulation of the thermal emission process [149–156]. The thermal emission spectra of metasurfaces, which are directly connected to their spectral absorption properties through Kirchhoff's law [157], can be engineered with tremendous flexibility through nanostructuring. This ability can impact diverse application areas, including solid state lighting, sensing, thermal imaging, thermophotovoltaics, and personal/commercial heat management [158]. However, further progress will be reliant on the availability of ultrathin elements that can easily be patterned and exhibit strong, electrically tunable absorption resonances.

For display applications, especially in wearables we need to identify lightweight materials that can be grown over large areas and easily be incorporated/patterned into metasurfaces. Materials and structures need to be identified that can enhance and control emission for displays with extreme pixel densities exceeding 10^4 pixels per square inch [159]. Wavefront manipulating optics needs to be developed that can handle the light field at wavelengths across the visible range and can assist in reducing aberrations in imaging systems comprised of miniature projectors and multiple cascaded optical components.

For application in chemical, biological, and environmental sensing, it would be very helpful to identify flat optical elements that deliver high-quality-factor (high-Q) optical resonances that can enhance light–matter interaction to e.g., increase sensitivity to subtle changes in refractive index or produce strong Raman signals. Several structures have recently been pursued in this direction that rely on the creation of bound states in the continuum, breaking symmetries in highly symmetric resonators, or the excitation of quasi-guided modes [11, 160–164].

4 2D, or not 2D, that is the question

In the design and optimization of next generation metasurfaces, it is important to realize that their ultimate physical and practical limitations can always be traced back to the properties of the materials and building blocks that the metasurfaces are constructed from. The current metasurface scene is dominated by truncated waveguides and plasmonic- or Mie-resonant antennas. The latter two can afford strong scattering and absorption with the relevant cross-sections often exceeding the geometric cross sections [165, 166]. The emerging metasurface applications, however, demand much more. From the aforementioned discussion, it is clear that we need metasurface materials and building blocks that are easily grown and patterned over large areas, straightforward to stack, and offer strong, tunable, nonlinear, ultrafast optical responses. The current building blocks can be hard to accurately place and stack over larger areas and into complex 3D architectures. Their tunability also tends to be very limited as the magnitude of most electroabsorption and electrorefraction effects in bulk (3D) metals and semiconductors are very weak [167]. In thinking what the ideal tunable, compact, stackable metasurfaces may look like, it is worth investigating the possibility of creating metasurfaces from atomically thin materials.

Concurrently with the prolific developments on metasurfaces, there has been incredible progress in the synthesis, handling, and understanding of 2D van der Waals (vdW) materials as is detailed in several comprehensive review articles [168–170]. The ability to realize these atomically-thin layers with wafer-scale uniformity is demonstrating their commercial potential [171, 172]. These materials also display a fascinating diversity of quantum, collective, topological, nonlinear, and ultrafast behaviors. It is exciting to think how such materials may open up new functions for metasurfaces. They come in the form of atomically thin sheets of insulators (e.g., hexagonal boron nitride), semiconductors (transition metal dichalcogenides [TMDCs], such as molybdenum disulphide) and semimetals (e.g., graphene). Some hard-to-realize properties in metamaterials also appear naturally in the 2D materials. For example, hyperbolic dispersion [173] and negative refraction [174] can be attained in graphite materials. Weak vdW forces bind these sheets together and this facilitates facile separation and stacking [175] to create heterojunctions and complex multilayer systems [176]. As their surfaces are naturally passivated without dangling bonds, they can also be easily integrated with other electronic and photonic elements. For these reasons, the opportunities for incorporating them in nanophotonic devices seem virtually limitless [177–179].

The suitability of 2D materials for incorporation into flat optics is in part founded on their natural ability to provide strong light–matter interaction. For example, due to its unique band structure, the absorption of graphene can be linked to the fine structure constant $\alpha_0 \approx 1/137$ as $\pi\alpha_0 = 2.3\%$ across a wide spectral range [180]. As doped graphene supports long-lived plasmons [181, 182], the absorption can be further increased to near-unity values by patterning a graphene layer [183]. In 2D semiconductors, the resonant excitation of excitons, electron-hole pairs bound by the Coulomb force, can also give rise to sharply peaked absorption features, just below the bandgap of a semiconductor [184]. A number of 2D semiconductors exhibit very high (~10%) absorption by a single layer near excitonic resonances, well exceeding that of many 3D semiconductors [185, 186]. The reduced screening in these 2D materials leads to exciton binding energies of hundreds of meV and they can consequently be harnessed in nanophotonic devices operating at room temperature [187–190]. Polar 2D dielectric materials, such as hBN, support strong optical phonon resonances with long lifetimes. As a result, they can support low-loss and highly confined phonon-polariton modes [191, 192] and can also be patterned to create low-loss metasurface building blocks.

The 2D materials also offer impressive tunability with various external stimuli. Graphene's optical properties can easily be altered by changing the Fermi energy through chemical or electrostatic gating, which is significantly more challenging in conventional metals and semiconductors. Black phosphorus, an attractive material for mid-infrared optoelectronics, also offers notable tunability of its optical properties with electrical gating [193–195] and affords ultrafast, dynamic polarization-dependent optical responses [196]. Many years of research on electro-optical modulation, also indicate that the strongest, high-speed modulation of materials optical properties is achieved by manipulating excitons [197]. Exciton resonances can also effectively be tuned over several 100 meV with changes in the materials composition, environmental index [198–200], electric/magnetic fields [198, 201], and strain [202, 203]. The suppression of exciton states through carrier injection [204–206] can have an even stronger impact on the material optical properties. Effective electrical modulation in graphene [207, 208] and 2D semiconductors materials [209] has already been demonstrated at speeds exceeding tens of GHz. Given the very high carrier mobility of graphene at 200,000 $cm^2/(V \cdot s)$ at room temperature and picosecond photocarrier relaxation processes, there is significant room for much higher speeds and ultrafast optical modulation [210]. The rapid ongoing improvements in the growth, processing, and encapsulation of high-quality 2D materials is certain to also further improve the tuning performance.

The noted superior electronic and highly tunable optical properties of 2D materials raise the intriguing question whether they can be used to create dynamic and atomically thin metasurface components. The creation of metasurfaces from graphene first arose from the question what good conductors are for plasmonics [211]. Since then a wide range of metasurfaces with graphene strips, discs, split-ring-resonators, and cones have been developed for the mid-IR and THz ranges [212]. They have demonstrated possibilities to achieve unity absorption, cloaking, polarization control, wavefront manipulation, and been applied to quantum communication, nonlinear optics, and sensing [179, 212]. Based on recent measurements showing near-unity reflectivity from monolayers of $MoSe_2$ [213, 214], it is clear that the interaction with single layer 2D semiconductors can also be extremely strong. Given the strong light–matter interaction, a variety of passive optical elements has already been realized by patterning multilayer 2D TMDCs [215, 216], multilayer graphene [217], 200-nm-thick graphene oxide [218], and even monolayer TMDCs [219]. Complex patterning and integration of graphene with subwavelength cavities have also been pursued to create metasurface devices capable of performing dynamic functions, such as beamsteering [220]. Most recently, active modulation of atomically thin WS_2 lenses has been demonstrated by turning "on" and "off" excitonic resonances in the visible spectral range by electrical gating [148]. This work is now opening the door to create electrically-tunable flat optics for the shorter wavelengths, where phonon resonances in polar dielectrics and plasmon resonances in graphene are not accessible.

Higher diffraction efficiencies and new functions for flat optics based on 2D materials may become achievable by stacking. The 2D materials are easily stacked to create metamaterials with new engineered properties that go beyond those of the individual building blocks. The atomically thin layers are certainly much smaller than the relevant wavelength of light, suggesting that standard homogenization should be allowed to guide the design. However, one should keep in mind the possible strong interactions (some not electromagnetic) between the different layers that can modify the optical properties of the constituent layers. The transdimensional properties of few-layers systems could prove to be a rich ground for discovery by itself [221, 222]. Stacking has already been used to create negative index materials [223], one of the major milestones in metamaterials. Heterostructures can also support highly confined plasmons that facilitate creation of metasurface elements that offer single photon nonlinearities due to the extraordinarily strong light–matter interaction [224].

5 The imminent fusion of 2D metasurface optics and 2D vdW metasurfaces

Achieving high efficiencies for manipulating light with 2D materials is challenging, as the interaction length/time with free-space optical beams is short. Smart geometries, such attenuated total internal reflection setups [225] and integration with cavities [226–228] has been pursued to access strong light–matter coupling physics [229–231]. Plasmonic [232–237] and Mie-resonant [238–240] antennas as well as metasurfaces [241, 242] can also naturally be integrated with 2D materials of the same dimensionality to enhance light–matter interactions without notably altering the properties of the 2D materials. This has been successfully pursued to enhance light absorption, scattering, and emission phenomena [234], whereas conventional metasurfaces can help improve the performance and add functionality to 2D materials, and the reverse has also proven to be extremely valuable. For example, the electrically tunable properties of graphene have been used to activate otherwise-passive plasmonic antennas and metasurfaces [116, 243, 244]. It is clear that there will be many beneficial synergies in bringing together the two classes of 2D materials.

6 Conclusions and outlook

A brief look at the history and tremendous opportunities for flat optics, paint a bright future for the field. The value of combining the rich physics of 2D materials with the design flexibility of 2D metasurfaces is already clearly evident. Much research will be needed on how to best integrate these distinct material classes to allow easy fabrication and maximum synergy. It is exciting to watch the growing importance and diversity in optical resonances that are being used in flat optics, starting from the geometry-controlled plasmon and Mie resonances to materials-based exciton and phonon resonances. As flat optics continues to get flatter, it appears that there is still plenty of room at the bottom.

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