Flat Optics With Metasurfaces and Their Applications

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Flat Optics with Metasurfaces and Their Applications

ABSTRACT

At the heart of numerous developments in contemporary optics and photonics is the progressive understanding of the wave-matter interaction and our ability to manipulate or control it. This has in turn been largely driven by the discovery and use of metamaterials: synthetic, man-made materials whose constituent building blocks are engineered at a size scale similar to the characteristic wavelength of the system or phenomenon being studied. As such, these metamaterials often possess exotic properties that go beyond conventional, naturally occurring materials. However, their fabrication and large-scale reproduction have proven to be prohibitively challenging. Metasurfaces are two-dimensional analogs of metamaterials and are consequently much more compatible with standard industry fabrication processes, such as those commonly used in the electronics and semiconductor industries, while still expanding on many of the functions of conventional materials. This thesis presents how, in an optical context, single layer metasurfaces can perform at a level similar to their traditional refractive or diffractive counterparts in a variety of imaging and spectroscopic applications. More importantly, it details how metasurfaces can outperform these conventional optical elements by incorporating advanced dispersion engineering techniques in the design process, resulting in devices with a highly compact form-factor capable of performing sophisticated functions such as aberration control, broadband achromaticity and polarization management.
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TO MY FAMILY, FRIENDS, AND ALL WHO HAVE STOOD BY ME
MY PH.D. JOURNEY STARTED OUT DIFFERENT FROM MOST: IT WAS NEVER REALLY A GOAL OF MINE TO attend graduate school, to spend years working on esoteric projects that may or may not in the end extend the boundary of human knowledge by an infinitesimal amount. On the contrary, I have always thought of myself as a hands-on person who was more suited to working on application-focused projects and bringing them to fruition than pondering about abstract academic problems.

Of course, this was a false dichotomy. I later came to realize that in many cases the two cases were intertwined: sometimes the former necessitated the latter; in other cases, the latter led to the former. I would like to express my heartfelt thanks to Professor Ertugrul Cubukcu, who admitted me to his research group at the University of Pennsylvania when I was but a lowly undergraduate who knew next to nothing about nanophotonics. He showed a faith in me, in my ability to learn quickly and to make things work, that often exceeded my own expectations of myself. By treating me as an intellectual equal, Prof. Cubukcu gave me the opportunity to experience firsthand what it was like to perform research right at the cutting edge of science. I was not sidelined by typical first-year undergraduate projects; instead, I was entrusted with full-fledged inquiries funded by multi-year grant proposals, whose success was very much in doubt and hinged upon the perseverance and ingenuity of the student involved. I learnt invaluable lessons from these experiences and I am forever indebted to him.

Even so, I was not completely sold on attending a Ph.D. upon completion of my Bachelor’s degree. Many research groups I talked to gave the impression that the student was but a small cog in a big machine, there to serve as a pair of helping hands for several years before moving on with his or her life, without perhaps ever seeing the complete, big picture and appreciating the fruits of their labor.
One stood out, however: the group of Professor Federico Capasso at Harvard. In his own words, “you might be smart individuals, but I am here to train you to become professionals. I am here to teach you what it means to be a scientist.” To me, this is what a Ph.D. education is all about: learning to be independent, develop new lines of inquiry and see it through with the highest standards of rigor and transparency. I count myself incredibly lucky to have met Federico and have him as my mentor. His passion for science, education, and above all, sheer professionalism have left indelible marks on not just me, but also generations of students before me; I know Federico will similarly influence many more future batches of eager young minds.

Within the Capasso group, several individuals have contributed immeasurably to making my stay a memorable and transformative experience. I would like to thank Dr. Reza Khorasaninejad for not only mentoring and guiding me when I was still new to the group, but also being a role model in research and in life. He is one of the most passionate and energetic scientists I know, and he has been a key source of motivation for me in my early years at Harvard.

My heartfelt thanks goes out to Dr. Wei Ting Chen, whose patience and meticulousness in conducting scientific experiments are truly admirable. He set the standards to which I constantly aspire to, and he has shown time and again that sheer force of will does allow us to overcome many of the difficulties we face today, be it in research or in life. He has also been a great friend and I treasure the time we spent together.

I would also like to acknowledge other members of the Capasso group, both past and present, who have on multiple occasions acted as a sounding board for my ideas, or contributed to research projects I have been involved in. I thank Dr. Yao-Wei Huang, for being a partner in the cleanroom during all those late nights when there was too much to do and too little time to do it. His innate optimism and cheerful nature has helped immeasurably in getting through numerous projects and tough deadlines. There is Noah Rubin, who although also graduated from Penn (we even took multiple classes together!), could not be more dissimilar from me in terms of how we worked and our approach to research (and possibly to life too). Nevertheless, he is a deep intellectual and I very much respect
that. Then there is Zhujun Shi, who saw the light and joined our group after a year of laboratory rotations and is one of the smartest and most creative people I have met at Harvard. I am proud that I had to chance to recommend her to Federico, and I know that she will go on to achieve much in life. Shoutouts also go to Joon-Suh Park for his expertise in nanofabrication and unswerving dedication to work and attention to detail; Jaewon Oh, whose easygoing and carefree demeanor belied an inquisitive mind; Jared Sisler, who worked so meticulously and diligently as an intern in our group that he received multiple offers from top schools in the country for graduate study; Dr. Michele Tamagnone, for being an invaluable resource when I needed it (and also for a seemingly endless supply of gummy bears); Aun Zaidi, for never ceasing to amaze me in what he could do even back when he was an undergrad, and also Dr. Steve Byrnes, Dr. Xinghui Yin, Dr. Fei Yi, Dr. Jason Reed, Dr. Patrice Genevet, Dr. Antonio Ambrosio, Daniel Wintz, Kundan, Rob, George Guo and many others.

It is often said that the people who are the closest to you, are the ones who are most easily overlooked. I would like to take this opportunity to thank my parents, who have in their own ways stood by me all these years, despite their sometimes having (well-founded) misgivings about my decisions. I am grateful to them and I could not have asked for anything more.

I would like to say a special thank you to Jeeun Lee, who has become the most important person of my life.

Last but not least, I would like to thank my thesis committee members, Professor Marko Loncar and Professor Evelyn Hu, for their invaluable comments and suggestions.
1

Introduction

1.1 Optical metasurfaces

The quest to understand and attain greater control over our physical surroundings often boils down to the availability of the right materials. In ancient times, each important epoch in human civilization was named for the defining material for that era, such as stone, bronze and iron. Today, the discovery of semiconductors has laid the foundation for the entire electronics industry; advancements in glass making have made possible optical fiber cables without which there would be no Internet, and so on.
In nature, the repertoire of available materials is often limited by fundamental chemical or physical properties. Metamaterials offer a tantalizing possibility to overcome these limitations, as they are comprised of artificially engineered constituent building blocks that are of a similar size scale to the characteristic wavelength of the phenomenon being studied. As such the wave-matter interaction is enhanced, which allows metamaterials to possess exotic properties surpassing that of conventional materials\textsuperscript{5-21}.

In optics, this is especially relevant since light is one of the main ways we perceive and make sense of the world around us. For example, in addition to seeing objects in the visible spectrum using the naked eye, humans use long wavelength infrared light, X-rays and high energy γ-rays to study elementary particles\textsuperscript{22}, explore solid-state physics\textsuperscript{23} and even probe distant galaxies\textsuperscript{24}. In all these applications it is necessary to be able to shape the wavefront of light and control its spatial distribution, or otherwise manipulate the degrees of freedom of an electromagnetic ray.

Recently, much attention has been devoted to the study of optical metasurfaces (hereafter referred to as simply metasurfaces), which are the two-dimensional analogs of metamaterials. These are comprised of arrays of subwavelength scale scatterers whose optical properties are carefully tailored to the task they are required to perform\textsuperscript{25-83}. These are much more amenable to large scale fabrication with standard semiconductor industry processes\textsuperscript{84} by virtue of the fact that they are single-layer devices; in many ways, one can imagine them as a digitized, single-mask version of analog optical components\textsuperscript{85}.

From a theoretical perspective, metasurfaces are significantly more challenging to treat than conventional bulk materials or even metamaterials. Natural materials can be described by their bulk dielectric permittivity $\varepsilon$ and magnetic permeability $\mu$ values\textsuperscript{86-87}, while effective medium theory can be used to extract similar bulk parameters for most metamaterials\textsuperscript{88-90}. For metasurfaces which possess a reduced dimensionality, however, effective medium theory does not always apply. Instead, they can be more rigorously modeled by using a generalized version of the electromagnetic boundary conditions, sometimes known as the generalized sheet transition conditions (GSTCs)\textsuperscript{91-94}.
Figure 1.1: (a) A generalized metasurface, where individual scatterers are substituted by their effective electric and magnetic surface susceptibilities. GSTCs enable one to model and predict rapid changes in electromagnetic fields across a metasurface. (b) The metasurface allows one to change the behavior of light abruptly at an interface. (c) A gradient metasurface consisting of an ensemble of subwavelength optical resonators. Here the paraxial approximation has been made and the metasurface was designed with only phase as the key variable. (d) Such an ensemble of scatterers will also result in generalized reflection and refraction of light, due to the local phase gradient.

\[ \hat{z} \times \Delta \mathbf{H} = j \omega \mathbf{P}_\parallel - \hat{z} \times \nabla \mathbf{M}_z \]  
\[ \Delta \mathbf{E} \times \hat{z} = j \omega \mu \mathbf{M}_\parallel - \nabla \mathbf{M}_z \left( \frac{P_z}{\varepsilon} \right) \times \hat{z} \]  
\[ \hat{z} \cdot \Delta \mathbf{D} = -\nabla \cdot \mathbf{P}_\parallel \]  
\[ \hat{z} \cdot \Delta \mathbf{B} = -\mu \nabla \cdot \mathbf{M}_\parallel \]  

Here \( \Delta \) represents the difference between the fields on the two sides of the interface (or metasurface), and \( \mathbf{P} \) and \( \mathbf{M} \) are the surface electric and magnetic polarization densities, which are in turn obtained from local electric and magnetic induced dipole moments in the plane. In order to express the GSTCs using macroscopic quantities, one needs to use the surface susceptibility tensor (a six by six quantity).
to relate the induced currents and fields on the metasurface, accounting for co- and cross-polarization
coupling between electric and magnetic induced currents\textsuperscript{91-93}:

\begin{align}
\mathbf{P} &= \varepsilon \hat{\chi}_{ee} \mathbf{E}_{av} + \hat{\chi}_{em} \sqrt{\mu \varepsilon} \mathbf{H}_{av} \quad (1.5) \\
\mathbf{M} &= \hat{\chi}_{mm} \mathbf{H}_{av} + \hat{\chi}_{me} \sqrt{\mu} \mathbf{E}_{av} \quad (1.6)
\end{align}

Here $\mathbf{E}_{av}$ and $\mathbf{H}_{av}$ denote the average fields. A schematic illustration is provided in Fig. 1.1. It is clear
that designing metasurfaces to satisfy Eq. (1.5) and (1.6) is a tremendous task, due to the large number
of degrees of freedom inherent in the system and the non-trivial nature of accurately extracting the
susceptibility tensor in simulation models.

Practically, however, one can neglect the longitudinal field components and consider only the
transverse electric and magnetic fields acting on a metasurface. This is similar to making a paraxial
approximation, which applies when the angles of incidence and reflection/refraction for the
metasurface are small\textsuperscript{95}. Empirically it has been found that the approximation holds even at reasonably
large angles of approximately 60 degrees\textsuperscript{96}. In this case, the local electric fields can be characterized by
their phase and amplitude for each (orthogonal) polarization state. The metasurface then reduces to an
interface with a spatially varying phase and amplitude profile, and is sometimes referred to as a
“gradient metasurface”\textsuperscript{27,82,95}. Although this concept arose from a simplification of the susceptibility
tensor, it is worth considering its implications in greater detail.

It is known from Fermat’s Principle that light always travels the extremum path\textsuperscript{87,97}; in other
words, this path is stationary to perturbations, such as the introduction of a slowly varying, non-zero
phase gradient due to a metasurface\textsuperscript{98}. The difference in total accumulated phase of the paths travelled
by the light with and without the perturbation is zero. This understanding leads to the generalized
Snell’s law of refraction\textsuperscript{82}:

\[ n_t \sin \theta_t - n_i \sin \theta_i = \frac{\lambda_0 \frac{\partial \phi}{\partial x}}{2\pi} \quad (1.7) \]

where $\theta_i$ and $\theta_t$ denote the incident and transmitted angles respectively, $n_i$ and $n_t$ are the refractive
indices of the respective surrounding medium, and $\lambda_0$ is the vacuum wavelength. An illustration is
provided in Fig. 1.1(c) and (d). Essentially, light is bent abruptly at the interface due to the presence of the local phase gradient in order to preserve the property of travelling along a stationary path. Intuitively, one can interpret this result as a statement of conservation of momentum: a phase gradient of the form $\frac{\partial \Phi}{\partial x}$ imparts a non-zero transverse momentum to the impinging light, whose resultant wavevector is consequently modified. The light therefore travels in a different direction dictated by the new wavevector. To take this concept a step further, by imparting a spatially varying phase gradient whose exact form depends on the application in question, and introducing additional degrees of freedom such as amplitude and polarization variations, one can realize a host of new and intrinsically flat optical devices using metasurfaces such as phase plates\textsuperscript{35,99-101}, collimators\textsuperscript{102-103} and flat lenses for diverse applications ranging from holography to displays\textsuperscript{33,73,104-108}. These devices generally possess a highly compact form factor, are able to perform targeted wavefront shaping and even accommodate multiple disparate functions without increasing spatial footprint.

1.2 Design principles of metasurface devices

The general design procedure for a metasurface device is shown in Fig. 1.2. In general, the phase profile of the metasurface is the variable of interest since it dictates the direction light is travelling towards, and is responsible for interference effects which tend to dominate a given optical response. The design procedure begins with the calculation of the target phase profile $\Phi(x, y)$ at a given design wavelength $\lambda_d$. $x$ and $y$ represent the spatial coordinates along the surface of the device. As previously mentioned in Chapter 1.1, the goal is to impart a spatially dependent phase delay to impinging light in order to shape the output wavefront. For example, in the case of a singlet metasurface lens (hereafter referred to as a metalens), the phase function can be written\textsuperscript{30}:

$$\Phi(x, y) = -\frac{2\pi}{\lambda_d} \left( \sqrt{x^2 + y^2 + f^2} - f \right)$$

where $f$ is the focal length of the lens. This arises from a simple geometric derivation where light rays
originating from all over the metalens need to meet and interfere constructively at the focus, as illustrated in Fig. 1.2(a). The resultant hyperbolic phase profile provides a decreasing phase delay from the center of the metalens to its edge, which exactly compensates for the difference in optical path lengths for rays originating at each location of the metalens.

For more complicated optical functions, such as a doublet lens for an increased field of view or a spectrometer, many other variables must be considered, such as the correction of coma, distortion, astigmatism and field curvature. Consequently, it becomes impractical to write down an analytical expression for the target phase profile. More advanced algorithms and various wave-optics or ray-tracing software can be used to obtain numerical approximations of the optimal phase profile using a customized figure of merit, such as minimizing the wavefront aberration function (defined as the optical path difference between the actual output wavefront versus an ideal spherical wavefront).

The next step is to build up a library of nanostructures together with their corresponding phases (and if necessary, other properties such as amplitudes, polarization behavior etc.) using full-wave simulation solvers. There are several different approaches to imparting a desired phase using a nanostructure, two of which are shown in Fig. 1.2(b): one approach is to make use of optical resonances, which can occur in both metallic nanoparticles (known as localized surface plasmon resonances) as well as dielectric ones (Mie resonance)\(^40,109-110\). Note that in this case, one needs a minimum of two distinct resonances whose resonance wavelengths are in close proximity, in order to achieve full \(2\pi\) phase coverage of the incident light. This can be achieved in many ways, such as metallic V-shaped antennas\(^28,82\), split ring resonators\(^111\), high-index dielectric nanoparticles of varying geometries\(^40,109-110\) and more. Another approach is to utilize structural birefringence and the geometric phase: by using appropriately designed anisotropic nanostructures, such as rectangular nanofins which possess a different effective refractive index along the long and short axes, one can locally achieve the behavior of a half-waveplate\(^30,112\). When these elements are rotated successively, the handedness of incident circularly polarized light can be converted and a net phase imparted to the cross-polarized component; this is known as the Pancharatnam-Berry phase or geometric phase\(^113-114\). This will be
Figure 1.2: (a) The target phase profile is obtained via an analytical expression in closed form or via series expansions derived from raytracing software. The example of the metalens singlet shows how its phase profile $\Phi(r)$ can be simply calculated by
$$\Phi(r) + \frac{2\pi}{\lambda d} r^2 + f^2 = \Phi(0) + \frac{2\pi}{\lambda d} f.$$ The equal sign implies that the rays from different lens coordinate $r$ have to arrive at the focus in phase. $\Phi(0)$ can be assumed to be zero as phase is relative. The phase profile for the bottom two cases (doublet metalens and metasurface spectrometer) becomes more complicated, especially since the phase profile is not symmetric for the metasurface spectrometer. (b) A library of nanostructures with their corresponding phases and higher-order dispersion terms is established via full-wave electromagnetic solvers. The first subplot shows the transmitted amplitude and phase of nanofins with various rotation angles under left-handed circularly polarized light incidence. The second subplot shows how the phase can be controlled by varying the diameters of nanopillars. (c) The phases of elements from the library is matched to the target phase via optimization algorithms to determine the best overall fit for all elements across the metasurface. The insets show the top views of two metalenses implemented based on geometric (top) and propagation (bottom) phases, respectively.

explained in greater detail in Chapter 2. A third approach (Fig. 1.2(b), bottom panel) would be to use isotropic nanostructures, such as circular disks or pillars, where a portion of the incident light is coupled to the fundamental optical mode of the structure. Essentially, the nanostructure behaves as a truncated dielectric waveguide; the phase acquired by the light upon exiting the nanostructure will be given by the product of the wavevector, effective mode index, and the height of the nanostructure.

The effective mode index, and thus the phase, can be continuously tuned within a certain range by varying the geometrical dimensions of the nanostructure.

All the methods listed above have their own advantages and disadvantages, such as polarization.
dependency, amplitude modulation and tolerance for nanofabrication imperfections. The onus falls upon the designer to identify the most suitable nanostructures for a particular application. For example, it may be advantageous to use anisotropic nanofins for high magnification imaging lenses in microscopes, because it would guarantee an accurate phase profile (which is determined by only the rotation of the elements, thereby making it comparatively robust to fabrication errors) with little to no amplitude modulation between the identical nanoscale elements; however, in this case the device would be necessarily sensitive to input circular polarization. For applications where no polarization sensitivity is desired or allowed, a different approach must be taken\textsuperscript{46,54}.

The final step of the metasurface design process is to digitize the target phase and identify a nanostructure from the library whose imparted phase matches the target as closely as possible. Typically this fit needs to be optimized via an algorithm for each and every spatial coordinate of the metasurface device\textsuperscript{45}. Inclusion of additional degrees of freedom (such as higher order derivatives of the phase, amplitude, and polarization) require significantly more computational resources and more efficient algorithms. Fig. 1.2(c) shows two possible layouts of generic metalenses, comprised of circular nanopillars and nanofins. The circular pillars are observed to have larger diameters closer the center of the metalens and become progressively smaller in size radially towards the edges. This behavior repeats itself in “zones” since the target phase is wrapped every $2\pi$. This trend also exists for the geometric phase metalens where the nanofins are rotated periodically. The choice of the geometrical parameters for the nanostructures at each spatial coordinate was carefully optimized and determined by a best-fit algorithm such as the one aforementioned.

It is worth pointing out that in this short overview of the metasurface design process, it is implicitly assumed that the optical properties of each individual nanostructure are derived from illumination occurring at normal incidence, under periodic boundary conditions. As was alluded to in Chapter 1.1, significant discrepancies occur as one goes past the paraxial limit at large angles. Additionally, while the assumption of periodicity implies that the Nyquist sampling criterion can be easily fulfilled (by requiring the sampling rate to be approximately equal to or greater than the
wavelength divided by the largest bending angle) since the inverse of the periodicity is equal to the sampling rate\textsuperscript{50}, this approach fundamentally neglects interactions between neighboring structures. In most practical cases, the errors resulting from this approximation can be minimized by using high-index dielectric nanostructures, where the fundamental electromagnetic mode is tightly confined to within the structure. Another solution is to place constraints on the target phase profile, by requiring that it change slowly with respect to spatial coordinates, and that the geometrical parameters of the nanostructures should similarly not change too rapidly. In cases where both do not apply, care must be taken to include the full interactions into consideration.

1.3 Thesis Outline

The theoretical and experimental results in the following chapters expand upon the concepts introduced in the previous sections. The main theme is to highlight some of the most promising areas of application of metasurface devices. Chapter 2 details metalenses that can perform diffraction limited imaging suitable for high resolution microscopy, at a level similar to commercial microscope objectives. It also presents a metalens capable of imaging under illumination with different helicities of circularly polarized light, something not possible with conventional lenses without adding multiple discrete optical elements. Chapter 3 introduces the concept of an off-axis metalens, and demonstrates how it can lead to ultra-compact miniature spectrometers and with built-in sensitivity to circular polarization. Chapter 4 shows how the control over circular polarization can be further enhanced using planar chiral metasurface structures, something not previously thought possible due to symmetry constraints. Chapter 5 addresses a longstanding criticism of metasurface devices – that they are inherently single wavelength and limited to operation within a narrow bandwidth. It details how one can engineer the dispersion of light in metasurfaces, in addition to common variables such as phase and amplitude, and illustrates how this method can be a potential game-changer by making possible broadband achromatic metalenses, aberration-corrected miniature meta-spectrometers and more.
2.1 Structural birefringence and geometric phase of light

In Chapter 1.2 it was briefly mentioned that one of the ways of implementing a target phase profile was to make use of structurally birefringent elements. In other words, these structures possess twofold rotational symmetry, resulting in different effective refractive indices along their long and short axes. Figure 2.1 illustrates a typical building block used for this approach: a nanofin with a rectangular cross-section.
Figure 2.1: (a) – (e) Schematic and (f) scanning electron micrograph of nanofins used to form a metalens. The nanofins are characterized by their height \( H \), lengths of the long and short axis (\( L \) and \( W \) respectively), a periodicity \( S \) and a rotation angle \( \theta \) with respect to one another. Successive rotations impart a geometric phase to cross-polarized light equal to twice the rotation angle. Scale bar: 300 nm.

In general, light propagating through a rotated nanofin can be written:\(^4\)

\[
\frac{\tilde{t}_{\|} + \tilde{t}_{\perp}}{2} \pm i\frac{\tilde{t}_{\|} - \tilde{t}_{\perp}}{2} \exp(\pm i2\theta) \left( \frac{1}{\mp i} \right)
\]

Here we have used circular polarization basis, denoted using the standard Jones vector notation. \( \tilde{t}_{\|} \) and \( \tilde{t}_{\perp} \) denote the complex transmission coefficients through the long and short axes of the nanofins respectively, while \( \theta \) refers to the rotation angle of the nanofins with respect to one another.

Eq. (2.1) can be most easily understood by considering the incident light to be circularly polarized; the two terms in Eq. (2.1) then clearly correspond to co- and cross-polarized components of the output light. The cross-polarized or converted light acquires opposite handedness and experiences a phase shift of \( \arg \left( \frac{\tilde{t}_{\|} - \tilde{t}_{\perp}}{2} \right) \pm 2\theta \), while the remaining co-polarized light maintains its polarization and experiences a different phase shift of \( \arg \left( \frac{\tilde{t}_{\|} + \tilde{t}_{\perp}}{2} \right) \). The phase delay of \( \pm 2\theta \) arises from the successive rotation of these structurally birefringent elements and is known as geometric or Pancharatnam-Berry phase; it applies only to the cross-polarized light\(^{113-114}\). This can be straightforwardly derived by applying a rotation operation to the Jones matrix of a half-waveplate (i.e. a circular polarization converter)\(^{112}\). As a result, an anisotropic nanofin (or any other structure with two-fold symmetry) can impart a precise phase delay from 0 to \( 2\pi \) purely by performing successive rotations, such as the case
shown in Fig. 2.1(f). The length and width of the nanofin are usually chosen so that the phase difference between $\tilde{r}_l$ and $\tilde{r}_s$ is $\pi$ at the design wavelength $\lambda_d$. In this case it behaves as an ideal half-waveplate and the transmission amplitude of $\frac{\tilde{r}_l-\tilde{r}_s}{2}$ at $\lambda_d$ is maximized, although it falls off at different wavelengths. Note that since identical nanostructures are used throughout the metasurface device, there is no amplitude modulation between the elements, which might lead to interference effects and cause degradation of the image quality. For this reason, this approach is preferred for high-quality imaging applications, when there is no requirement placed on the polarization state of light. The subsequent sections present in detail some of the performance characteristics of metalenses employing this approach of phase implementation.

2.2 High NA diffraction limited microscope objective with a metalens

Typical high numerical aperture (NA) objectives consist of precision engineered compound lenses which make them bulky and expensive, limiting their applications and hindering their integration into compact and cost-effective systems. Singlet planar lenses with high NA in the visible wavelength spectrum are in particularly high demand due to their potential widespread applications in imaging, microscopy, and spectroscopy. Although visible planar lenses can be realized by numerous diffractive components, high NA and high efficiency are usually not attainable simultaneously in these devices because their constituent structures are of a similar size scale as the incident wavelength of light, which precludes an accurate phase profile with sufficient sampling.

The phase profile of a typical metalens, such as the one shown in Fig. 2.1(a), has previously been given by Eq. (1.8). This implies that, for an implementation method using rotated nanofins, each rotation angle is given by

$$\theta(x, y) = -\frac{\pi}{\lambda_d} \left( \sqrt{x^2 + y^2 + f^2} - f \right)$$

(2.2)

i.e. the desired rotation angle is half of the required phase at each point on the metalens.\(^{30}\)
As previously mentioned in Chapter 2.1, in order to maximize the polarization conversion efficiency (and in turn, the overall efficiency of the metalens), the nanofins should operate as half-waveplates: the phase retardance of light transmitted along the long and short axes should be $\pi$. This can be optimized via any Maxwell’s equations solvers. Figure 2.2(a) shows that conversion efficiencies as high as 95% can be achieved for an arbitrary design wavelength via tuning of nanofin parameters. This was possible by using titanium oxide as the material for the nanostructures: it is lossless in the entire visible spectrum and has a sufficiently high refractive index of approximately 2.4 to ensure a high degree of light confinement within the nanofins. Details of the fabrication process are presented in the Appendix A.1115.

In this demonstration, three distinct metalenses with NA = 0.8 corresponding to high magnification microscope objectives were fabricated with design wavelengths of 660 nm, 532 nm and 405 nm respectively. A representative image of the final metalens as seen under an optical microscope is shown in Fig. 2.2(b). Their focal spot profiles were characterized and measured using a custom-built microscopy setup (Appendix A.2); the results are plotted in Fig. 2.3(a)-(c) and (g) to (i). For each metalens, a camera image of the intensity profile at the focal plane is shown, together with a corresponding vertical cut. The full-width-at-half-maximum (FWHM) value, which is indicative of the
 or focal spot size, is obtained from the latter. This is compared against the diffraction limit at each design wavelength \( \frac{\lambda}{2 \times NA} \). Additionally, similar measurements were performed for a commercial microscope objective designed for visible wavelengths (Nikon, NA = 0.8) with the same setup under identical conditions, and the results are presented in Fig. 2.3 (d) – (f) and (j) – (l).

For each respective design wavelength, highly symmetric focal spots were observed for the metalenses. Furthermore, their focal spots were all diffraction limited. The same is not true for the commercial objective, where significant aberrations are observed to occur, particularly at green and blue wavelengths (Fig. 2.3 (e) and (f)). This can be understood because conventional high-NA objectives are designed to image under broadband illumination. As such, wavefront aberrations need to be corrected for multiple wavelengths over a range of angles of incidence to meet industry standards for the required field of view. This is typically implemented by cascading a series of precisely aligned compound lenses. Fabrication imperfections in each individual optical lens and residual aberration errors, particularly spherical aberration, could result in a focal spot size that is larger than theoretical predictions. In contrast, the metalenses were designed to possess a phase profile that is free of spherical aberration under normally incident illumination, which results in a diffraction limited spot at the respective design wavelengths. For example, the theoretical root mean squares of the wave aberration function (WAFRMS) for the metalenses designed for 405, 532 and 660 nm are 0.049\( \lambda \), 0.060\( \lambda \) and 0.064\( \lambda \) respectively. These values are very close to the condition for a perfect spherical wavefront\(^{116} \). The Strehl ratios for these metalenses were also determined from the measured focal spots; they were found to be close to 0.8, consistent with the observed diffraction-limited focusing. The calculations are explained in greater detail in Appendix A.3.

It is important to note that although the metalenses were designed at specific wavelengths, one is still able to observe wavelength-scale focal spots even at wavelengths away from the design. For example, for the metalens designed at \( \lambda_d = 532 \) nm, focal spot sizes of 720 and 590 nm were measured at wavelengths of 660 and 405 nm respectively. This is shown in Fig. 2.4. The broadening of the focal
Figure 2.3: (a)-(c) Measured focal spot intensity profiles of the metalens designed at (a) 660 nm, (b) 532 nm, and (c) 405 nm. (d) – (f) Measured focal spot intensity profiles of the objective (100× Nikon CFI 60, NA = 0.8) at wavelengths of (d) 660 nm, (e) 532 nm, (f) 405 nm. (g) – (i) Corresponding vertical cuts of the metalenses’ focal spots. Metalenses designed at wavelengths of 660, 532 and 405 nm have FWHMs of 450, 375 and 280 nm respectively. The symmetric beam profiles and diffraction limited focal spot sizes are related to the quality of the fabricated metalenses and accuracy of the phase realization. (j) – (l) Corresponding vertical cuts of the focal spots of the objective, at wavelengths of (j) 660 nm, (k) 532 nm and (l) 405 nm. FWHMs of the focal spots are labeled on the plots. These values are ~ 1.5 times as large as those measured for the metalenses.
spots with respect to the theoretical diffraction-limited values arises from chromatic aberrations: metasurfaces are inherently dispersive due to structural dispersion, and the ideal spherical-aberration free phase profile given by Eq. (1.8) has an explicit dependence on the wavelength.

As a result, chromatic aberrations in these metalenses are more pronounced than traditional refractive lenses, which leads to a wavelength dependent focal length. This is generally not an issue for laser-related imaging, microscopy, and spectroscopy because monochromatic sources with narrow linewidths are used. For example, in Raman microscopes/spectrometers, a 532 nm laser with a linewidth of a few picometers is common. In this case, the linewidth-induced broadening of the focal spot size and change in focal length is negligible.
The focusing efficiencies of the metalenses were also characterized and are shown in Fig. 2.5(a). This quantity is defined as the fraction of power contained in a focal spot, normalized to the incident power on the metalens. The metalens designed at $\lambda_d = 660$ nm has a focusing efficiency of 66% and remains above 50% in most of the visible range, while the metalens designed for 532 nm has a peak focusing efficiency of 73%. Additionally, the beam intensity profile of the latter (an $x$-$z$ cross section, following the coordinates in Fig. 2.1) within a 40 $\mu$m span of the focal point is illustrated in Fig. 2.5 (b). The negligible background signal is indicative of excellent phase realization, as the beam converges to a diffraction limited spot, and also shows the high conversion efficiency of each nanofin.

![Figure 2.5](image)

**Figure 2.5:** (a) Measured focusing efficiency of the metalenses designed at wavelengths of 660 nm and 532 nm. (b) Intensity distribution in dB of the $x$-$z$ plane, showing the evolution of the beam from 20 $\mu$m before and after the focus. This measurement was performed on the metalens designed at $\lambda_d = 532$ nm. The wavelength of incident light was 532 nm.

For the metalens designed at 405 nm wavelength, a measured focusing efficiency of 86% was achieved. This measurement is not reflected in Fig. 2.5(a) because it was performed using a monochromatic diode laser (Ondax Inc.); the shortest wavelength of the tunable laser used for the previous measurements (SuperK Varia) was approximately 470 nm.

To demonstrate the use of the metalens for practical imaging, a different metalens with a diameter of 2 mm and focal length of 0.725 mm (NA = 0.8) was fabricated. First, the imaging resolution was characterized using the 1951 United States Air Force (USAF) standard resolution test chart as the target object. The measurement configuration is presented in greater detail in Appendix A.4. The
image formed by the metalens is shown in Fig. 2.6(a). The light source was a tunable laser (SuperK Varia) set at 530 nm with a bandwidth of 5 nm. As the resulting image was larger than the charge-coupled device (CCD) camera, it was projected onto a translucent screen and subsequently photographed with a digital single-lens reflex (DSLR) camera (Canon). The smallest features in the resolution target are lines with widths of 2.2 \( \mu \text{m} \) and center-to-center distances of 4.4 \( \mu \text{m} \) (the bottom element in the highlighted region in Fig. 2.6(a)). A similar image quality can be achieved at wavelengths in the visible spectrum, as shown in Appendix Fig. A.5. Images of the smallest features were taken separately with a CCD camera shown in Fig. 2.6 (b) – (e), at wavelengths of 480, 530, 590, and 620 nm respectively. It is clear that the metalens can resolve these micrometer-sized lines. A similar experiment was repeated using a Siemens star target; it was found that all features can be resolved over the entire visible wavelength range (Appendix Fig. A.6 and A.7). Note that the different magnifications of the images observed at different wavelengths are due to focal length variations as a function of wavelength, since the metalens is chromatic, as discussed previously. In this particular setup corresponding to Appendix Fig. A.4, the tube lens had a focal length of 100 mm, which yields a magnification of 138× at 530 nm (the design wavelength). For wavelengths of 480, 590 and 620 nm, magnifications of 124×, 152× and 167× were obtained respectively by comparing the ratio of the image sizes formed on the CCD camera to the known physical size of the USAF test object.

To characterize the effects of chromatic aberration, the same object at 530 nm was imaged without changing the distance between the metalens and the object, while varying the bandwidth of the source from 10 to 100 nm (which was the limit of the tunable laser). These results are shown in Fig. 2.6(f) – (i). Although the image quality slightly degrades as a result of the increased bandwidth, the smallest features can still be resolved even at the maximum bandwidth of 100 nm. Finally, for a comparison of the imaging quality to that of a conventional objective, a H-shaped object comprising of arrays of holes with gaps of ~ 800 nm was fabricated via focused ion beam (FIB) milling. An SEM micrograph of the object is shown in Fig. 2.6(j). The image formed by the metalens (Fig. 2.6(k)) has
Figure 2.6: (a) Image of 1951 USAF resolution test chart formed by the metalens taken with a DSLR camera. Laser wavelength is set at 530 nm. Scale bar: 40 μm. (b) – (e) Images of the highlighted region in Fig. 4A at wavelengths of (b) 480, (c) 530, (d) 590, and (e) 620 nm. Scale bar: 5 μm. (f) – (i) Images of the highlighted region in (a), at a center wavelength of 530 nm and imaged with different bandwidths: (f) 10, (g) 30, (h) 50, and (i) 100 nm. Scale bar: 5 μm. (j) Nanoscale target prepared via focused ion beam (FIB). The smallest gap between neighboring holes is ~ 800 nm. (k) Image of the target object in (j) formed by the metalens. (l) Image of target object formed by the commercial state-of-the-art objective. Scale bar: 10 μm for (j) – (l). (m) Image formed by the metalens, showing that holes with subwavelength gaps of ~450 nm can be resolved. Scale bar: 500 nm.
comparable quality to the one formed by the 100x Nikon objective (Fig. 2.6(l)) with the same NA = 0.8. The change in image sizes comes from the difference in the magnification of the imaging systems. The resolution limit of the metalens can be also be seen from the fact that four holes with subwavelength spacings of ~ 450 nm between one another can be well resolved (Fig. 2.6(m)). This value agrees with the measured modulation transfer function of the metalens, presented in Appendix A.5 and Fig. A.8.

2.3 Chiral imaging with metalenses

Objects are visible because they scatter light. Decoding spectral and polarization properties of scattered light can reveal valuable information about texture, orientation, and even the constituent materials of an object. Although humans can partially perceive spectral information as color, our eyes are blind to the polarization states. Modern polarization imaging systems require cascading lenses with several optical components such as beamsplitters, polarizers, and waveplates. Polarization-resolved imaging and spectroscopy have versatile applications ranging from remote and environmental sensing to biological studies. For example, resolving chirality is essential for extracting structural and functional properties of biochemical species at both molecular and bulk levels. Besides costly and sizable configurations, cascading optical components also reduces the image quality and spatial resolution. Moreover, adding spectral resolution capabilities to an imaging system mandates the use of a dispersive element, integrated filters, or multiple cameras leading to further complexity and an increased instrument size. Here, we demonstrate a multispectral chiral lens (MCHL) that integrates the functionality of polarization and dispersive optical components into one device. This ultrathin lens overcomes the limitations of bulk optics and simultaneously provides chiral and spectral information across the visible spectrum without the requirement of additional optical components. The device can be utilized for both high-quality imaging and spectroscopy of bulk and microscopic scale specimens where the spatial resolution is limited by the NA of the MCHL.
Figure 2.7: (a) The building block of the MCHL consists of two nanofins on a glass substrate. (b) Top-view of the building block where $S_x = 300$ nm and $S_y = 600$ nm. The blue and green nanofins impart the required phase profile required to focus RCP light and LCP light, respectively. Phase realization is based on the geometric phase by means of nanofin rotation. (c) Top view of a nanofin denoting its length $L = 250$ nm and width $W = 80$ nm. (d) Schematic diagram illustrating the imaging principle of the MCHL where LCP and RCP light from the same object at coordinates $(x_{ob}, y_{ob}, z_{ob}) = (0, 0, -18 \text{ cm})$ are focused into two spots, $(x_{imL}, y_{imL}, z_{imL})$ and $(x_{imR}, y_{imR}, z_{imR})$, respectively. Spiral arrows indicate helicity of incident light. Coordinates of these spots are wavelength dependent due to the dispersive design of the MCHL. For the design wavelength of 530 nm these values are $x_{imL} = -2.34 \text{ mm}$, $y_{imL} = 0.184 \text{ mm}$, $z_{imL} = 29.90 \text{ mm}$, $x_{imR} = 2.34 \text{ mm}$, $y_{imR} = 0.184 \text{ mm}$, and $z_{imR} = 29.90 \text{ mm}$. (f) Top-view SEM image of the fabricated MCHL. Scale bar: 600 nm. The two interlaced arrays of nanofins are false-colored. (g) Side-view SEM image of a portion of the MCHL showing the high aspect-ratio TiO$_2$ nanofins on a glass substrate. The MCHL has a diameter of 3 mm. Scale bar: 600 nm.

The MCHL utilizes a metasurface to achieve chiral imaging. The basic building blocks of the MCHL are comprised of two nanofins shown in blue and green in Fig. 2.7. The required phase for focusing light is imparted based on the Pancharatnam-Berry phase via rotation of nanofins, similar to Section 2.2 (Fig. 2.7(b))$^{30,113-114}$. The focusing efficiency is maximized when each nanofin acts as a half-wave plate, that is, it converts circularly polarized input light into transmitted light with opposite helicity. This is accomplished by suitably designing the nanofin dimensions such as width, length and height (Fig. 2.7(c), (d)). The schematic diagram of the MCHL, consisting of two interlaced arrays of blue and green nanofins on a glass substrate, is shown in Fig. 2.7(e). While the nanofins indicated by the color blue (located at $(x_R, y_R, z_R)$) impart the required phase profile for focusing incident right-
circularly polarized (RCP) light, the nanofins in green (located at \((x_L, y_L, z_L)\)) focus left-circularly polarized (LCP) light. As a result, for an object located at \((x_{ob}, y_{ob}, z_{ob})\) the MCHL forms two side-by-side images with opposite helicity: the RCP image of the object is formed at \((x_{imR}, y_{imR}, z_{imR})\) and the LCP image at \((x_{imL}, y_{imL}, z_{imL})\).

In designing the MCHL, the object is assumed to be a point source emitter with linear polarization, which is equivalent to an equal superposition of both RCP and LCP components. Considering first the RCP component: the center of the MCHL (i.e. origin of the coordinate system) is set as the reference point. The phase delay \(\varphi_d\) due to the difference in the optical path from the object to the image through the nanofin at position \((x_R, y_R, z_R)\) can thus be written as\(^{122}\):

\[
\varphi_d(x_R, y_R, z_R) = \frac{2\pi}{\lambda_d} (\Delta D_{ob} + \Delta D_{im} - f) \tag{2.3}
\]

where

\[
\Delta D_{ob} = \sqrt{(x_R - x_{ob})^2 + (y_R - y_{ob})^2 + (z_R - z_{ob})^2}
\]

\[
\Delta D_{im} = \sqrt{(x_R - x_{imR})^2 + (y_R - y_{imR})^2 + (z_R - z_{imR})^2}
\]

\[
f = f_1 + f_2
\]

\[
f_1 = \sqrt{x_{ob}^2 + y_{ob}^2 + z_{ob}^2}
\]

\[
f_2 = \sqrt{x_{imR}^2 + y_{imR}^2 + z_{imR}^2}
\]

and \(\lambda_d = 530\ \text{nm}\) is the design wavelength. The reference optical path denoted by \(f\) is the distance from the object to the reference point defined previously \((f_1 = 18\ \text{cm})\) plus the distance from the reference point to the image \((f_2 = 3\ \text{cm})\). \(\Delta D_{ob}\) and \(\Delta D_{im}\) are distances between the nanofin at \((x_R, y_R, z_R)\) and the object/image, respectively. In order to form an image, this phase difference \(\varphi_d(x_R, y_R, z_R)\) must be compensated by each nanofin. This is accomplished by rotating the nanofins by an angle equal to exactly half the required phase difference, identical to what was presented in Chapter 2.2. A similar derivation is also valid for the LCP component of the point source, except different arrays of nanofins located at the position \((x_L, y_L, z_L)\) compensate the optical path difference such that the image is formed at \((x_{imL}, y_{imL}, z_{imL})\). The nanofins are also rotated in the opposite direction (i.e. multiplying the previous
Figure 2.8: (a) Simulation results using ray-tracing methods show the MCHL focusing incident light from a point source into a focal spot. The rays in this figure begin after passing through the MCHL. The MCHL focuses different wavelengths into spatially separated spots enabling multispectral imaging. (b) Measured focal length as a function of wavelength. Here, focal length is defined as the distance from the focal spot to the center of the MCHL \((x=0, y=0, z=0)\). (c) Two images formed by the MCHL into the field-of-view of a color camera. The object was the facet of a single mode fiber emitting broadband light linearly polarized by a polarizer. The bandwidth is 100 nm centered at a wavelength of 550 nm. (d) Images formed by the MCHL when the polarization of the emitting light is set to left-circularly polarized by adding a quarter-wave plate after the polarizer. (e) Images formed by the MCHL when the emitting light is switched to right-circularly polarized by adjusting the quarter-wave plate appropriately. The gain and exposure time of the camera were fixed for panels (c) – (e). Also, the camera was saturated at the focal point making it possible to visualize the out of focus colors. For panels (c) – (e), the scale bar is 0.5 mm. (f) Magnified view of the left image in panel (c). Scale bar: 0.150 mm. (g) Measured extinction ratio of the MCHL as a function of wavelength.

results for RCP by negative unity). In these equations, the glass substrate of the MCHL is neglected; in principle this results in slight aberrations that are, however, negligible for the intended demonstration.
The MCHL is fabricated using atomic layer deposition of titanium dioxide, similar to the process used for the high NA metalenses in Chapter 2.2 previously\textsuperscript{115}. More details can be found in the Appendix A.1. Scanning electron microscope (SEM) images of the fabricated lenses are shown in Fig. 2.7(f), (g). The theoretical focusing characteristics of the metalens was studied using ray-tracing techniques, by applying the phase gradient introduced by the nanofins onto the incident rays. Figure 2.8 (a) shows that the MCHL focuses all incident rays of one given circular polarization state (either RCP or LCP) emanating from a point source into a focal point. Here, the position of the focus shifts with changing wavelengths of the incident light, as shown in Fig. 2.8(b). This is associated with the off-axis focusing characteristics of the MCHL, as well as the intrinsic dispersion of metasurface-based lenses. A more complete discussion on off-axis focusing characteristics can be found in Chapter 3.

Experimentally, this feature can be directly verified and observed by imaging the facet of a single mode optical fiber. Figure 2.8 (c) shows how the MCHL simultaneously focuses and disperses a broadband beam emitted from the fiber, resulting in a rainbow-like image. The beam had a center wavelength of 550 nm with a bandwidth of 100 nm. Because of the wavelength dependence of the focal length, the MCHL focuses green light while all other colors are out of focus. This effect is more clearly seen in the magnified image in Fig. 2.8 (f). However, by adjusting the camera position, one can selectively focus onto other wavelengths; this is shown in Appendix A.6 and Figs. A.9 and A.10.

Figure 2.8(c) also shows that for a linearly polarized light source, there are two identical images within the camera’s field-of-view, corresponding to the LCP and RCP components of incident light. It is notable that according to the phase profile of the MCHL, the array of nanofins that focuses one particular helicity will also simultaneously diverge the light with the opposite helicity (virtual image), which contributes to the background noise. However, this effect is negligible as shown in Fig. 2.8(c) and Appendix Fig. A.10, as no higher order diffraction effects nor noticeable background noise was observed. Subsequently, the chiral response of the MCHL is examined by setting the polarization state of the incident light to LCP. As can be seen from Fig. 2.8 (d), the image on the right disappears with a corresponding increase in intensity of the image on the left. The opposite effect was observed by
reversing the helicity of incident light (Fig. 2.8(e)). This chiral response can be quantified by measuring the extinction ratio (ER)\textsuperscript{117}, which is defined as the ratio of optical power of the right (left) image to the left (right) image under RCP (LCP) illumination. Figure 2.8(g) plots the values of the ER measured across the visible spectrum. A high ER of 15 dB together with the engineered dispersive properties of the MCHL makes it possible to perform multispectral chiral imaging.

For chiral imaging across the visible wavelength range, a large biological specimen, *Chrysina gloriosa*, a beetle that naturally exhibits strong circular dichroism (CD) at green wavelengths\textsuperscript{123}, was chosen. The imaging setup is shown in Appendix Fig. A.11. Figure 2.9(a) shows that the MCHL simultaneously forms two images of the beetle on the camera chip with opposite helicity. The very strong CD of the beetle is evident by comparing images of the beetle on the right- and left-handed side, as the beetle’s exoskeleton strongly reflects LCP light while absorbing more RCP light. Images of the beetle under blue and red LED illumination are shown in Fig. 2.9(b) and (c), respectively. The slight background noise partially comes from the lower efficiency of the MCHL at red and blue wavelengths in addition to the lower reflectivity of the beetle in this region. As a control experiment, an achiral object, a one-dollar coin, was also imaged with the same experimental set-up. As expected, Fig. 2.9(d) shows two nearly identical images formed by the MCHL.

Finally, the CD of different body parts of the beetle was measured, using a modified experimental configuration shown in Appendix Fig. A.12. First, laser light was focused onto the beetle’s thorax. The focal spot has a size of \(\sim 100 \mu\text{m}\) to avoid sample damage. The scattered light is captured by the MCHL, which forms two spots with different intensity corresponding to the two circularly polarized states \((I_R \text{ and } I_L)\) on the camera. The CD is calculated by while sweeping the wavelength. This measurement was subsequently repeated on its leg. The results in Fig. 2.9(e) show a large CD which peaks around the green region of the spectrum. In this instance, the MCHL was used to image a chiral beetle but it can be more generally utilized to image a wide range of chiral objects. Ultimately, the spatial resolution is determined by the NA of the MCHL. Here, in order to provide a large field-of-view to image the entire beetle, the chosen focal length was large, resulting in a low NA of 0.05. The
Figure 2.9: (a) The MCHL forms two images of the beetle, *Chrysina gloriosa*, on the color camera. The left image is formed by focusing left-circularly polarized light reflected from the beetle and the right image is from right-circularly polarized light reflected from the beetle. Illumination was provided by green LEDs paired with 10 nm bandpass filter centered at 532 nm. (b), (c) Images formed by the MCHL under (b) blue (10 nm bandpass filter centered at 488 nm) and (c) red LED illumination (10 nm bandpass filter centered at 620 nm). (d) Images of a one-dollar coin under green LED illumination (LED was paired with 10 nm bandpass filter centered at 532 nm). (e) Circular dichroism from two different parts of the beetle as a function of wavelength.
image resolution can be readily improved by reducing the focal length or increasing the diameter of
the MCHL.

2.4 Conclusions

This Chapter has presented several examples of metasurface lenses being applied to practical imaging
applications and shown that they can perform at a sufficiently high level to not only be relevant outside
of an academic laboratory setting, but also be potential game-changers. The visible range metalenses
with NA = 0.8 and efficiencies as high as 86% can provide diffraction-limited focal spots at arbitrary
design wavelengths, which make them ideal devices for use in optical lithography, laser-based
microscopy, and spectroscopy. In addition to having a magnification as high as 170× and being
capable of resolving structures with subwavelength spacing, the intrinsically compact configuration of
these metalenses can enable portable, handheld instruments for many applications. Although the
metalenses in their present form are subject to chromatic aberrations, the latter can be corrected with
approaches such as dispersive phase compensation and other dispersive engineering techniques, to be
further discussed in detail in Chapter 5. This chapter also highlights how chiral imaging capabilities
and spatially resolved chiral spectroscopy can be performed with a single planar lens. Conventionally,
this cannot be achieved using single optical components unless multiples of them are stacked together
in a bulky system. Providing multiple functionalities while retaining a planar and compact geometry is
the key competitive advantage of metalenses, and metasurface devices in general.
Ultra-compact chiral spectrometer

3.1 Off-axis metalenses for spectroscopy

A basic but essential tool for many optical instruments is the spectrometer. The ability to decode changes to the spectrum of light in response to external stimuli is central to applications such as environmental monitoring of pollutants and the detection of specific antibodies in disease diagnostics\textsuperscript{124-125}. Furthermore, advanced techniques such as circular dichroism spectroscopy are widely used to obtain additional circular polarization (CP) information in order to distinguish between optical isomers during the manufacture of many pharmaceutics and agrochemicals\textsuperscript{126-127}.
Conventional approaches to acquire spectral and circular polarization information require cascading multiple components such as non-polarizing beam splitters, waveplates, and polarizers, or composite prisms comprised of naturally birefringent crystals (e.g., Fresnel rhomb\textsuperscript{128}) paired with a spectrometer. These spectrometers usually consist of focusing mirrors and a grating turret\textsuperscript{129} (Fig. 3.1). They suffer from insufficient grating dispersion to allow for large spatial separation within a short (e.g. mm to cm) light propagation distance, which places a lower limit on their physical size in order to achieve sufficient spectral resolution. The necessity of using a turret to mount gratings with different dispersions in order to tune/adjust spectral resolutions also adds significant complexity and bulkiness to the system. While handheld and miniaturized spectrometers do indeed exist\textsuperscript{130-132}, they are typically limited to a single, fixed spectral resolution and lack polarimetric capabilities. Similarly, while there have been efforts to utilize metasurfaces for spectroscopy, these devices typically suffer from these same limitations or a low spectral resolution\textsuperscript{133-134}.

![Figure 3.1: Left to right: Schematic diagrams showing a conventional grating spectrometer, an on-axis dispersive metalens, and a dispersive off-axis metalens. For a conventional spectrometer, the focusing and dispersive (grating) elements are separated. By design, the focusing mirror is achromatic and light with different wavelengths is focused onto the same detector plane, with lateral displacement resulting solely from the grating dispersion. In a typical on-axis metalens, the focal lengths change with wavelength and occur at different depths. When adapted to an off-axis configuration, the focusing angle also changes with wavelength in addition to the longitudinal focal length shift. They thus both contribute to the transverse displacement of the focal spot along a given plane.](image)

These are strong motivations to develop compact spectrometers capable of good spectral performance and helicity-resolving capabilities, i.e., distinguishing right/left circularly polarized (RCP/LCP) light. This can be achieved by using appropriately designed single-layer off-axis metalenses operating at visible wavelengths\textsuperscript{55}. These metalenses are comprised of identical nanofins which are geometrically rotated with respect to one another (Fig. 3.1). Since they are intended to
operate at visible wavelengths, the metalenses are fabricated using atomic layer deposition (ALD) of
titanium oxide (TiO$_2$) using the process described in Appendix A.1. By integrating multiple off-axis
meta-lenses with different parameters (focal length, spatial size, position, and focusing angle) onto the
same substrate, a highly versatile ultra-compact spectrometer with variable resolution, spectral range,
and minimal spatial footprint can be achieved. Due to the intrinsic CP sensitivity of metalenses based
on the geometric phase, such a spectrometer is also capable of resolving the helicity of light in a single
measurement. The device can thus be called a chiral spectrometer by virtue of this functionality.
Finally, their CMOS-compatible fabrication process is amenable to monolithic integration with camera
sensors and large-scale production.

The key advantages of using off-axis metalenses is apparent by examining Fig. 3.1. First and
foremost, the focusing and dispersive elements in a conventional spectrometer (focusing mirrors and
grating respectively) can now be combined in a single planar metasurface. Secondly, a significantly
higher dispersion can be achieved with an off-axis metalens. This can be understood by examining the
equation for its phase profile:

$$\varphi(x, y) = 2\pi - \frac{2\pi}{\lambda_d} \left( \sqrt{(x - x_f)^2 + (y - y_f)^2 + z_f^2} - f \right)$$

(3.1)

where $x_f$, $y_f$ and $z_f$ are the coordinates of the focus, $f = \sqrt{x_f^2 + y_f^2 + z_f^2}$ is the focal length, and $\varphi$
and $\lambda_d$ are the required phase and design wavelength respectively. For an off-axis lens, one can
introduce the off-axis angle $\alpha$ by requiring $x_f = f \sin \alpha$, $z_f = f \cos \alpha$, $y_f = 0$; this yields an off-axis
lens in the $x$-$z$ plane (i.e. a cylindrical lens) such as the case shown in Fig. 3.1. By substituting Eq.
(3.1) into the phase gradient term in the generalized Snell’s Law (Eq. (1.7)), one obtains the result:

$$\Delta \alpha \approx \sin^{-1} \left( 1 + \frac{\Delta \lambda}{\lambda_d} \right) \sin \alpha - \alpha$$

(3.2)

where $\Delta \alpha$ is a small change in the off-axis angle caused by a small change in wavelength $\Delta \lambda$. One can
see that for small $\Delta \alpha$, it increases with a larger initial off-axis angle $\alpha$. In conventional gratings, $\alpha$ is
limited by e.g. shadowing effects to be approximately a few tens of degrees; this in turn limits the

dispersion of the grating, resulting in a long optical path to achieve the desired spectral resolution. For
off-axis metalenses, one is not a priori limited in the maximum angle; this allows the metalens to
achieve a significantly higher intrinsic dispersion.

In addition, when $\alpha$ is non-zero, a small change in the wavelength of incident light results in an
additional lateral displacement of the focal spot $f \Delta \alpha$, whose magnitude is strongly dependent on both
$f$ and $\alpha$. This can be exploited to further increase the dispersion of the entire system, since in
conventional spectrometers, the focal length change arising from the focusing mirrors is negligible so
that any contribution to the dispersion arises solely from the change in grating diffraction angle. The
resultant sum of these displacements $|\Delta \vec{r}|$ (Fig. 3.1) and its variation can be quantified by the net
dispersion of the metalens $\frac{|\Delta \vec{r}|}{\Delta \lambda}$ along an appropriately placed camera plane$^55$:

$$\frac{|\Delta \vec{r}|}{\Delta \lambda} = \sqrt{\left(\frac{\Delta f}{\Delta \lambda}\right)^2 + f^2 \left(\frac{\Delta \alpha}{\Delta \lambda}\right)^2}$$ (3.3)

The minimum resolvable wavelength difference $\delta \lambda_{\text{min}}$ at the design wavelength $\lambda_d$, i.e. the
spectral resolution of the spectrometer, corresponds to a focal spot displacement along the camera
plane given by the Rayleigh criterion:

$$\delta \lambda_{\text{min}} = \frac{\Delta \lambda}{|\Delta \vec{r}|} \times 0.61 \frac{\lambda_d}{NA}$$ (3.4)

where $\Delta \lambda/|\Delta \vec{r}|$ is the reciprocal dispersion obtained from Eq. (3.3) and NA refers to the numerical
aperture of the off-axis metalens. However, in a spectrometer, the pixel size of the camera introduces a
further constraint on the spectral resolution since the pixel size defines the sampling rate. If the pixel
size is large compared to the Rayleigh limit, then according to the Nyquist sampling theorem, aliasing
occurs, which prevents one from resolving two points that are in close proximity. Since in general at
least 3 pixels are required to distinguish two points, the final spectral resolution of the spectrometer at
the design wavelength is the convolution between these two effects and can be written as$^{55,135}$:

$$\delta \lambda_{\text{min}} = \frac{\Delta \lambda}{|\Delta \vec{r}|} \times 0.61 \frac{\lambda_d}{NA} + \frac{\Delta \lambda}{|\Delta \vec{r}|} \times \frac{3D}{M}$$ (3.5)
where $D$ and $M$ are the camera pixel size and magnification of the spectroscopic imaging system, respectively. From this discussion, it is evident that by tailoring the off-axis focusing angle as well as the focal length, one can achieve a desired dispersion, and hence spectral resolution, at any design wavelength using metalenses. However, there is a trade-off between the spectral resolution and spectral range. For a system with a fixed detector size, using a higher NA metalens for a higher spectral resolution results in more chromatic aberrations and astigmatism (e.g. broadening of the focal spot size) for a given change in wavelength, which limits the spectral range of the spectrometer.

Another challenge associated with using high NA lenses is to locate a common plane where multiple wavelengths are in focus, due to the smaller depth of focus. Alternatively, one can lower the NA while increasing the dispersion to maintain similar resolution for a wider spectral range. In this case, since the dispersion is larger, the area of the active region of the camera sensor will pose the final limit to the working wavelength range.

3.2 Characterization of spectral and polarization properties

For general spectroscopic applications, which require a large working wavelength range and high resolving power within a spectrum of interest, several off-axis metalenses can be integrated on the same substrate to provide spatially separated focal spots in the same field of view of a camera. Each metalens possesses different spectral resolutions and spectral range and selectively focuses light with opposite CP states. In this way, one can realize within a single device of area less than 2 cm by 1 cm, the equivalent of multiple distinct gratings (with different wavelength ranges and resolutions), focusing lenses, and CP elements.

A representative SEM image of an off-axis metalens is shown in Fig. 3.2(a). For the final device (Fig 3.2(b)), four metalenses were fabricated on the same substrate: two with large ($\alpha=60^\circ$) focusing angles with a NA of 0.1 for high spectral resolution (R1 and L1) and two at small focusing angles of $25^\circ$ with a NA of 0.022 for a larger spectral range (R2 and L2). The two metalenses fabricated for each
Figure 3.2: (a) Scanning electron microscope image of a fabricated off-axis metalens. (b) Photograph of a fabricated device with four separate metalenses labeled R1, R2, L1 and L2. These metalenses are designed based on the phase profile in Eq. (3.1) but rectangular in shape to keep all focal spots within the active region of the camera. The primary focusing/dispersion direction is along the horizontal axis of the image. The letters R and L refer to the helicity of light focused by each meta-lens; 1 and 2 indicate the parameters used for the lens design. R1 and L1 have a NA of 0.11, focusing angle $\alpha = 60^\circ$, and a focal length of 20.1 mm, while R2 and L2 have an NA of 0.022, focusing angle of $\alpha = 25^\circ$, and a focal length of 11.1 mm. Physical dimensions of the lenses are $9 \times 0.1$ mm (R1 and L1) and $0.5 \times 0.8$ mm (R2 and L2). Scale bar: 5 mm. (c) CMOS camera image after magnification using a 10 × objective, of the focal spots generated by the metalenses for circularly polarized incidence. The upper and bottom focal spot corresponds to metalens R1 and R2. Both scale bars: 25 $\mu$m. (d) Horizontal cuts of the focal spots in (c), showing diffraction limited focusing.
NA are sensitive to opposite helicities of incident circularly polarized light: as a result of the use of the geometric phase in the design, any one single metalens would focus light of a chosen handedness while defocusing the other. Note that these metalenses are designed according to the phase profile dictated by Eq. (3.1), with care taken to avoid spatial overlapping of focal spots from different metalenses by proper placement on the substrate. The final shape of the metalenses is rectangular instead of circular to keep the focal spots within the active region of the camera; this results in the asymmetrical shape of the focal spots. The primary focusing/dispersion direction is along the horizontal axis of Fig. 3.2(c).

The focal spots of the metalenses were characterized using a narrow line with (~pm) diode laser (Quantum Optics) at the design wavelength of 532 nm. The laser beam is coupled into a fiber and subsequently collimated (Thorlabs RC04APC-P01) before being incident on the metalenses. An objective lens (Mitutoyo M Plan Apo 10×, NA 0.28) paired with a tube lens of focal length $f = 200$ mm and a CMOS camera (Edmund EO-5012) with a small pixel size of 1.67 $\mu$m were used for accurate characterization. Measured results are in good agreement with theory: close to diffraction limited focal spots with full-width half maximum (FWHM) of 2.3 $\mu$m and 11.6 $\mu$m are observed for the high and low NA metalenses, respectively (Fig. 3.2(c) and (d)). Similar values were obtained for metalenses designed for the opposite helicity. Additionally, Fig. 3.2(c) shows that the divergent light of opposite helicity introduces negligible background noise to the system. To build a compact spectrometer, the metalenses were paired with a CMOS camera (Thorlabs DCC1545M) (Fig. 3.3(a)). It has a pixel size of 5.2 $\mu$m and an active area of $6.66 \times 5.32$ mm$^2$. The camera plane is placed so that the quality of focal spots (i.e. shape and symmetry) is optimized for two distinct wavelengths ($\lambda = 532$ nm and 660 nm). The images of the focal spots corresponding to the NA = 0.1 and NA = 0.02 metalenses under linearly polarized illumination at $\lambda = 532$ nm, as well as their horizontal cuts, are shown in Fig. 3.3(b). Note that here the FWHM of the focal spot profiles are limited by the camera’s pixel size, in contrast to the characterization shown in Fig. 3.2(c).
Figure 3.3: (a) Photograph of the meta-spectrometer consisting of meta-lenses and a camera. (b) Measured intensity profiles of focal spots of (left) and their corresponding horizontal cuts (right) from the camera shown in (a), under linearly polarized light from a diode laser at $\lambda = 532$ nm. Top and bottom panels correspond to the focal spots of the $\text{NA} = 0.1$ and $\text{NA} = 0.02$ lenses, respectively. Scale bars: 40 $\mu$m (top) and 30 $\mu$m (bottom).

One often cited concern about metasurface focusing and dispersive elements is chromatic aberration. Since the required phase profile is wavelength dependent, at wavelengths different from the design, the focal spot will be aberrated and its position will be shifted both laterally and longitudinally (along the focusing axis). These aberrations are more pronounced for higher NA lenses due to the smaller focal spot size. In general, in order to maintain high spectral resolution for as wide a range of wavelengths as possible, the focal spot quality (i.e., its size and shape) should be preserved for that range. This requires both a suitable NA in terms of lens design and appropriate placement of the detector plane such that it intersects with the focal spots within their depth of focus. Here, we specifically designed the small diameter meta-lenses (labelled R2 and L2 in Fig. 3.2(b)) for a large spectral range while maintaining a reasonable spectral resolution. Ray-tracing simulations of their performance at 470 nm, 532 nm, and 660 nm are shown in Fig. 3.4(a). These simulations provide a guideline to identify the optimal position of the camera plane for minimizing the focal spot simultaneously at different wavelengths within the working range. This range is defined as the
Figure 3.4: (a) Ray tracing calculations for the low NA meta-lenses (R2 and L2 in Fig. 3(b)), which were designed for \( \lambda = 532 \text{ nm} \), at wavelengths of 470 nm, 532 nm, and 660 nm. A plane which simultaneously intercepts the focal planes at each of these wavelengths is shown. (b)–(d) Simulated spot diagrams color-coded to match their corresponding wavelengths. The ideal, aberration-free Airy disks are plotted as a reference. Good focal spot quality is preserved over a large wavelength range. (e) Measured spectra from a supercontinuum laser with 5 nm bandwidth using the meta-lens spectrometer, and (f) a commercial handheld spectrometer. The center wavelengths used varied from 480 nm to 700 nm in increments of 20 nm. (g) Measured dispersion values for the NA = 0.1 (blue) and NA = 0.02 (green) meta-lenses about their design wavelength of 532 nm.

spectrum over which the focal spots are still close to diffraction-limited along the dispersion direction. This criterion is more stringent than typically used in the industry, but we nevertheless find, under this definition, the spectral range of our meta-lenses to be almost 200 nm.

The aforementioned focal spot sizes are computed using a commercial software OpticStudio (Zemax LLC). Figures 3.4(b)–(d) show the spot diagrams corresponding to the intercept points on the
camera plane (black dashed line in Fig. 3.4(a)) for rays passing through the meta-lens. The colors of the spots correspond to their wavelength, i.e., 470 nm (blue), 532 nm (green), and 660 nm (red).

To visualize the spreading of the points, the ideal Airy disk at each chosen wavelength is plotted as a reference. These were calculated by using the Huygens–Fresnel principle to propagate the complex electric fields at the meta-lens to its corresponding focal plane. For wavelengths away from the design wavelength $\lambda_d = 532$ nm, the focal plane is defined by a normal vector and a point, corresponding to the propagation direction of the chief ray (the normal vector) as well as the intersection of the chief ray and paraxial rays (the point). This definition was used in the calculations since due to chromatic aberrations, not all rays have a common intersection at wavelengths different from the design wavelength. From Figs. 3.4(b)–(d), it is clearly seen that the focal spots within 470–660 nm lie mostly within their Airy disk along the dispersion direction. Deviations are within the pixel size of our camera (5.2 $\mu$m). Note that this only applies to the primary focusing direction of the meta-lenses (horizontal axis in Figs. 3.4(b)–(d)).

Subsequently, a supercontinuum laser (SuperK Varia, NKT photonics) with its bandwidth set to 5 nm for wavelengths ranging from 480 nm to 780 nm in steps of 10 nm was used to characterize the performance of the meta-spectrometer and compare it against that of a commercial handheld spectrometer (OceanOptics USB4000 UV-VIS). The known center wavelengths of the supercontinuum laser input were used to calibrate the metaspectrometer system and account for the slight non-linear dispersive effects across this broad wavelength range (Appendix B.1). Figures 3.4(e) and 3.4(f) illustrate the measured spectrum by the meta-spectrometer and the commercial spectrometer, respectively. Camera images of the focal spots are shown in Appendix Fig. B.2. One can observe close to identical spectral lineshapes with FWHM between 5 and 8 nm across the visible. This validates the claim for the working range of the low NA meta-lenses; additionally, no significant linewidth broadening effects are observed together with the increase in wavelength within our previously defined working wavelength range from 470 nm to 660 nm. Beyond this range, due to aberrations in the focal spots, one can observe a significant, consistent increase in FWHM which is larger than the commercial
This is in good agreement with the previously discussed theoretical results in Figs. 3.4(b)–3.4(d). These results also offer further evidence that by a proper design a planar meta-lens can achieve comparable or even superior performance to traditional lenses.

Next, the dispersion and spectral resolution of the meta-spectrometer were quantified. Figure 3.4(g) plots the measured peak intensity positions of the focal spots as a function of wavelength for both high and low NA meta-lenses about their design wavelength of 532 nm. These dispersion values are calculated by tracking the change in these positions as the wavelength is varied with higher precision in steps of 1 nm. As expected and consistent with the earlier calibration results in Fig. B.1 in the Appendix, the observed dispersions are approximately linear within a range of 30-40 nm. These values are found to be 59 $\mu$m/nm and 24 $\mu$m/nm for meta-lenses with NA of 0.1 and 0.022, respectively. Using Eq. (3.5), we determine the intrinsic experimental spectral resolution achievable to be 0.05 nm and 0.46 nm for meta-lenses with corresponding NAs of 0.1 and 0.022. In practice, for this meta-lens spectrometer, the spectral resolution is limited by the pixel size of the camera (5.2 $\mu$m); the detector-limited attainable spectral resolution therefore becomes (Eq. (3.5)) 0.31 nm and 1.11 nm, respectively. It is important to note that the values achieved here are a proof of concept, given various experimental constraints. In principle, by using a suitable camera with a larger active region, one can significantly improve the working range while retaining high spectral resolution. As an example, it is possible to design meta-lenses of similar sizes (R2 and L2 in Fig. 3.2) with a spectral range covering the entire visible (400-800 nm) where all the focal spots lie within their respective diffraction limited Airy disk. Further details and simulated results are presented in Fig. B4 in the Appendix.

Finally, the CP resolving capability of the metaspectrometer was characterized. A complete description of any arbitrary polarization state can be made using the Stokes parameters $S_0$, $S_1$, $S_2$, and $S_3$ which correspond to the intensities of incident, vertically polarized, 45° polarized, and circularly polarized light, respectively. Here, emphasis is placed on $S_3$ because the chiral optical response (circular dichroism) of most materials is weak compared to its linear counterpart. Furthermore, conventional approaches to measuring $S_3$ as a function of wavelength using a single planar device are
challenging due to the requirement of separating light in both spectral and polarization domains. Although various nanostructures are known to behave as miniaturized CP filters, they lack the ability to resolve spectral information\textsuperscript{136-138}. By using the off-axis meta-lenses based on rotated nanofins, the resulting phase profile intrinsically focuses one helicity while defocusing the other. This functionality thus enables one to resolve the helicity of incident light in a single measurement by having two separate meta-lenses to focus RCP and LCP on the same chip.

![Figure 3.5](image)

**Figure 3.5:** (a) Stitched camera images of the focal spots from the meta-lenses for right circular polarized (RCP), linearly polarized (LP), and left circularly polarized (LCP) incident light. Intensities are normalized to the same color scale and the illumination wavelength was 532 nm. The intensities of the RCP and LCP spots are observed to be slightly different due to fabrication imperfections as they are focused by different meta-lenses. Scale bar: 100 \(\mu\)m. (b) Normalized \(S_3/S_0\) Stokes parameter as a function of wavelength measured by the meta-spectrometer (blue), a commercial polarimeter (red), and theoretical calculation (black) from the known waveplate retardance.

Figure 3.5(a) illustrates the variations in intensities of the focal spots upon changing incident light polarization (from RCP to linear to LCP). Differences in the intensities of the focal spots for RCP and LCP light arise from fabrication errors since they originate from different meta-lenses (R1 and L1 versus R2 and L2, as shown in Fig. 3.2(b)). To control the polarization of the incident light, a polarizer (Thorlabs GTH10) and a zero-order single wavelength (532 nm) quarter waveplate (Thorlabs WPQ05M-532) were placed in the collimated beam path right before the meta-lenses. Next, various \(S_3\) Stokes parameters (normalized to incident light intensity \(S_0\)) at different wavelengths were obtained using the meta-lenses and compared to the results of a commercial
polarimeter as well as analytical calculations (Fig. 3.5(b)). These various elliptically polarized states at
different wavelengths are generated by changing the wavelength of input light (SuperK Varia). Due to
the deviation in optical retardance of the single wavelength (532 nm) quarter waveplate (characterized
and provided by Thorlabs), various elliptical polarization states can be generated across a broad range
in the visible. Measured values upon appropriate calibration (following the approach in Ref. 139) are
seen to be in good agreement with both the values measured by a commercial polarimeter (Thorlabs
TXP) and analytical predictions based on known retardance data. For wavelengths close to $\lambda = 532$ nm
where the retardance of the waveplate is almost exactly $\lambda/4$, the measured values are
slightly greater than unity (approximately 1.01) likely due to experimental noise (e.g., pixel noise
and limited dynamic range of the camera) in the measurements$^{140-141}$. Although here only the quantities
$S_3/S_0$ were obtained due to the chiral response of the meta-lenses designed with the geometric phase, in
principle, by adding linear grating elements, the full Stokes parameters can be resolved$^{142-143}$.

3.3 Conclusions

In this Chapter, an ultra-compact meta-spectrometer based on the integration of multiple planar off-
axis metalenses at visible wavelengths has been presented. This device has several advantages over its
traditional grating-based counterparts: first, it combines the functions of a focusing and dispersive
element in a single planar structure, which eliminates the need for rotating turrets or focusing mirrors.
Second, in terms of performance, it surpasses conventional blazed grating elements as one can achieve
extremely large dispersions which are otherwise unattainable. In addition, the integration of several
metalenses with different NAs on one substrate allows for multiple different spectral resolutions and a
flexible working wavelength range with no further increase in system bulk or complexity. Finally, the
meta-lenses can provide extra information about the circular polarization state of incident light, which
is not attainable for conventional devices without the use of additional optical elements (e.g., polarizer
and waveplates). The use of dielectric TiO$_2$ as the working material also renders it compatible with
existing CMOS processes where large-scale production could take place in a single lithographic step or be monolithically integrated with sensor technologies. We envision numerous potential applications in health care, environmental sensing, and related areas for this technology.
Giant intrinsic chiro-optical activity in planar dielectric nanostructures

4.1 Chiral metamaterials and metasurfaces

An object is said to be chiral if it cannot be superposed with its mirror image via rotation or translation operations alone. This geometric or structural chirality is an intrinsic part of the natural world and manifests itself in numerous forms, ranging from molecules like amino acids\textsuperscript{144} to macroscale objects such as quartz crystals\textsuperscript{145} and even entire living organisms\textsuperscript{123}. Due to its profound implications for various disciplines, particularly pharmaceutics, the study and manipulation of chiral media has been a
highly active field of study in recent years.

In the context of optics, chiral objects interact with circularly polarized (CP) light in different ways depending on their handedness; they are said to exhibit optical chirality and are characterized by circular dichroism and circular birefringence. The latter is also known as optical activity. However, chiro-optical responses in most naturally occurring compounds are very small due to the fundamental mismatch in size scale between molecules and the wavelength of incident light.

Suitably designed three-dimensional (3D) metamaterials comprising arrays of chiral nanostructures possess chiro-optical responses several orders of magnitude greater than their naturally occurring counterparts. Circular dichroism on the order of several tens of percent, as well as negative refractive indices due to chirality have been observed at THz and GHz frequencies. However, complexities associated with 3D fabrication and a general lack of suitable, low-loss materials at optical wavelengths have so far limited their application.

The planar counterparts to these chiral metamaterials, chiral metasurfaces, overcome these challenges but exhibit chiro-optical responses that are significantly weaker and become vanishingly small close to normal incidence. This is due to fundamental symmetry considerations: in planar structures, there always exists a plane of reflection symmetry perpendicular to the surface normal which renders them geometrically achiral. Symmetry-breaking effects due to the substrate are known to be negligible in terms of their contribution to the strength of the eventual chiral effect. As such, one approach to achieve strong optical chirality is to vary the material composition such as in quasi-3D multilayered or oligomer structures. An alternative method that stays true to the planar geometry is to break the symmetry via external experimental configurations, that is, illuminating at oblique incidence and observing the resultant zeroth-order beam. One can also observe a chiral response in higher orders of diffracted light from planar chiral structures at normal incidence. Although these planar structures can lead to exotic chiro-optical effects such as circular conversion dichroism and asymmetric transmission due to the reversal of their handedness when illuminated from opposite sides, it is crucial to note that they remain fundamentally distinct from the ‘true’ geometric chirality.
characteristic of 3D objects; following established nomenclature, the former will be called ‘extrinsic chirality’\textsuperscript{156} and its 3D counterpart ‘intrinsic chirality’.

In this Chapter, we overcome these inherent limitations associated with chiral metasurfaces by using planar dielectric gammadion nanostructures whose radiation patterns are dominated by exceptionally strong electric and magnetic higher-order multipole responses in the visible spectrum\textsuperscript{53}. Contrary to conventional wisdom, the chiroptical response of these planar structures is characterized by intrinsic chirality; it manifests strongly at normal incidence and does not change when the incidence direction is reversed. Numerical simulations show that one can obtain close to unity circular dichroism in transmission, with more than 95\% of incident light of a chosen helicity being transmitted at \( \sim 540 \) nm; experimentally we achieve \( \sim 80\% \) circular dichroism and circular birefringence exceeding 100 000\(^\circ\)/mm (in units of polarization rotation per unit thickness). These values are comparable to or exceeding 3D geometrically chiral metamaterials (Fig. 4.1), which is unprecedented for planar structures. In this comparison, we have used the un-normalized circular dichroism values given by the raw differential transmittance (i.e. transmitted power) to ensure an accurate representation of device performance.

To date, gammadion structures have been shown to possess circular dichroism on the order of a few percent\textsuperscript{158-159,164}. Other works involving planar structures have made use of the interaction between electric dipole and quadrupolar modes to realize chiro-optical behavior and high-quality factor modes in the mid-infrared regime\textsuperscript{167}, but its anisotropic nature results in non-negligible circular polarization conversion such that the measured output cannot be attributed to a purely chiral response. In our design, the motif and all its relevant geometrical parameters are optimized to ensure the absence of circular polarization conversion and that higher-order multipoles (up to the magnetic octupole) are dominant. This engineering of the multipoles results in unprecedented performance values in terms of near-unity transmission and circular dichroism, particularly in the visible spectrum, and thus showcases the full potential of planar chiral nanostructures.
Figure 4.1: Representative examples of chiral metamaterials of varying designs and their measured performances at their respective operating frequencies. The performances are characterized by their circular dichroism (unnormalized, calculated from the differential transmittance $\Delta T$) and circular birefringence, where applicable. Different colors correspond to different geometries of the chiral structures, that is, 3D (blue) and planar (green). Generally, the former possesses significantly stronger chiro-optical effects due to intrinsic chirality. The experimental results from this work using planar structures that exhibit intrinsic chirality are represented by the red triangle.

4.2 Origins of chiroptical response in nanostructures

We first look at the origin of chiro-optical responses in various metamaterials in greater detail. From an electromagnetic source perspective, this can be understood by considering the superposition of radiated electric fields from electric and magnetic multipole moments generated in a structure upon illumination (Fig. 4.2(a) – (c)). When these moments are perpendicular to each other and lie on two orthogonal planes, optical activity in the form of polarization rotation can only occur at oblique angles,
where the radiated fields are nonparallel. On the other hand, coplanar, orthogonal moments have parallel electric field components and thus no rotation of the field can occur. As a result, at least some components of electric and magnetic moments that are parallel to each other must exist, such that their resultant electric fields are aligned perpendicularly, leading to polarization rotation and hence optical activity. This intuitive picture is a generalization (to oblique incidence) of what is known as the Rosenfeld criterion\textsuperscript{168} in the relevant literature, which states that the condition for chiro-optical activity can be written as \( \mathbf{p} \cdot \mathbf{m} \neq 0 \), where \( \mathbf{p} \) and \( \mathbf{m} \) are the net electric and magnetic dipole moments, respectively. Conventional methods of achieving strong chiral responses thus involve the use of either planar nanostructures under oblique incidence or intrinsically chiral metamaterials (Fig.4.2(a), (b)). In the former, due to the planar geometry and deeply subwavelength thickness, only tangential currents exist, and the resultant net magnetic moment (\( \mathbf{m} \)) therefore always points out of plane, orthogonal to the in-plane net electric moment (\( \mathbf{p} \)) (Figure 4.2(a)). Consequently, the required superposition of radiated fields can only occur off-normal (where the radiation from \( \mathbf{m} \) is non-zero). This is true regardless of whether chiral or achiral shapes are used. While in principle the presence of a substrate should break the symmetry, such effects are known to be negligible in terms of magnitude. In contrast, for 3D chiral structures, the geometry admits both normal and tangential currents, leading to the generation of in-plane magnetic moments (\( \mathbf{m}_\parallel \)) with a non-zero component parallel to the in-plane electric moment (\( \mathbf{p}_\parallel \)), resulting in strong chiro-optical behavior at normal incidence (Fig. 4.2(b)).

A natural extension to this concept would be to consider the possibility of in-plane magnetic moments existing in planar chiral structures. For this to occur, the structures need to possess a finite thickness on the order of the excitation wavelength in the material and, in the case of dielectric materials, have sufficiently high refractive indices to support out-of-plane electric displacement currents that generate in-plane magnetic moments\textsuperscript{40,109-110} (Fig. 4.2(c)). Further details on the physical origins of such modes are provided in Appendix C.1. In this case, the resultant chirooptical response would be characteristic of intrinsic chirality, that is, it can be observed at normal incidence and does not change when the direction of incidence is reversed. This is a restatement of the fact that
extrinsically chiral

Figure 4.2: (a) – (c) Schematic illustrations of the operating principle of a representative (a) planar extrinsic (pseudo) chiral metasurface, (b) 3D intrinsic metamaterial and (c) optically thick planar structure with intrinsic chirality under their respective illumination conditions (red arrows). The insets show magnified views of the structures overlaid with the relative orientations of electric (\(\mathbf{p}\), green arrow) and magnetic (\(\mathbf{m}\), blue arrow) multipole moments generated under such illumination. These moments are drawn as simple vector arrows for ease of illustration. In (a), due to geometrical constraints, the net electric and magnetic moments are always orthogonal. In (b) and (c), the chiral structures can support in-plane magnetic moments (\(\mathbf{m}_\parallel\)) that have a non-zero component along the direction of the in-plane electric moment (\(\mathbf{p}_\parallel\)).

objects (Fig. 4.2(a)) change their handedness or sense of twist depending on the observer’s perspective, whereas intrinsically chiral ones do not (Fig. 4.2(b)). Although the existence of these in-plane magnetic moments in high-index dielectrics such as silicon has been well studied using achiral nanoparticles of various geometries to realize highly directional nanoantennas and Huygen’s metasurfaces\(^{34,39,169-170}\), their chiral counterparts have only been superficially explored. We emphasize that these planar structures are fundamentally distinct from 3D intrinsically chiral objects (such as the helix) as they still possess a plane of reflection symmetry in the normal direction, and a traditional symmetry argument\(^{171}\) would therefore suggest that they cannot exhibit intrinsic chiral behavior regardless of thickness and refractive index. This neglects the possibility for vertical, out-of-plane electric displacement currents in planar geometries due to propagation effects.
4.3 Device design and characterization

We chose a common motif, the gammadion, as our planar chiral nanostructures (Fig. 4.3(a), (b)). Periodic arrays are fabricated using electron beam lithography and atomic layer deposition of titanium oxide, a relatively high-index dielectric lossless in the visible spectrum. Intuitively, their chiral behavior arises from the orthogonal arrangement of arms with respect to each other and the resulting linear polarization conversion (Fig. 4.2(c) and 4.3(a)); excitation with, for example, \(x\)-polarized light results in an induced displacement current and radiated electric fields along the \(y\) axis due to the geometry of the structure. Note that the choice of these structures with fourfold (\(C_4\)) symmetry ensures the absence of linear birefringence, that is, the amplitude transmission coefficients are related by \(t_{xx} = t_{yy}, t_{yx} = -t_{xy}\), such that circular polarization conversion into both right circularly polarized (RCP) and left circularly polarized (LCP) (\(t_{xx} - t_{yy} \pm i(t_{xy} + t_{yx})\), respectively) is zero. The latter quantity is distinct
from but can be often misconstrued as circular dichroism.

Additionally, we note that at normal incidence for \( C_4 \) symmetric structures, reciprocity ensures that there is no dichroism in reflection\(^{159} \), and as a result, diffraction (into higher orders) must exist in order for dichroism to be observed in transmission. In the design of our nanostructure array, the periodicity \( \Lambda \) (Fig. 4.3(a), (b)) was chosen to be 500 nm, such that the targeted free space wavelength (540 nm) is between the periodicity and effective wavelength in the substrate. The performance of our structure under RCP and LCP light at normal incidence is schematically illustrated in Fig. 4.3(a); RCP light transmits in the zeroth order, whereas LCP light is completely diffracted into the first order (here 46\(^\circ\)). The angle at which this occurs can be controlled by adjusting the periodicity. Here, we have included a waveguide layer of thickness 300 nm to further increase the circular dichroism in the zeroth-order transmission, analogous to the design of guided or leaky mode resonance structures\(^{172} \). Essentially, at resonance, part of the incident wave of the chosen helicity (RCP) is coupled into the guided mode, which then slowly leaks from the waveguide and interferes with the applied wave to produce a sharp filtering response. For the other helicity (LCP) due to chirality of the gammadion structure, the effective index of the mode is different, and thus, light does not couple efficiently into the waveguide. As a result, the structure exhibits a background response typical of the multilayered geometry. The same is true for other wavelengths away from the resonance. Further details, including the optimization of performance with and without the waveguide layer, can be found in Appendix C.2. However, we emphasize that the structure is not reliant upon this waveguide layer in order to exhibit its chiroptical behavior; the origin of this response can be traced back to the electromagnetic modes (multipoles) excited purely within the structure. It is readily apparent from Appendix Fig. C.3 and C.4 that field profiles similar to Fig. 4.3 exist within the structure, even without the waveguide layer. The latter exists only to improve overall transmission efficiency and circular dichroism by modulating the transmission envelope.
Figure 4.4: Multipole decomposition. (a), (b) The far-field intensity $|E_x|^2$ with explicit contributions from each multipole for (a) LCP and (b) RCP incidence. Only the two strongest contributions, the toroidal quadrupole ($T_O$, purple) and magnetic octupole ($M_O$, yellow), as well as the standard electric ($P$, cyan) and magnetic dipole responses ($M$, red) are shown for clarity. (c), (d) Tuning of the spectral position and strength of (c) the toroidal quadrupole and d magnetic octupole response by changing the aspect ratio of the structure. Here, the length and width of each arm of the gammadion were kept constant at 370 and 74 nm, respectively, while the height was varied from 300 to 400 nm. Each curve is offset by 8 units along the y axis. The shaded regions indicate the region of interest, where the resonances are tuned with height of gammadion.

A closer analysis of these modes excited in the gammadion reveals the presence of higher-order multipole moments. Using finite-difference time-domain simulations (Lumerical Inc.), we computed the in-plane component of the magnetic fields (here $H_x$) and the vector sum of corresponding generating currents ($J_z$ and $J_y$) for both incident circular polarization states across a vertical cut through the middle of the structure ($y$-$z$ plane) at 540 nm (Fig. 4.3(c) and (d)). For the RCP case, $H_x$
exhibits three antinodes opposite in sign within the structure, indicative of an octupole dominated response (Fig. 4.3(d)). In contrast, under LCP illumination at the same wavelength, fewer antinodes of opposite sign are observed (Fig. 4.3(c)), characteristic of a mixture of lower-order multipole (quadrupolar) moments. This is a direct result of the chirality of the structure, which leads to significantly different current distributions and far-field radiation patterns for the two circular polarization states. As mentioned in the previous section, these field distributions remain the same even without the underlying TiO$_2$ waveguide layer, indicating that the chirality arises from the planar structures themselves. It is important to note that the use of higher-order multipoles is essential to achieve a strong chiro-optical response due to the diffraction requirement for chirality in this configuration. Although engineering only the dipolar response is sufficient for achiral, subwavelength devices such as Huygen’s metasurfaces operating in the zeroth order, manipulation of the radiation into large angles is necessary here. One should therefore make use of several higher-order multipoles, whose primary radiation directions are off-normal.

To study the contributions of each multipole in more detail, we performed multipolar decomposition using the charge-current expansion framework to represent the origin of electromagnetic radiation in the structure by point-like multipole sources$^{173-175}$. We considered three families of multipole excitation: electric, magnetic, and toroidal and performed the expansion up to the electric/magnetic octupole and toroidal quadrupole. Toroidal excitations can be treated explicitly within such a charge-current expansion framework, distinct from the usual far-field expansion using vector spherical harmonics that are comprised only of electric and magnetic coefficients. They comprise a family of elementary electromagnetic sources distinct from their electric and magnetic counterparts, that is, they are associated with radial current density of the form $\mathbf{r} \cdot \mathbf{J}$, in contrast to charge and azimuthal current density ($\mathbf{r} \times \mathbf{J}$), which correspond to electric and magnetic multipole moments, respectively. These toroidal moments have drawn significant attention recently by being responsible for exotic phenomena such as non-radiating anapoles$^{176}$ and have spurred research into novel devices ranging from toroidal lasers to sensors$^{175}$. 

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Intensities of the zeroth-order radiated electric field $|E_x|^2$ for a single gammadion structure (in the presence of its neighbors) for various multipoles are shown in Fig. 4.4(a) and 4.4(b). Here, the excitation light source was CP and normally incident. Only the two strongest contributions as well as traditional electric and magnetic dipolar responses are shown for clarity. A complete set of multipole responses is provided in Appendix Fig. C.5. We observe strong dichroism in the zeroth-order electric field at ~ 540 nm, where the higher-order multipole radiation peaks for RCP but exhibits a strong dip for LCP. As expected from the field distribution shown in Fig. 4.3(d), the magnetic octupole plays a large role in the chiro-optical response under RCP illumination. However, it is revealed that the dominant factor is actually the toroidal quadrupole (Fig. 4.4(b)). This affirms the importance of considering toroidal moments as an independent source of radiation (rather than subsumed under the electric multipole). It is also interesting to note that the electric and magnetic dipole contributions in this case are very much weaker than their higher-order counterparts; they are almost negligible for both input polarizations, which affirms chirality as a concept beyond the usual dipolar approximation. This provides further evidence of the importance of generating and tailoring higher-order multipole responses in the design of metamaterials and metasurfaces in general. We emphasize that these strong high-order multipolar contributions are only possible due to the geometry of the structure; the thickness (on the order of the incident wavelength) supports in-plane magnetic moments necessary for generating magnetic and toroidal responses, whereas the proximity of the individual ‘arms’ in the gammadion enables interactions between the induced charges and currents, thereby giving rise to higher-order multipoles. This provides an intuitive picture for how the overall strength of the multipoles present in the system can be tuned by varying both the length of the individual arms and the thickness, that is, by changing the aspect ratio. This is analogous to the tuning of magnetic and electric dipoles shown earlier in Appendix Fig. C.1. Here, in Fig. 4.4(c) and 4.4(d), by varying the height for a fixed length and width of the gammadion (370 and 74 nm, respectively), we show that a similar relationship between aspect ratio and multipole strength holds true. Note that in order to achieve large chiro-optical responses, the phases of the multipoles must also be considered such that
their radiation interferes constructively (destructively) under RCP (LCP) illumination; hence, the optimal thickness was chosen to be 340 nm instead of the lower values that appear to have larger multipole strength but smaller overall zeroth-order transmittance. This fact is verified in Appendix Fig. C.6, where the overlap between the dominant multipole radiation and simulated net zeroth-order transmittance as well as a color plot of the circular dichroism are shown.

Figure 4.5: Far-field angular radiation patterns ($|E_x|^2$). (a) – (d) Top: 3D far-field angular radiation patterns and bottom: their 2D projections for the (a) electric dipole ($E_{11}$), (b) magnetic dipole ($M_{11}$), (c) magnetic octupole ($M_{31}$) and (d) toroidal quadrupole ($T_{21}$), respectively. For clarity, only the forward radiation direction ($z \leq 0$ half-space) is shown. The subscripts denote the corresponding spherical harmonic, $Y_{lm}$, used to calculate the electric field. Here, the $m = \pm 1$ harmonic is chosen as it is the dominant contribution to radiation along the $z$-direction, corresponding to zeroth-order transmission from the structure. Each point on the 2D projections obeys $k_x^2 + k_y^2 + k_z^2 = k_0^2$ ($k_0 = 2\pi/\lambda$ is the wavevector), which defines the radiation direction, and the magnitude is given by the color bar. The black lines demarcate where $k_z = 0$, that is, the radiation only occurs in the $x$–$y$ plane.

Figure 4.5 illustrates the 3D and projected 2D angular radiation patterns ($|E_x|$) of both conventional dipoles and dominant multipoles that contribute to zeroth-order radiation in the structure. Here, they are expressed in the spherical harmonic basis (instead of Cartesian), following the approach used in Ref.73 and are classified by their source (electric (E), magnetic (M) and toroidal (T)) as well as a pair of indices $lm$ specifying the order and degree of the spherical harmonic being considered. Note that dipole, quadrupole and octupole modes correspond to $l = 1, 2$ and 3, respectively, and their associated $m$ values range from $-1$ to $1$. Since only $m = \pm 1$ harmonics have non-vanishing contributions
to the zeroth order and the radiation patterns from \( m = +1 \) and \(-1 \) are identical, in Fig. 4.5 we show only the case for \( m = +1 \). Additionally, in the 3D figures, only the forward scattered (transmitted) radiation corresponding to the \( z \leq 0 \) half-space is illustrated for clarity. The 2D projections are analogous to what would be seen upon capturing the 3D radiation patterns with a microscope objective and imaging its back focal plane. The axes correspond to the normalized wavenumbers \((k_x/k_0 \) and \( k_y/k_0 \)) such that every point on the image satisfies \( k_x^2 + k_y^2 + k_z^2 = k_0^2 \). This defines the direction of radiation, and the colormap specifies its magnitude. From these plots, one sees that it is the difference in intensities of the \( M_{31} \) and \( T_{21} \) modes that lead to the large circular dichroism in the zeroth-order radiation under RCP (LCP) illumination of the structure.

We experimentally characterized the chiro-optical properties of our planar nanostructures using a home-built microscopy setup. The light source is a supercontinuum laser, and the detectors used in turn are a commercial spectrometer and polarimeter. Figures 4.6(a) and (b) illustrate the measured and simulated zeroth-order transmittance for both RCP and LCP light across the visible spectrum (450–700 nm). Note that these values are normalized to the transmitted light through an unpatterned glass substrate (with the TiO\(_2\) layer) to remove contributions from reflection due to the substrate as well as interference effects caused by the TiO\(_2\) thin film. The unnormalized circular dichroism spectrum given by the raw differential transmittance \((\Delta T)\) is shown in Fig. 4.6(c). We observe that \( \sim 87\% \) of RCP light is transmitted in the zeroth order, with a difference in transmittance (i.e., circular dichroism) of almost 80\% at a wavelength of 540 nm. Incident LCP light is almost completely diffracted into the first order (here 46°) by design as dichroism in reflection is forbidden by the \( C_4\) symmetry and there is no absorption. These values agree well with the simulations; a slight discrepancy of \(<10\%\) is likely due to structural imperfections during fabrication, which mainly arise from proximity effects during electron beam lithography and deviations in the resist thickness. These could lead to resultant band broadening and lower efficiencies. It should also be noted that even in the simulations, residual reflection (\(\sim 5\%\)) remains after accounting for substrate effects: this occurs since the structures used in the current
Figure 4.6: (a), (b) Experimental a and simulated b zeroth-order transmittance spectra for the planar chiral nanostructures for both RCP (red) and LCP incident light (blue). (c) Experimental (solid line) and simulated (dotted line) circular dichroism spectra, defined as the differential transmittance between RCP and LCP from the results shown in (a) and (b), respectively. (d) Circular birefringence spectra (also known as the optical rotary dispersion) of the gammadions, comprising azimuth rotation angles from polarimetric data (green circles) and finite-difference time-domain (FDTD) calculations (dotted line). (e) Polarization ellipses of the zeroth-order transmitted polarization states at various wavelengths under linearly polarized incidence. Their ellipticity and azimuthal angles are obtained from the experimental data shown in (c), (d).

scheme lack sufficient degrees of freedom to be impedance matched to free space after optimizing for chiro-optical behavior. In principle, this can be overcome with the use of more complicated structures using multiple coupled resonances. In addition, some of the spectral features are below the resolution of the spectrometer (~1.5 to 2 nm), such as those at 560 and 610 nm in Fig. 4.6(a) and 4.6(b), resulting in them being truncated and thus being different from the simulated results. As is characteristic of an
intrinsically chiral response, besides exhibiting strong chiral behavior at normal incidence, the
handedness of the structure is also invariant to the propagation direction of light (see Appendix Fig.
C.7)— when the sample is reversed, the results are identical (to within experimental noise), as shown
in Fig. 4.6(a) and 4.6(b).

The circular birefringence was measured by performing polarimetric measurements with linearly
polarized input; the azimuthal angle of the (generally elliptical) output polarization state corresponds
to the polarization rotation due to circular birefringence. Figure 4.6(d) shows the resultant rotation as a
function of wavelength (also known as the optical rotary dispersion). Note that due to the minimum
bandwidth achievable for the supercontinuum laser (~5 nm) and the lack of spectroscopic capabilities
in the polarimeter, sharp features in the spectrum cannot be resolved. However, by overlaying
simulated results with experimental data, we observe close agreement overall. Furthermore, a peak
polarization rotation of ~ 60° for a combined structure and waveguide thickness of ~ 600 nm is
achieved (~100 000°/mm, in units of degrees of rotation per unit length). To put this result in
perspective, the polarization rotation of most naturally occurring chiral media is on the order of
degrees per mm; to the best of our knowledge, this is also significantly larger than state-of-the-art
chiral metasurfaces, including those operating at oblique incidence (Fig. 4.1).

4.4 Conclusions

Although recent advances in the field of metamaterials have enabled a wide variety of planar optical
devices such as lenses, holograms and q-plates\textsuperscript{30,32,45,48-49,99} as well as digital or programmable
metamaterials\textsuperscript{12,18} that center on the principles of phase and amplitude modulation, for true polarization
control at the level of a single nanostructure, chiral structures are indispensable. Although there has
been much seminal work done in this area by the community, much of it has centered on either
extrinsic or lossy plasmonic devices or demonstrated only at GHz/THz frequencies, where the size
scales make it possible to engineer various 3D, complex structures. Crucially, there is a lack of
compelling demonstrations of high-performance planar chiral devices at optical wavelengths. Moreover, certain subtleties regarding the underlying distinction between planar and 3D intrinsic chirality have yet to be made explicit. In this Chapter, we have presented planar chiral nanostructures with intrinsic chirality by using the propagation effect and measure close to unity circular dichroism (80%) and giant polarization rotation (100 000°/mm) at normal incidence for the visible spectrum. This was achieved by tailoring the higher-order multipole response, which plays a significant role in off-normal scattering compared to the usual dipolar approximation used to characterize chiral phenomena. This could pave the way for the widespread adoption of planar chiral nanostructures in numerous applications ranging from polarization optics to telecommunications, given their single step lithographic nature and easily scalable fabrication process.
Dispersion engineering and aberration control

5.1 Optical dispersion

Optical dispersion results from the variation in the index of refraction of a material with the frequency of incident light. Accurate control over this property is crucial in numerous industrial and research applications. For example, it mitigates pulse spreading in optical fibers which are the backbone of modern telecommunications\textsuperscript{177-178}, enables pulse shaping in ultrafast laser optics\textsuperscript{179-180}, and ensures faithful image reproduction in imaging systems by reducing chromatic aberrations. This has made possible various technologies behind widely used commercial products ranging from state-of-the-art
microscopes to cameras, as well as various metrology instruments that are indispensable to modern manufacturing processes.

Historically, after Sir Isaac Newton used a prism to disperse sunlight into different colors (i.e. wavelengths), he realized that a similar effect due to the inherent dispersion in glasses led to image blurring in lenses. Since it is impossible to make a glass with a constant refractive index for all wavelengths of light, he resorted to using reflective mirrors exclusively in his design of telescopes. In the 1730s, it was discovered that a doublet lens made of flint and crown glasses comprising different types and amounts of metal oxides in a fused silica matrix could significantly reduce chromatic aberrations. This is because introducing an additional lens made of slightly different materials provides an extra degree of freedom to compensate for the dispersion of the entire system. Nevertheless, it was not until the late 1800s that the combined efforts of Carl Zeiss, Ernst Abbe and Otto Schott in achromatic lens design and glass-making techniques led to the development of multi-wavelength, color-corrected microscope objectives that are still widely used today.

This example highlights the difficulties inherent to the approach of correcting chromatic aberration by altering material composition. A continuous range of desired dispersion values cannot be achieved over a large bandwidth and allowed combinations of glasses/dopants must be explored extensively, which is incredibly time-consuming and challenging in terms of fabrication. To date, there are only about a thousand types of different glasses that are readily available from major glass suppliers such as Schott, Hoya and Ohara. This includes overlapping cases where glasses from one company possesses very similar optical properties to those from another.

In mobile phone cameras, the camera module typically consists of about 6 plastic aspherical lenses made of different materials to correct both chromatic and monochromatic aberrations. Precise alignment of these lenses is another great challenge and the difficulty increases with the number of lenses. Although many companies are able to mass-produce the injection-molded plastic lenses, only a few can perform precise alignment with high yield for mobile phone cameras. Together, these factors represent a technological and manufacturing challenge to the quest of further shrinking the form factor.
of high-end achromatic optical components.

In sharp contrast to traditional refractive optics, optical metasurfaces consisting of sub-wavelength nanostructures can introduce an effective refractive index and dispersion dictated mainly by the geometrical parameters and arrangement of the structures, rather than material composition. Recent advances in nanofabrication have gradually improved the efficiency of metasurfaces up to 90% in some cases\textsuperscript{30,185}, which makes them viable candidates for a variety of practical applications. Importantly, they have a uniform height profile resulting in essentially a flat surface topology. This significantly reduces the challenges in alignment as one can adapt well-developed techniques that are widely used in semiconductor foundries\textsuperscript{84}. As such, these metasurfaces are well-positioned to act as a platform for designing dispersion-tailored optical components, and they can either function as independent optical elements or work in tandem with refractive optics to achieve achromaticity. In this Chapter, we first show that by judicious design of single-layered nanofins on a surface, it is possible to simultaneously control the phase, group delay and group delay dispersion of light, thereby achieving a transmissive achromatic metalens with a large bandwidth\textsuperscript{45}. This design does not involve spatial multiplexing or cascading. Secondly, we demonstrate how this technique allows one to design an aberration-corrected, ultra-compact meta-spectrometer with significantly improved spectral range of operation without needing any additional optical elements\textsuperscript{60}. This technique is general and can be widely employed in the design of other micro and nanostructures and has the potential to revolutionize the nascent field of metasurfaces.

5.2 Dispersion engineering in metalenses

As an initial example, consider the achromatic metalens shown in Fig. 5.1(a). The relative phase provided by the metalens elements (with respect to its center) follows Eq. (1.8), which is the standard hyperbolic phase profile for focusing at a given wavelength. Written explicitly to illustrate this frequency dependence, Eq. (1.8) becomes:

\[ \text{relative phase} = \frac{2\pi}{\lambda} \left(R - x^2 + y^2\right) \]

where \( R \) is the radius of curvature, \( x \) and \( y \) are the coordinates, and \( \lambda \) is the wavelength. This equation can be used to design metalenses with tailored dispersion characteristics.
\[ \varphi(r, \omega) = -\frac{\omega}{c} \sqrt{r^2 + F^2} - F \]  

(5.1)

where \( \omega, c, r \) and \( F \) refer to the angular frequency, speed of light, radial coordinate of the metalens, and its focal length, respectively. This spatial and frequency dependent phase profile \( \varphi(r, \omega) \) implies that at a given \( r \), the metalens provides different transverse wavevectors \( k_r = \frac{\partial \varphi(r, \omega)}{\partial r} \) so that different wavelengths of light are deflected by the same angle. Equation (5.1) can be Taylor-expanded near a design frequency \( \omega_d \) as follows:

\[ \varphi(r, \omega) = \varphi(r, \omega_d) + \frac{\partial \varphi(r, \omega)}{\partial \omega}\bigg|_{\omega=\omega_d} (\omega - \omega_d) + \frac{\partial^2 \varphi(r, \omega)}{\partial \omega^2}\bigg|_{\omega=\omega_d} (\omega - \omega_d)^2 + \cdots \]  

(5.2)

Equation (5.2) indicates that to achieve achromatic focusing within a given bandwidth \( \Delta \omega \) around \( \omega_d \), an optical element placed at a radial coordinate \( r \) needs to satisfy not only the zeroth order phase profile \( \varphi(r, \omega_d) \), but also the higher-order derivative terms which determine the dispersion of the metalens. \( \frac{\partial \varphi(r, \omega)}{\partial \omega} \) and \( \frac{\partial^2 \varphi(r, \omega)}{\partial \omega^2} \) are known as the relative group delay and group delay dispersion, and are typically on the order of femtoseconds (fs) and femtoseconds squared (fs\(^2\)) in the visible.

Conventional diffractive lenses only satisfy the required phase, that is, the phase profile at a design frequency. The neglect of these derivative terms results in chromatic effects. An intuitive interpretation of each term in Eq. (5.2) is shown in Fig. 5.1(a). The first term leads to a spherical wavefront (yellow line in Fig. 5.1(a)). The group delay term compensates for the difference in the arrival times of the wavepackets at the focus, while the higher-order derivative terms (group delay dispersion, and so on) ensure that the outgoing wavepackets are identical. The net effect is the minimization of the spread in the arrival times of wavepackets at the focus to ensure that they constructively interfere. The smaller the time spread, the larger the bandwidth achievable. Therefore, to realize diffraction-limited focusing for a broad bandwidth, both phase and group delay, as well as higher-order terms, need to be considered.
Figure 5.1: (a) Schematic of an achromatic metalens. To realize achromatic focusing, the phase profile \( \varphi(r, \omega) \) must satisfy Eq. (5.1). The metalens is designed to provide spatially dependent group delays such that wavepackets from different locations arrive simultaneously at the focus. The yellow line shows the spherical wavefront. (b) Required relative group delays as a function of metalens coordinate. The focal length is parametrized as \( F(\omega) = k \omega^n \). Note that the NA is a function of wavelength for \( n \neq 0 \) due to the change in focal length. Depending on the value of \( n \), the metalens can be designed as achromatic \((n = 0)\) or chromatic with focal length inversely proportional to wavelength \((n = 1, \text{dispersion similar to Fresnel lenses})\), or proportional to wavelength \((n = -1)\). The case of \( n = 2 \) exhibits stronger dispersion. These metalenses have a diameter of 20 \( \mu \text{m} \) and a focal length of 49 \( \mu \text{m} \) at \( \lambda = 530 \text{ nm} \). (c) Required relative group delay dispersion of the same metalenses.

To account for the dispersion of a metalens, the focal length \( F \) in Eq. (5.1) can be parametrized as:

\[
F = k \times \omega^n \tag{5.3}
\]

where \( k \) is a positive constant and \( n \) is a real number. The dispersion of a metalens can thus be
designed to arbitrary specifications by substituting different values for \( n \). The metalenses with \( n \) equal to 0 and 1 are referred to as achromatic and diffractive metalenses hereafter. The diffractive metalens possesses a focal length shift similar to Fresnel lenses. From Eq. (5.3), the positive (negative) values of \( n \) imply that shorter (longer) wavelengths are focused farther from (closer to) the metalens, respectively. The larger the absolute value of \( n \), the farther the separation between the focal spots of two wavelengths, resulting in stronger dispersion. Figures 5.1(b), (c) show the required relative group delays and group delay dispersions as a function of radial coordinate for metalenses with numerical aperture (NA) = 0.2 at \( \lambda = 530 \text{ nm} \). For \( n = 2 \) and \(-1\), they require nonnegligible group delay dispersion to precisely control the focal length shift and achieve diffraction-limited focusing. Note that the required group delay and group delay dispersions are relatively small for \( n = 1 \). This agrees with the previous observation that a diffractive metalens (NA = 0.8) implemented using geometric phase can still focus light with a focal spot size approximately equal to a wavelength\(^{30}\).

In order to implement dispersion engineering in practice, the number of degrees of freedom in the constituent elements needs to be increased. We utilized coupled phase-shift elements comprised of two nanofins in close proximity, which behave as coupled waveguides. Their geometrical parameters are defined in Fig. 5.2(a), and scanning electron microscope images of a fabricated metalens are provided in Fig. 5.2(b). It has been previously shown that coupled waveguides can support tunable dispersion, for example, near-zero group delay dispersions for a wide bandwidth\(^{186-187}\). The optical properties of a nanofin has been given in Eq. (2.1); note that the phase acquired by the cross-polarized component of light is determined only by the rotation angle of the nanofin and is independent of all other parameters. Thus, the target phase profile is essentially decoupled from the required group delay and group delay dispersion, which are controlled by the term \( \tilde{t}_l - \tilde{t}_g \). Figure 5.2(c) shows phase spectra for a nanofin with different rotation angles. The slope is approximately linear within a given bandwidth and is independent of the rotation angle of the nanofin. This property allows one to design achromatic metalenses with a large bandwidth.
Figure 5.2: (a) Schematic of a metalens element. The element consists of one or more TiO$_2$ nanofins of varying dimensions but equal height $h = 600$ nm, evenly spaced by a distance $p = 400$ nm. The gap between nanofins is $g = 60$ nm. The length $l$, width $w$, height $h$ and rotation angle $\alpha$ are also shown; the subscripts denote the left and right nanofin, respectively. The nanofins are rotated with respect to the center of the square ($400 \times 400$ nm$^2$). (b) Scanning electron micrograph of a region in the fabricated metalens. Scale bar: 500 nm. (c), (d) Simulation results for a single nanofin. (c) Phase plots as a function of frequency for different rotation angles for nanofins with $l = 250$ nm and $w = 80$ nm. (d) A comparison of polarization conversion efficiency for different nanofin lengths from FDTD calculations (solid lines) versus MODE Solutions (dashed lines). The lengths of the nanofins are labelled; they have a constant width $w = 80$ nm. (e) Phase spectra and polarization conversion efficiencies for five different elements showing the tunability of the group delay by changing the lengths and widths of nanofins. The shaded region marks the design bandwidth of 120 nm. Each colored curve corresponds to its element schematically shown on the right. The parameters $(l_1, w_1, l_2, w_2)$ of each nanofin are labelled; all dimensions are in nanometers. The elements in the colored squares are located at different radial positions from the edge of the metalens (red square) to the center (purple square), such that the corresponding group delay (slope of the phase versus angular frequency plot) increases from the edge to the center. This ensures achromatic focusing as illustrated in Fig. 5.1(a).

In order to obtain further physical insight into the dispersion design, each TiO$_2$ nanofin can be regarded as a truncated waveguide. Neglecting end reflections, the phase of the transmitted light after passing through the structure at a given coordinate $r$ is $\phi(r, \omega) = \frac{\omega}{c} n_{\text{eff}} h$, where $n_{\text{eff}}$ and $h$ represent the effective index and height of the nanofin, respectively. The derivative with respect to frequency

$$\frac{\partial \phi(r, \omega)}{\partial \omega} = \frac{1}{c} n_{\text{eff}} h + \frac{\omega}{c} \frac{\partial n_{\text{eff}}}{\partial \omega} h$$

(5.4)

yields the group delay: this is the ratio of the nanofin height to group velocity, which can be controlled by the nanofin dimensions and/or material used. Figure 5.2(d) shows a comparison of polarization...
conversion efficiency using the eigenmode solver and finite-difference time-domain (FDTD) methods. The good agreement verifies the validity of treating the nanofins as short waveguides. At higher frequencies, the observed deviations result from the excitation of higher-order modes and resonances within the nanofins. Figure 5.2(e) shows the phases and polarization conversion efficiencies of five different nanofin elements. Their group delays were obtained using linear fitting of the phase spectra within a bandwidth of 120 nm, centered at 530 nm. This ensures that the group delay of an element fulfills the requirement shown in Fig. 5.1(b) and its group delay dispersion is close to zero for at least the 120 nm bandwidth being considered. However, as seen later in this Chapter, simulations and experimental results show that the metalens focal length is only weakly dependent on wavelength beyond this bandwidth, up to 670 nm. Note that for low NA, the required range of group delay is proportional to the product of lens radius and NA. We designed and implemented metalenses with nanofin dimensions corresponding to a group delay range of about 5 fs; see Appendix D.1 for a plot of polarization conversion efficiencies versus group delays.

5.3 Achromatic focusing and imaging

To demonstrate the versatility of this approach, we designed and fabricated an achromatic metalens \((n=0)\) as well as two other metalenses with \(n = 1\) and 2. They all possess a NA = 0.2 at wavelength \(\lambda = 530\) nm. For \(n = 1\), that is, a regular diffractive metalens, the phase profile was imparted by identical nanofins using the geometric phase. The achromatic metalenses were designed by digitizing the required phase and group delay, which were then implemented by selecting elements from a library of various nanofin parameters. For \(n = 2\), we also selected elements with group delay dispersion close to the required ones. The measured normalized focal length shifts and their theoretically predicted values from \(\lambda = 470\) nm to 670 nm are shown in Fig. 5.3(a). The latter was calculated by propagating the fields generated by the nanofins using Fresnel–Kirchhoff integration, neglecting the actual
Figure 5.3: Measured focal length shifts and intensity distributions of metalenses. (a) Experimentally measured normalized focal length shifts (symbols) compared to simulations (lines) for metalenses with different values of \( n \). Incident light was collimated and at normal incidence. The metalenses were designed at wavelength \( \lambda = 530 \) nm with NA = 0.2 and focal length of 63 \( \mu \)m. (b) – (d) Measured intensity distributions in linear scale (in false colors corresponding to their respective wavelengths) in the \( x-z \) plane. Panels (b), (c) and (d) correspond to metalenses with \( n = 0 \) (achromatic), 1 and 2, respectively. The wavelengths of incidence are denoted on the left. The direction of incidence is towards the positive \( z \)-axis. (e), (f) Normalized intensity profiles along the white dashed lines of (c) and (b) for diffractive and achromatic metalenses, respectively. The white dashed lines pass through the center of focal spots in the case of \( \lambda = 470 \) nm. Scale bars: 2 \( \mu \)m.
coupling between metalens elements. Experimentally, the focal lengths at different wavelengths were obtained by measuring their intensity profiles (point spread functions) along the propagation direction (z axis) of the incident beam in steps of 1 μm, as shown in Fig. 5.3(b), (d). The z coordinate corresponding to the peak intensity value gives the focal length for a given wavelength.

The performance of these diffractive and achromatic metalenses were characterized in terms of their focal spot profiles (Fig. 5.3(e), (f)). They were measured at the focal plane corresponding to an illumination wavelength of 470 nm (white dashed lines in Fig. 5.3(b), (c)). The diffractive metalens shows significant defocusing when the wavelength is larger than 550 nm (Fig. 5.3(e)). In contrast, the focal spots of the achromatic metalens at different wavelengths are diffraction-limited: their Strehl ratios are larger than 0.8 and the deviations of the full-width at half-maximum (FWHM) are within 5% of the theoretical values (see Appendix D.2). It is important to note that although this achromatic metalens was originally designed for a bandwidth of 120 nm centered at 530 nm, due to the negligible focal length shift compared with its depth of focus (λ/NA^2), the metalens maintains its focal spot profile for almost the entire visible spectrum from 470 nm to 670 nm. This achromatic metalens can also focus incoherent white light under a broadband halogen lamp illumination (see Appendix Fig. D.3).

We also fabricated achromatic and diffractive metalenses with larger diameters (NA = 0.02, diameter = 220 μm) and compared their imaging qualities. The focal spot profiles of the achromatic metalens are shown in Fig. 5.4(a). Figure 5.4(b) shows the images of a standard United States Air Force resolution target obtained from the achromatic metalens, under various laser illumination wavelengths with a bandwidth of 40 nm, corresponding to the bandwidth of a typical LED light source. Control experiments using the diffractive metalens are shown in Appendix D.4. The test target was fixed at the focal plane corresponding to illumination at λ = 470 nm. In Fig. 5.4(b), a slight decrease of contrast in the images at red wavelengths is observed since the feature size of the target (~15 μm) is close to the diffraction limit of the achromatic metalens; there is also a decrease in efficiency of the metalens at red wavelengths.
Figure 5.4: Focal spot profiles and imaging using an achromatic metalens under different illumination wavelengths. The achromatic metalens has a diameter of 220 μm and NA of 0.02. The light source is a supercontinuum laser with tunable center wavelength and bandwidth. (a) Experimentally measured focal spot profiles. The illumination wavelengths with about 5 nm bandwidth are labeled on the top. Scale bars: 20 μm. (b) Images of 1951 United States Air Force resolution target formed by the achromatic metalens. The resolution target (Thorlabs, R1L1S1N) was fixed at the focal plane corresponding to an incident wavelength $\lambda = 470$ nm. The line widths of the upper and lower rows of bars are 15.6 and 14 μm, respectively. Scale bar: 100 μm. (c) Images of the same United States Air Force target using the achromatic metalens under an illumination bandwidth of 200 nm centered at 570 nm. (d) The image of the Siemens star pattern under the same illumination condition in (c). The outer radius is 500 μm. The metalens provides about 50× magnification. Scale bars, 200 μm. Appendix Fig. D.4 shows that a chromatic metalens of same diameter and NA generates significantly blurred images, in contrast to the achromatic lens.

Additionally, we demonstrate white-light imaging using a broadband illumination source (white-light laser) from 470–670 nm. The images of the United States Air Force target and Siemens star are
shown in Fig. 5.4(c), (d). They show that the chromatic aberration is well-corrected even under white-
light illumination, and that the metalens can achieve high imaging quality over a few square
millimeters, corresponding to a 30° field of view. Note that the patterns at the center of the United
States Air Force target and the Siemens star have feature sizes smaller than the resolution of the
achromatic metalens. The focal spots of the metalens for different angles of incidence were measured
and found to possess Strehl ratios larger than 0.8, up to an incident angle of about 15° (see Appendix
D.5). The color of these white-light images has a bluish tinge because of the wavelength-dependent
efficiency of the metalens. We measured about 20% efficiency around 500 nm, against a theoretically
predicted value of 50% (see Appendix D.6). This deviation likely results from fabrication errors and
the coupling between metalens elements. The latter results from the fact that the phase of each element
(coupled waveguides) was obtained via simulations using periodic boundary conditions. This
approximation ignores near-field coupling to adjacent metalens elements, thereby introducing a
perturbation on the wavefront and an attendant reduction in efficiency. This effect can be taken into
account with comprehensive full lens simulation followed by optimization to increase efficiency. Note
that the metalens efficiency is lower compared to previous works30,46, because to cover a larger range
of group delay, some low polarization conversion efficiency elements must be chosen (see Appendix
D.1). The metalens efficiency can be increased by introducing more complicated nanostructures to
increase the freedom of design parameters, or by choosing highly efficient elements at the expense of
reducing metalens diameter. To illustrate this point, we also fabricated and measured two metalenses
with smaller diameters with efficiencies of about 40% (Appendix Fig. D.6).

This design principle can be applied to other regions of the electromagnetic spectrum. In addition,
as mentioned previously, realizing achromatic metalenses with larger diameters and higher numerical
apertures requires a larger range of group delay supported by various combinations of nanofins with
different dimensions. This can be realized, as shown in Eq. (5.4), by different dispersion engineering
approaches or by simply increasing the height of the nanofins. The authors of Ref188 have
demonstrated TiO₂ nanostructures with ~4.5 μm height corresponding to a group delay of about 37 fs.
Cascading layers of metalenses can further increase the total group delay; moreover, this introduces an additional degree of freedom to correct for monochromatic aberrations (coma, field curvature, astigmatism, and so on) within a large field of view\textsuperscript{189-190}. Finally, one can combine a metalens that acts as an aberration corrector with a refractive spherical lens. This is particularly promising as one would be able to simultaneously correct chromatic and monochromatic aberrations of the spherical lens, while maintaining the benefits of a large lens aperture and a small chromatic focal length shift. An example is given in Appendix D.7 for a commercial low-cost spherical lens (LA4966, Thorlabs Inc.) with a diameter of 5.4 mm and NA = 0.1 attached to a dispersion-engineered metalens. Calculations show that the focal length shift of the refractive metalens doublet is similar to that of triplet lenses if one incorporates group delay dispersion engineering in the design of the metalens. Moreover, the metalens enables the spherical lens to achieve diffraction-limited achromatic focusing across the visible spectrum from 450 nm to 700 nm, a feat that traditionally requires cascading of many refractive lenses of various glass materials\textsuperscript{181}. The required group delay for such a metalens is about 22 fs, only four times larger than what we can realize with 600-nm-height nanofins. A plot of the achievable group delay and group delay dispersions within the current library is presented in Appendix D.8.

5.4 Aberration corrected spectroscopy with metalenses

In Chapter 3, we saw how metasurfaces could be efficiently utilized to simultaneously focus and disperse light, thereby integrating the functionality of both the grating and focusing lens in a single optical component. In addition, by engineering the off-axis focusing angle, very large dispersions could be obtained; this allows one to achieve a miniature spectrometer with sub-nanometer resolution with a working distance of only a few centimeters from the lens to the detector\textsuperscript{55}. It should be noted that this working distance is not fundamentally limited in any way; it is dependent on the desired specifications of the metalens, and in principle could even be much smaller, enabling monolithic
integration with detectors.

However, these off-axis focusing metalenses face the same challenges as their refractive and diffractive counterparts in terms of possessing an intrinsically curved focal plane and experiencing aberrations such as astigmatism and field curvature for wavelengths different from the design. This fundamentally limits the spectral resolution and the spectral range of operation. Here we show how dispersion engineering techniques, such as those described in Chapter 5.2, can be applied to off-axis metalenses such that they can maintain nanometer resolution across approximately the entire visible spectrum.

Figure 5.5: (a) Schematic of a coupled nanofin structure with unit cell size \( p \), and length, width, and height characterized by \( l, w, \) and \( h \), respectively. The latter parameters are varied to obtain a library of elements with different dispersion properties. The entire unit cell can be rotated by an angle \( \alpha \) to provide a phase shift of \( 2\alpha \) for left-handed circularly polarized light, using the geometric phase principle. (b) Schematic of an off-axis metalens comprising these coupled nano-fin structures. The focal spot is located at \((x_f, 0, z_f)\). We also define a primed coordinate system centered on the focal spot. The rainbow-colored line indicates the flat focal plane of the lens for different wavelengths in the visible. (c), (d) Scanning electron micrographs of a fabricated aberration-corrected off-axis metalens. Note that the direction with the largest phase gradient is along the horizontal \( x \)-axis. Scale bars: 1 \( \mu \)m

Figure 5.5 shows the schematic images and scanning electron micrographs of an aberration-corrected, off-axis metalens. It was fabricated by electron beam lithography and atomic layer
deposition of titanium oxide (see Appendix A.1). Its constituent unit cell \((400 \times 400 \text{ nm}^2)\) comprises one or more rectangular nanofins of height 600 nm (Fig. 5.5(a), (b)). The gap between the nanofins is fixed at either 60 or 90 nm. As detailed in Chapter 2.1, due to its form birefringence, each successive unit cell can be rotated to impart the desired phase using the geometric phase concept. Additionally, multiple nanofins in close proximity within a unit cell behave analogously to coupled waveguides and can support electromagnetic modes with tunable dispersion\(^{186-187}\) (see Chapter 5.2). Their optical properties are determined by varying the length \((l)\) and width \((w)\) of the fins (Fig. 5.5(a)). This choice of elements enables us to decouple the output phase of light from GD and its higher order derivatives, so that we can independently specify both a target phase profile as well as its behavior at wavelengths different from the design.

The off-axis metalens has dimensions of \(800 \times 250 \mu\text{m}\) with a focal length \((f)\) of 4 cm and focusing angle \((\theta)\) of 25° at the design wavelength of 470 nm (Fig. 5.5(b)). This acts as both the focusing and dispersive element in a traditional spectrometer since different wavelengths of light are focused at different spots along the plane of the detector, enabling one to obtain spectral information. This is schematically illustrated by the rainbow-colored bar in Fig. 5.5(b). Scanning electron micrographs of the fabricated device are shown in Fig. 5.5(c), (d). Note that the metalens is essentially a rectangular cut from a circular metalens with a radial phase profile; as a result, phase gradients are present along both \(x\) and \(y\) directions. However, by design the primary focusing will be along the \(x\)-axis. The phase gradient imparted along \(y\) is therefore small, as evidenced by the small difference in rotation angles of the unit cells in the vertical direction (Fig. 5.5(c), (d)).

In order to achieve off-axis focusing, the phase profile of the metalens must satisfy Eq. (3.1). Similar to Chapter 3.1, \(x_f = f(\omega)\sin\theta(\omega)\) and \(z_f = f(\omega)\cos\theta(\omega)\), where we have written out explicitly the frequency dependence of both the focal length \(f\) and the off-axis angle \(\theta\). Since we intend to design it as an aberration corrected metalens, we wish to tailor \(f(\omega)\) such that all the focal spots for different wavelengths lie along a plane orthogonal to the \(x\)-axis and parallel to the \(z\)-axis. This choice
Figure 5.6: Ray-tracing simulations of (a) a regular Berry-phase lens and (b) the aberration-corrected metalens. The metalenses were designed with focal length $f = 40$ mm and focusing angle $\alpha = 25$ degrees at wavelength $\lambda = 470$ nm. The focusing planes for each case are indicated by bold lines; the dashed line in (a) is horizontal and meant as a reference to the curved focal plane. (c) Diagrams of the focal spots at their respective focal planes calculated from ray-tracing for the Berry phase lens and (d) aberration-corrected metalenses, respectively, under wavelengths of 470, 530, 590, and 660 nm illumination. The oval outline represents the diffraction-limited focal spot size; it is oval due to the rectangular dimensions of the metalens. Labeling of the coordinates follows the scheme shown in Figure 5.5(b). The asymmetric broadening of the focal spot along the $z'$-axis is due to astigmatism as the incident wavelength changes.

makes alignment easier and prevents background light from reaching the detector. Thus one can choose an initial design frequency $\omega_d$, which then establishes the form $f(\omega)$ and $\theta(\omega)$ should take:

$$f(\omega)\sin\theta(\omega) = f(\omega_d)\sin\theta(\omega_d)$$

Here $\omega_d$ was chosen to be 470 nm. As a result, one can design the dispersion as $f(\omega) = \frac{\omega}{\omega_d} \cdot f(\omega_d)$; and $\sin\theta(\omega) = \frac{\omega_d}{\omega} \cdot \sin\theta(\omega_d)$. This choice of focal plane ensures that minimal dispersion engineering
is needed along the $y$-direction, so that the metalens can be scaled up to match the size of entrance slits of spectrometers in order to capture light more efficiently (see Appendix D.9). Equation (5.5) thus dictates the frequency dependent phase profile of an off-axis aberration-corrected metalens, which is noticeably different from prior metalens spectrometers. This phase profile can be implemented by Taylor expanding Eq. (3.1), in a manner similar to that shown in Chapter 5.1 (Eq. (5.2)). As discussed previously, by an appropriate choice of parameters for the coupled nanofins within a unit cell, one can separately implement both the desired phase and GD/GDD profiles.

Figure 5.6 compares the simulated results of an off-axis metalens designed using the procedure described above to that of a traditional Berry phase lens where only the phase profile (zeroth-order derivative term in Equation (5.2)) is implemented. The Berry phase lens represents a control sample for the case without dispersion engineering and performs similarly to an off-axis focusing Fresnel lens. Both lenses have the same dimensions and specifications. From the ray-tracing results in Fig. 5.6(a) and (b) (calculated using Zemax OpticStudio), one observes that the Berry phase lens focuses along a curved surface while the aberration-corrected metalens focuses along a plane orthogonal to the $x$-axis, accurately fulfilling the design based on Eq. (5.5). The latter thus avoids the problem of focal spot broadening due to projection on a planar surface, such as a camera. It is important to note that by implementing the GD and GDD terms (Eq. (5.2)), one also corrects for the wavelength dependent aberrations, such as astigmatism, of the Berry phase lens. Astigmatism occurs in this case due to different foci of sagittal and tangential rays (incident rays passing through the $x$- and $y$-axes of the lens, respectively) as incident wavelengths deviate from the design. In an off-axis configuration, changes in incident wavelength will result in unequal shifts of the sagittal and tangential focal planes and contribute to an asymmetric broadening of the focal spot. This is observed in Fig. 5.6(c), as the focal spots at wavelengths away from the design wavelength of 470 nm rapidly broaden and become aberrated along the $z'$-direction. Consequently, they are no longer diffraction limited beyond 530 nm, which would severely limit the spectral range of a system using the Berry phase lens. In contrast, using our current approach (i.e., the aberration-corrected metalens) the phase profile remains close to the
ideal requirement at multiple different wavelengths due to the presence of the dispersion terms. As a result, we can achieve diffraction-limited focusing across the entire visible spectrum (Fig. 5.6(d)) while staying true to a single layer metasurface design, without the use of additional correcting optics.

Next we experimentally characterized the performance of our metalens, again using a traditional Berry phase lens as a control. The results are shown in Fig. 5.7. We illuminated each lens with collimated monochromatic laser light at 488, 532, 632, and 660 nm, and captured the resulting focal spots with a CMOS camera (Thorlabs DCC 1545M) situated along the $z'$-axis.

![Aberration corrected metalens](image)

![Berry phase lens](image)

**Figure 5.7:** Experimental characterization of the focal spots and their respective (normalized) intensity cross-sections of (a) – (d) the aberration-corrected metalens, and (e) – (h) the Berry phase lens used as a control under monochromatic laser illumination at 488, 532, 632, and 660 nm. Both lenses were fabricated on the same sample and the data were measured simultaneously using a single camera. Scale bars: 100 μm.
We observed that the focal spots of the aberration-corrected metalens remain well defined with a single peak for all four wavelengths across the visible (Fig. 5.7(a)–(d)), with a hint of broadening and asymmetry around 660 nm. In contrast, the focal spots of the Berry phase lens become significantly aberrated beyond 532 nm wavelength, with strong side-lobes appearing which would severely degrade the overall spectral resolution of the system (Fig. 5.7 (e) – (h)). These results agree well with simulations (see Appendix D.10). We note that close to the design wavelength (i.e., at 488 nm), the Berry phase lens has a higher quality focal spot with a slightly narrower full-width-at-half-maximum (FWHM). We attribute this to fabrication errors and coupling effects between neighboring unit cells in the aberration-corrected metalens since the structures are brought within closer proximity (compared to a single nanofin in the Berry phase lens) as they are rotated, thereby perturbing the optical modes and causing additional phase or dispersion variation not initially accounted for during the design. However, this can be mitigated by expanding the range of parameters of the simulation library in the design step. Furthermore, the difference in FWHM of the focal spots at 488 nm is only about 6 μm, which is approximately the size of one camera pixel (5.2 μm).

We subsequently characterized the dispersion and efficiency of the aberration-corrected metalens, as shown in Fig. 5.8. These properties were measured using a collimated supercontinuum laser source (SuperK Varia, NKT Photonics), with wavelengths ranging from 450 to 700 nm in steps of 5 nm. To obtain the dispersion, at each wavelength the focal spot position was captured using the camera and the shift in spatial position (compared to the focal spot at the previous wavelength) was recorded; the results are plotted in Fig. 5.8(a). We obtained a reciprocal linear dispersion ($\Delta\lambda/\Delta r$) of 0.013 nm μm$^{-1}$; this translates into an intrinsic spectral resolution of 0.77 nm at 488 nm, based on Eq. (3.5). In other words, by using a numerical aperture, aberration-corrected metalens with a working distance of 4 cm to the detector, we can achieve nanometer spectral resolution across 200 nm in the visible. More specifically, we find a spectral resolution of 0.96, 1.01, 1.06, and 1.14 nm at 488, 532, 632, and 660 nm incident wavelengths, respectively. The calculated spectral resolution across the entire bandwidth of interest is presented in Appendix D.11.
The efficiency of the metalens as a function of wavelength is defined as the power contained in the focal spot, normalized to the incident power passing through an aperture with the same size as the lens (800 × 250 μm) and the glass substrate. These values are generally low (<15% in Fig. 5.8(b)), partly due to the lack of degrees of freedom in the coupled nano-fin elements to independently optimize for efficiency, in addition to implementing the phase, GD and GDD profiles. Implementing an accurate phase and dispersion profile is the top priority since this dictates the resolution and performance of the lens. This results in a situation where different nanofins across the lens may have extremely different efficiencies due to the design algorithm. Consequently, to avoid excessive amplitude modulation which introduces spurious, unwanted diffraction orders, elements with nearly uniform amplitude across the design bandwidth were chosen at the cost of overall efficiency\(^{45}\). This problem can be mitigated by adopting more complicated structures than coupled nanofins, which would provide more degrees of freedom during design and optimization\(^{83}\). We also attribute the loss in efficiency to a slightly reduced accuracy of fitting the phase profile (due to the requirement of fitting higher order terms), as well as fabrication imperfections.

Beyond a highly compact spectrometer with nanometer resolution across a broad bandwidth,
some applications also call for customized dispersion characteristics. For example, in Raman spectroscopy a linear dispersion in the frequency domain is sometimes desired to simplify calibration procedures and minimize error. Although the current dispersion of the aberration-corrected metalens spectrometer appears to be almost linear in frequency (Fig. 5.8(a)), it is not rigorously so. This is apparent upon inspection of Eq. (5.5): the derivative of \( z \) (spatial coordinate of the focal spot) with respect to frequency is not a constant. A straightforward way to proceed is to rewrite both the focal length and focusing angle of the metalens such that the derivative of the focal spot \( z \)-coordinate with respect to frequency is now a constant, while ensuring that the focal spots for different wavelengths remain along a plane. However, this would require a completely new phase profile and the required GD and GDDs would be very large (up to 1000s of femtoseconds squared), which would be almost impossible to realize in a real device. A similar challenge exists for the design of spatially large metalenses with high numerical aperture.

Figure 5.9(a) shows one way to overcome this challenge: by inserting an additional correcting metasurface, one can maintain the metalens phase profile while introducing comparatively small changes to the GD and GDD to slightly perturb \( f(\omega) \) and \( \theta(\omega) \), such that the focal spot position along the focal plane is strictly linear with respect to frequency. In practice, this can be a doublet or simply another metasurface placed in free space, as long as the distance between the two components is taken into account during the design step.

The calculated focal length shift of such a doublet is plotted together with our previous result for the singlet aberration-corrected metalens in Fig. 5.9(b). Although the curves are closely spaced, which is expected since the original dispersion is approximately linear (Fig. 5.8(a)), some corrections to the focal spot positions are introduced by the doublet such that the final dispersion (orange line in Fig. 5.9(b)) becomes strictly linear. By design, we require the focal spot positions at the beginning and end of the operating spectral range of the doublet to be identical to the singlet (blue line in Fig. 5.9(b)): this would ensure that only the intermediate focal spot positions are being corrected for, which translates into minimal GD and GDD required for the additional metasurface corrector. The phase profiles of the
Figure 5.9: (a) Schematic of a doublet, comprising of a metasurface corrector and the original aberration-corrected off-axis metalens. The corrector serves to impart GD and GDDs such that the focal spot positions of the metalens are linear in frequency. (b) Plots of the focal spot positions of the singlet metalens (blue) and the doublet (orange) as a function of frequency. (c) Plot of the required phase as a function of frequency for an element at the edge of the metasurface corrector (blue line), and the results of second- and third-order polynomial fits (orange crosses and black circles, respectively). (d) GD and GDD values required for elements across the middle of the metasurface corrector, along x and y directions (blue circles and orange crosses). Inset: magnified view of the dispersion required for elements along y, which is minimal, due to the choice of the orthogonal camera plane.

metasurface corrector for various wavelengths were determined by using optimization tools in the ray-tracing software Zemax OpticStudio. Figure 5.9(c) plots the required phase (along the x-axis) at the edge of this metasurface as a function of frequency. Note that at the extremes of the spectral range of interest, the required phase is approximately equal since we wish to retain the original focal spot
positions of the singlet metalens. The orange crosses and black circles in Fig. 5.9(c) correspond to second and third-order polynomial fits to this target phase, respectively; these fitting coefficients in turn define the values of GD and GDD needed to implement the phase profile. Within our current library of elements, we are only able to reliably engineer up to second-order terms (GDD), resulting in some small but nonzero phase error. However, in our lens design methodology, we are usually able to limit the resulting wavefront error to less than $0.075\lambda$ (the Maréchal criterion) such that the focal spot is still diffraction limited.

A representative sample of GD and GDD values needed for the metasurface corrector is plotted in Fig. 5.9(d). Each point represents an element which was chosen to satisfy the target phase profile across the spectral range of interest. Note that the range of GD and GDD values needed are approximately 5 fs and 140 fs$^2$, respectively. Based on our existing library of nanostructures used in this work and others which have GD and GDD ranges of 5 fs and 10 fs$^2$, we believe that these requirements can be met using nanofabrication technologies\textsuperscript{195}, or by using a hybrid lens comprising of both diffractive and dispersion-engineered metasurface elements. Additionally, in the metasurface corrector the dispersion engineering primarily takes place along the $x$-axis; this is similar to the original design of the aberration-corrected metalens where the choice of the focal plane (parallel to the $z$-axis) ensures that minimal dispersion is needed along $y$ (inset of Fig. 5.9(d)).

It is important to note that by incorporating third-order corrections (see Appendix D.11) to the phase, one would be able to fit the target phase profile more accurately and eliminate the small phase errors mentioned previously. This would ensure diffraction-limited focusing across the entire spectrum of interest and can be accomplished by introducing additional degrees of freedom to the nanostructures. In the current design, we have made use of anisotropic nanostructures for the metalens. Consequently, the spectrometer works only for one helicity of circularly polarized light. For applications where polarization insensitivity is crucial, one can use instead isotropic nanostructures such as pillars or squares. A similar approach of using more complicated shapes would be necessary for the angular dispersion engineering of other devices such as Fourier or sub-resolution metalenses\textsuperscript{196}. 
Another intriguing possibility is the use of topology optimization to design freeform shapes with these desired functionalities.

It is also interesting to comment on the related problem of expanding the spectral range of the device to the ultraviolet and infrared regimes. From a design perspective, this necessitates the correction of even higher orders of dispersion (e.g., third and even fourth-order derivatives of phase with respect of frequency). While this might be prohibitive for a single metalens due to design complexity, this is possible by the same technique of using a metacorrector in conjunction with a refractive or diffractive optics lens, as discussed previously. From a practical point of view, this would require the nanofabrication of materials with a very large bandgap which would remain low-loss in the ultraviolet region, such as transition metal fluorides, fused silica, and nitrides.

5.5 Conclusions

In this Chapter, we have demonstrated that by simultaneously controlling the phase, group delay and group delay dispersion, one can achieve broadband achromaticity and correct for various aberrations in metalenses. This represents a significant advance in the state of the art for metalenses, which have traditionally been limited in their applications due to bandwidth, particularly in the visible. In addition, we have shown how this approach can correct for chromatically induced astigmatism and field curvature in metalens spectrometers and essentially engineer the focal length to be an arbitrary function of frequency. This could be extremely useful in sophisticated instruments such as aberration-corrected spectrometers, as well as metalens doublets. We believe that this technology can lead to exciting advances and applications not just in spectroscopy, but also consumer/wearable optics products in general.
A.1 Fabrication of high NA metalenses

The metalenses were fabricated by a process shown in Fig. A.1. Electron beam resist was spin-coated on a fused silica substrate to produce a layer with thickness $t_{\text{resist}}$. Control of this thickness is important because it sets the height of the final nanostructures. The resist was patterned using electron beam
Figure A.1: Clockwise, from top left: Electron beam resist is spin-coated onto fused silica (or equivalent transparent substrate) with thickness $t_{\text{resist}}$, which ultimately sets the height of the final structure. In the next step, the inverse of the final metasurface pattern is imprinted into the electron beam resist via electron beam lithography, and subsequent the pattern is subsequently developed. The boxed area is an expanded cross-section of the maximum feature width, $w$. Initial TiO$_2$ deposition is then performed via ALD, which conformally coats sidewalls and the top of the resist and exposed substrate (side view). The TDMAT molecule used for ALD is also shown. A completed ALD of the TiO$_2$ yields a film thickness greater than half the width of the maximum feature size, $t_{\text{film}} \geq w/2$. Subsequently, the tops of the TiO$_2$ metasurface and residual resist are exposed after reactive ion etching (a back-etching process) with a mixture of Cl$_2$ and BCl$_3$ ions (top and side view). Finally, the dielectric metasurface is fabricated upon removal of remaining resist via chemical solvents (top and side view). Lithography and subsequently developed in solution to remove the exposed resist, yielding a pattern that is the inverse of the final metasurface design (note: this is true only because a positive resist was used in the first step). The sample would then be transferred to an atomic layer deposition (ALD) chamber set at 90°C. The purpose of this temperature is twofold: it produces the desired amorphous phase in the deposited TiO$_2$, and prevents deterioration of the nanoscale patterns in the resist by keeping it well below the glass transition temperature. During the deposition, the gaseous TiO$_2$ precursor TDMAT coats all exposed surfaces conformally. Because this process fills the exposed features from both sides, the total ALD film thickness required is $t_{\text{film}} \geq w/2$, where $w$ is the maximum width of all gaps in the pattern. The residual TiO$_2$ film is then removed via back-etching in a reactive-
ion etcher via a mixture of BCl₃ and Cl₂ gases and planarized. The etch depth is equal to \( t_{\text{film}} \) so that the underlying resist is exposed; this is finally removed via solvents to yield the metasurface device.

A.2 Characterization of high-NA metalenses

The focal spots of the meta-lenses were characterized using a custom-built microscope shown in Fig. A.2 consisting of a fiber-coupled laser source, linear polarizer, quarter-waveplate, and Olympus objective (100×, NA = 0.9) paired with a tube lens (\( f = 180 \) mm) to form an image on a CMOS camera (Edmund EO-5012). The sources used were lasers (Ondax Inc.) with linewidths less than 100 MHz. For efficiency measurements, a supercontinuum laser (SuperK Varia) was used as the source. The efficiency is defined as the ratio of the optical power of the focused beam to the optical power of the incident beam, as captured by a photodetector (Thorlabs S120C) located at the same position as the CMOS camera. The incident optical power was measured as the light passing through an aperture (aluminum on glass) with the same size as the meta-lens.

![Figure A.2](image)

**Figure A.2:** Schematic of the experimental setup used for characterizing the size of focal spot and point spread functions of the metalenses. The incident laser beam is collimated by a fiber collimator (Thorlabs RC04APC-P01) with a beam size diameter of 4 mm. The collimated beam then passes through a Glan-Thompson polarizer (Thorlabs GTH10) and a quarter-waveplate (Thorlabs AQWP05M-600) to generate circularly polarized light. An Olympus objective (100× magnification, NA=0.9) was used to image the light focused by a metalens. A tube lens with focal length \( f = 180 \) mm was used to form an image on a CMOS camera (Edmund EO-5012).

A.3 Calculation of Strehl Ratio

The Strehl ratios as shown in Fig. A.3 are calculated by comparing the measured intensity distributions in the focal spots to the theoretically calculated Airy disk profiles, assuming diffraction-limited focusing with a full-width-at-half-maximum FWHM of \( \lambda/2 \) NA. The intensities of the measured profiles are then normalized to the calculated ones to achieve the same energy within a given area. The
Strehl ratio is given by the ratio of maximum peak value of the measured intensity curve to that of the calculated one. Strehl ratios of 0.76, 0.78 and 0.77 are achieved for metalenses designed for wavelengths of 405 nm, 532 nm and 660 nm respectively at their design wavelengths.

A.4 Imaging experiments with metalenses

For imaging, the metalens was paired with a tube lens with focal length of 100 mm. The schematic of the set-up is shown in Fig. A.4. A collimated beam was passed through a diffuser to reduce laser speckles before being focused by a Mitutoyo objective (10×) to illuminate the target object. The distance between the object and metalens was adjusted based on illumination wavelength due to the latter’s wavelength-dependent focusing characteristics.

To explore the imaging performances of the metalenses at different wavelengths, images of the USAF test chart as well as a Siemens star were used. The results are shown in Fig. A.5 and Fig. A.6 respectively. Figure A.7 provides a zoomed-in view of the resolving capabilities of the metalens when imaging the Siemens star.

A.5 Modulation transfer function (MTF) measurement

The MTF measurement was performed using a standard slant-edge test (Thorlabs R2L2S2P) and is shown in Fig. A.8. The MTF value of 0.1 is a good guide to the limit of the resolving power of the metalens. Figure A.8 shows that this occurs at the spatial frequency of approximately 1000 line pairs per millimeter (lp/mm). This cut-off value corresponds to a resolution limit of 500 nm ($10^6/2\times$ cut-off spatial frequency).
Figure A.3: Strehl ratio calculations: diffraction-limited (theory) and measured focal spot intensity distributions for three metalenses designed at wavelengths of (a) 405, (b) 532 and (c) 660 nm respectively. Focal spots are measured at the corresponding design wavelengths.
Figure A.4: Schematic diagram of the experimental setup used for imaging by the metalens. The laser beam is collimated by a fiber collimator (Thorlabs RC04APC-P01) with a beam size diameter of 4 mm. The collimated beam then passes through a Glan-Thompson polarizer (Thorlabs GTH10) and a quarter-waveplate (Thorlabs AQWP05M-600) to generate right circularly polarized light. This beam is passed through a diffuser to reduce speckles and then focused by a Mitutoyo objective (10× magnification, NA=0.28) onto the target object. The metalens is placed one focal length away from the object and paired with a tube lens (f=100 mm) to form an image on a CCD camera (Point Grey, GX-FW-28S5C-C). To reduce background signals, a polarizer paired with a quarter-waveplate in cross polarization was used.

A.6 Characterization of multispectral chiral lens (MCHL)

The dispersive (wavelength dependent) response of the MCHL was studied in detail by imaging the end facet of a single mode fiber with broadband light from a supercontinuum laser (SuperK Varia). By adjusting the distance between the camera and the object (i.e. the fiber), different wavelengths of light can be focused. This is shown in Fig. A.9. Figure A.10 shows the entire field of view as seen by the camera, showing that there is very little background in the images produced by the MCHL.

A.7 Imaging with the MCHL

The measurement setup for imaging the beetle is shown in Fig. A.11. Several light emitting diodes (LEDs) were used together with appropriate bandpass filters to uniformly illuminate the large (~ several centimeter) specimen. Both LCP and RCP images formed by the MCHL were collected in a single snapshot by the camera.

A.8 Circular dichroism measurement using the MCHL

The measurement setup for obtaining circular dichroism of the beetle is shown in Fig. A.12. Light from the supercontinuum laser is coupled into a fiber, then collimated and focused by a thin lens onto different parts of the beetle. The MCHL collects the scattered light and directs it onto the camera.
Figure A.5: Image of the 1951 USAF resolution test chart formed by the metalens designed at $\lambda_d = 532$ nm with diameter $D = 2$ mm, and focal length $f = 0.725$ mm taken with a Cannon DSLR camera at 11 wavelengths of (a) 480, (b) 550, (c) 580, (d) 590, (e) 620 and (f) 640 nm. Scale bar: 40 μm.
Figure A.6: Image of a Siemens star formed by the metalens designed at $\lambda_d = 532$ nm with diameter $D = 2$ mm, and focal length $f = 0.725$ mm captured by a Canon DSLR camera at wavelengths of (a) 480, (b) 500, (c) 540, (d) 580, (e) 600 and (f) 620 nm. Scale bar: 50 μm. The blurring near the center results from projecting the images onto a translucent screen.
**Figure A.7**: Image of a Siemens star formed by the metalens designed at $\lambda_d = 532$ nm with diameter $D = 2$ mm, and focal length $f = 0.725$ mm captured by a CCD camera at wavelengths of (A) 480, (B) 540, and (C) 620 nm. Scale bar: 10 $\mu$m. The features near the center of the star target can be well resolved.
Figure A.8: Measured modulation transfer function (MTF) as a function of spatial frequency in line pairs per millimeter (lp/mm). The slant-edge target was imaged using the metalens designed at $\lambda d = 532$ nm with diameter $D = 2$ mm and focal length $f = 0.725$ mm. The line is to guide the eye.
Figure A.9: Dispersive response of the multispectral chiral lens (MCHL). (a) – (f): Images formed by the MCHL. The object was the facet of a single mode fiber. Input polarization was linear and center wavelength and bandwidth were 550 nm and 100 nm, respectively. The camera distance from the MCHL was varied in order to selectively focus certain colors. Scale bar: 0.5 mm.
Figure A.10: Absence of higher orders and background noise. (a) – (c) Full field of view images corresponding to the cropped images shown in Fig. 2.8(c), (d) and (e) respectively. Scale bar: 2 mm.
Figure A.11: Experimental setup for imaging the beetle, which was placed at a distance of 18 cm from the MCHL. Images were captured by a color camera placed 3 cm from the MCHL. Band-pass filters used for measurements shown in Fig. 2.9 have center wavelengths of 532 nm, 488 nm and 620 nm, with a bandwidth of 10 nm.

Figure A.12: Experimental setup for circular dichroism measurement. Output of the fiber-coupled laser is collimated and then focused down by a thin lens on different parts of the beetle. The scattered light is collected by the MCHL.
B.1 Calibration of off-axis metalenses

The calibration curve for the off-axis metalenses with NA = 0.022 is shown in Fig. B.1. This is necessary as the dispersion (i.e. the rate of change of displacement of the focal spot along the camera detector) of the lenses is not strictly linear with wavelength, but is instead a polynomial. This curve
defines the $\Delta \lambda$ that each pixel of the camera corresponds to for different incident wavelengths. One could also obtain the dispersion from this data by taking the slope of the curve at different wavelengths.

B.2 Images of focal spots in metaspectrometer

Figure B.2 shows the images of the focal spots corresponding to the metalenses with various specifications (i.e. size, NA and polarization), as directly seen by the camera in the metaspectrometer configuration when illuminated with linearly polarized light from a supercontinuum laser with 5 nm bandwidth. The camera was placed such that the shapes of the focal spots from all lenses were optimized for both 532 nm and 660 nm incident wavelengths.

Figure B.1: Calibration curve for the $\text{NA} = 0.022$ off-axis metalenses. Wavelengths are varied from 480 nm to 780 nm in steps of 10 nm using the supercontinuum laser. Data points correspond to the center (highest intensity region) of the focal spots on the detector. The red curve is a fitted curve using a 5th degree polynomial.
Figure B.2: Camera images of focal spots produced under illumination from a supercontinuum laser with bandwidths of 5 nm after passing through a linear polarizer, at a center wavelength of (a) 532 nm and (b) 660 nm. In (a) the focal spots of metalens with NA = 0.1 (top) and NA = 0.022 (bottom) originate from metalenses labeled R1 and R2, respectively. In (b) both focal spots belong to the low NA metalenses (with opposite helicities: R2 and L2) since the focal spots form the high NA lenses are no longer visible due to their limited range of operation.

B.3 Comparison of metaspectrometer with commercial handheld spectrometer

Figure B.3 compares the measurement results of the bandwidth (defined by the full-width-at-half-maximum) of input laser light from a supercontinuum laser, using the metaspectrometer and a commercial handheld spectrometer (OceanOptics USB4000 UV-VIS). The metalens spectrometer offers comparable or better performance within the range 480 nm – 660 nm. This agrees with the results shown in Fig. 3.4 (a) – (d). Above 660 nm the FWHM increases rapidly and is not accurate due to aberrations in the focal spots.
Figure B.3: Full-width half maximum (FWHM) of the input light from the supercontinuum laser, as measured by the metalens spectrometer (black squares) and commercial handheld system (red circles).

B.4 Improved design of metaspectrometer with different camera

Figure B.4 illustrates how the design of the metaspectrometer can be improved with a camera with a large active region, e.g. the Hamamatsu S10141 – 1108S. This improved design uses a metalens with a focal length of 30 mm with a focusing angle of 15°, and has a NA of 0.008. It possesses an intrinsic (lens-only) resolution of 0.48 nm; together with the detector-limited resolution of 0.55 nm, a spectrometer using this metalens can have a resolution of approximately 1 nm. However, it can possess a spectral range of operation throughout the entire visible spectrum, from 400 – 800 nm. This resolution limit and spectral range is comparable to existing compact spectrometers which possess significantly longer beam propagation lengths.
Figure B.4: (a) Simulated ray-tracing plot of a metalens designed for the camera with large active region. Dotted line indicates the optimal position of the camera, where it is approximately tangent to the focal points of the metalens throughout the spectral range of operation. (b) – (g) Calculated spot diagrams showing that the focal spots throughout the visible range are contained within the diffraction limited Airy disk (black ellipse). The elliptical Airy disk results from the rectangular shape of the lens (500 by 800 microns).
Supplemental Material: Giant intrinsic chiro-optical activity in planar dielectric nanostructures

C.1 Magnetic dipole response

The magnetic dipole response in high index dielectrics have been well studied in the literature. It originates from vertical displacement current loops in dielectric structures with finite thickness. In order
to observe a magnetic dipole resonance for a given wavelength, it is easy to see that the corresponding thickness of the structure should be $\lambda/n_{\text{eff}}$, where $n_{\text{eff}}$ is the effective index of the electromagnetic mode. For dipole modes $n_{\text{eff}}$ is generally close to the refractive index of the material. This thickness requirement can be understood as the condition for a standing wave in the cavity formed by the structure, as shown in Fig. C.1(a) – (c). It is important to note that this magnetic dipole moment occurs in-plane, and for achiral structures it is orthogonal to the in-plane electric dipole moment.

In contrast, the electric (dipole) resonance wavelength depends primarily on the charge separation which occurs within the length of the cavity. Therefore, by tuning the aspect ratio (width : thickness) of the structure one expects to be able to tune the spectral separation between the electric dipole and magnetic dipole resonances. For cylindrical geometries a spectral overlap occurs at aspect ratio $\sim 2$. This results in high transmittance in the forward-scattering direction due to the backscattered radiation from the electric and magnetic dipoles being out of phase; at a single frequency point the backscattered response will be completely canceled and the particle will only scatter forward. This is known as Kerker’s condition. We reproduce this result in Fig. C.1(d) using TiO$_2$ cylinders. This physical understanding of the origin and behavior of the magnetic dipole moment in dielectrics can be used to design higher order multipoles as well.
Figure C.1: (a) – (c), Schematic illustration of typical structures used to observe strong magnetic dipole resonances in high index dielectric materials, such as (a) a sphere, (b) cylinder/disk, and (c) a rectangular fin. In each of these cases the incident light excites a vertical displacement current loop ($j$) in the structure, which in turn generates an in-plane magnetic moment ($m$). (d) Colormap of the transmittance of a TiO$_2$ cylinder as a function of its height (fixed radius 130 nm). The spectral positions of the magnetic and electric dipole resonances can be tuned by adjusting the height and length of any given structure, i.e. their relative spectral separation is a function of the aspect ratio. There exists one frequency where the backscattered radiation from the magnetic and electric dipoles interfere destructively and completely cancel, resulting in high transmittance.

C.2 Role of waveguide layer and guided mode resonance

Figure C.2 illustrates the physics behind guided mode resonances occurring in the gammadion structures and underlying TiO$_2$ layer. The period of the gammadions (500 nm) is chosen such that diffraction occurs in transmission at the operating wavelength of 540 nm. Part of the incident light is coupled into the TiO$_2$ thin film which acts as a waveguide. This coupling of diffractive and waveguide modes results in guided, or leaky, mode resonances. To further confirm this fact, a simulation was performed with linearly polarized light (electric field perpendicular to the long axis) incident on a grating comprised of rectangular structures, reveals the presence of sharp spectral features at 510 nm and 570 nm, characteristic of guided mode resonances, as well as modulation of the transmittance.
envelope around 540 nm (where the transmittance ~ 100%). The presence of these spectral features result in an improved transmittance contrast. For the case of a chiral grating, light is coupled into the guided mode resonance and exhibits a transmittance dip for one helicity (here LCP) but not the other, due to their refractive index difference. In this way the use of an additional waveguide layer enhances the transmittance contrast between the two helicities and thus increases dichroism.

**Figure C.2**: (a) Schematic illustration (side-view) of a periodic array of gammadion nanostructures on a TiO₂ thin film supported by a silica substrate. The interference between diffracted and guided waves are shown. (b) Simulated transmittance spectrum of a TiO₂ grating (normalized to source power), comprising infinitely long rectangles on the same TiO₂ thin film and substrate. The widths of the stripes were chosen such that the fill factor was approximately the same as that of the gammadions. Illumination with linearly polarized light reveals the presence of sharp spectral features characteristic of guided mode resonances, as well as modulation of the transmittance envelope.

**C.3 Fields within the gammadion structure without waveguide layer**

Figure C.3 shows that qualitatively, the same multipolar excitations exist within the gammadion structure even without the underlying TiO₂ waveguide layer. In other words, the chiral response orginates due to the geometry of the structure, while the waveguide layer only serves to improve the transmission contrast and circular dichroism, as shown in Fig. C.2. Similarly, Fig. C.4 plots the simulated values of transmittance achieved for both helicities of incident light, with and without the waveguide layer. Again, we see that the circular dichroism and overall transmission efficiency for RCP is improved with the addition of the waveguide layer, but the effect still exists and is strong even without it.
Figure C.3: Magnetic field profile ($\text{Re}(H_z)$) of gammadions without the TiO$_2$ waveguide layer. Dominant multipole excitations can be seen to be the same as Fig. 4.3(c) and 4.3(d), i.e. quadrupolar (a) and octupolar (b) under left and right circularly polarized incident light respectively.

Figure C.4: (a) Simulated transmittance (black) and circular dichroism (orange) achieved with our chiral structures between 500 nm – 600 nm, as a function of waveguide layer thickness. The structure achieves maximum transmittance and circular dichroism at 300 nm waveguide thickness. (b) Optimizations of the structure as a function of its size (here the ratio of length to width is kept at 5:1) without the waveguide layer. The chiro-optical response is still evident but both transmittance and circular dichroism are weaker due to the lack of the waveguide layer which modulates the transmittance envelope.

C.4 Full multipole decomposition

Figure C.5 shows the full results of multipole decomposition, from the lowest order dipole terms to the electric and magnetic octupole terms. This complements Fig. 4.4(a) and 4.4(b).
Figure C.5: (a), (b) Zeroth order far-field intensity ($|E_x|^2$) for multipoles up to the magnetic/electric octupoles and toroidal quadrupole under (a) LCP and (b) RCP illumination. Note that the $y$-axis is in logarithmic scale.

C.5 Characterization of chiroptical response

Figure C.6 shows the expected transmittance spectrum and circular dichroism map derived from the summation of the multipole contributions shown in Fig. C.5 and Fig. 4.4(b). Primary contributions are the magnetic octupole and toroidal quadrupole modes.

Figure C.6: Characterization of chiro-optical response. (a) The simulated 0th order transmittance and individual multipole radiation spectra are overlaid. Excellent qualitative agreement is observed, indicating that the dominant multipoles in the system under RCP incidence are indeed the toroidal quadrupole ($T_Q$) and magnetic octupole ($M_O$). (b) Circular dichroism map of the gammadion structures as a function of thickness and wavelength, for a fixed length and width of 370 nm and 74 nm respectively. The optimized thickness is clearly seen to be 340 nm, due to the interference between the $T_Q$ and $M_O$ modes.
C.6 Chiroptical response when reversing direction of incident light

Experimental results for the transmittance and circular dichroism spectra through the sample are presented for both directions of incidence, i.e. forward and reversed. These are observed to be identical within experimental error, thereby proving that the observed chiroptical response is intrinsic in nature.

**Figure C.7:** (a) – (d), Experimental 0th order transmittance for (a) front and (b) back side incidence, as well as (c), (d) their circular dichroism spectra respectively. In the former case the light passes through the structure first and exits from the substrate, while in the latter the light passes through the substrate and exits from the structure. That these spectra are seen to be virtually identical is a hallmark of intrinsic chirality, here achieved with a planar structure.
Supplemental Material: Dispersion engineering and aberration control

D.1 Polarization conversion efficiency versus group delay of coupled nanofins

Figure D.1 plots the polarization conversion efficiency against group delay (first derivative of phase with respect to angular frequency) for a library of coupled nanofin elements. A filter was applied to remove the elements with conversion efficiencies lower than 5%. It can be observed that most of the
nanofins with high conversion efficiencies have group delays between 2 to 5 femtoseconds, as a result of the waveguiding effect being dominant (i.e. these values are limited by the height of the nanofin structures at 600 nm).

D.2 Focal spot characterization

Data for the Strehl ratio and full-width-at-half-maximum (FWHM) values for the achromatic metalens with a NA of 0.2 is shown in Fig. D.2. The corresponding focal spot profiles are shown in Fig. 5.3(f).

![Figure D.1: Simulated polarization conversion efficiencies and group delay for various coupled nanofin elements. Each dot on the graph represents an unique element with its x and y coordinates determined by the group delay and PC efficiency respectively. The group delay values are obtained by linearly fitting the phase plots at $\lambda = 530$ nm within a 120 nm bandwidth. Elements with R-squared values lower than 0.98 are dropped.](image)
Figure D.2: (a) Strehl ratio and (b) FWHM for the achromatic metalens with NA of 0.2. The dashed black line shows the theoretical FWHM of the Airy disk.

D.3 Focal spot under incoherent illumination

The focal spot profile for the achromatic metalens (NA = 0.2) under broadband incoherent illumination is shown in Fig. D.3. For additional verification, the spectrum of the light source was also measured using a spectrometer and presented in Fig. D.3. The focal spot size is observed to be slightly beyond the diffraction limit because the light guide used in the measurement is approximately 5 mm in diameter and the metalens is not corrected for the entire spectrum of the source. A pair of crossed circular polarizers was used in this measurement to remove the background.

D.4 Control experiment using achromatic and diffractive metalenses

Figure D.4 compares the measured focal spot profiles and images of the USAF resolution target using the achromatic and diffractive metalenses, which had identical NA = 0.02 and diameter of 220 μm under illumination with different wavelengths. One observes clear differences in the clarity and resolutions of the final images taken by the metalenses, indicating that the chromatic aberration is being corrected for in the achromatic metalens design.
Figure D.3: (a) Focal spot profile taken by a color CCD camera (UI-1540SE, IDS Inc.) for the achromatic metalens with NA = 0.2 under broadband incoherent illumination. (b) Normalized intensity of light source measured by a spectrometer (USB4000, OceanOptics) for a Tungsten source coupled to a light guide (OSL1, Thorlabs Inc.).

Figure D.4: (a) and (c) Focal spot profiles of achromatic and diffractive metalenses. Scale bar: 20 μm. These focal spot profiles were taken without re-focusing to visualize chromatic focal length shift. The bottom rows show intensity along the horizontal cut through the center of each focal spot. The illumination wavelength (with a bandwidth of about 5 nm) is denoted on the top. (b) and (d) Imaging using the metalenses. Schematic set-up is shown in Fig. S11. These images were taken at the focal plane for wavelength λ = 470 nm. Scale bar: 100 μm.
D.5  Focal spot characterization under oblique incidence

Figure D.5 presents the results of focal spot characterization and analysis for the achromatic metalens (NA = 0.02, diameter = 220 μm) under different angles and wavelengths of incidence. Note that the Strehl ratio indicates how close a focal spot is compared to the theoretical Airy disk profile. A Strehl ratio larger than 0.8 is commonly acknowledged as a requirement for diffraction-limited focusing.

![Figure D.5](image)

Figure D.5: (a) and (b) Experimentally measured full-width at half maximum (FWHM) and Strehl ratio for different angles of incidence and wavelengths. The theoretical values of FWHM are marked as dashed lines.

D.6  Efficiencies as a function of wavelength

The efficiencies of metalenses are studied and presented in Fig. D.6. The efficiency is defined by the power contained in the focal spot of the lens, divided by the power of light passing through an aperture with the same diameter. The former was measured by placing a power meter at the image plane of a
custom-built microscopy setup. An aperture with a size about the diameter of the Airy disk was used to filter out the background light. Theoretical values were obtained by taking the averaged polarization conversion efficiency of each element across the achromatic metalens. Although coupling effects between each nanofin element, as well as diffraction effects, are neglected, this gives an upper limit of efficiency.

![Figure D.6](image)

**Figure D.6:** (a) Measured (blue line) versus theoretical efficiencies (red and yellow lines) for the achromatic metalens (NA = 0.02, diameter = 220 μm). (b) Measured efficiencies of metalenses with the same NA of 0.02 but different diameters, which are labeled in the legend. The efficiency becomes higher for smaller diameters because the required group delay is smaller, which precludes the need for nanofin elements with low efficiency but high group delay (see Appendix D.1).

### D.7 Correcting aberrations using dispersion-engineered metalens

Using our approach, it is possible to correct not only monochromatic but also chromatic aberrations of a refractive lens. As an example, we chose a commercially available and low-cost plano-convex lens from Thorlabs Inc. Figure D.7(a) shows a raytracing diagram at wavelength λ = 530 nm. A metalens is attached to the planar side of the lens (depicted by the blue line). The diameter of the entrance aperture is 5.4 millimeters, and the refractive/metalens doublet has a numerical aperture of about 0.1. The frequency-dependent metalens was designed by the principle described in Fig. 5.1, i.e. it needs to provide various group delays and group delay dispersions such that all wavepackets from different lens
coordinates arrive at the focus together and with the same pulse shape. The green and black curves in Fig. D.7(b) show focal spot intensity profiles with and without the metalens, respectively. The metalens corrects the spherical aberration of the refractive lens by introducing a W-shaped phase profile similar to the well-known Schmidt plate (see the green curve in Fig. D.7(c)). Moreover, the phase profile of the metalens changes as a function of wavelength to correct chromatic aberration simultaneously. The required range of group delay and group delay dispersion for this frequency dependent phase profile is shown in Fig. D.7(d). These required values are only a few times larger than those provided by our current library shown in Fig. D.8(a) and can be readily achieved with modifications of the nanostructure design. The focal length shift is about 700 μm for the uncorrected lens, i.e. without the metalens (the orange curve of Fig. D.7(e)). Intriguingly, if one only corrects for group delay (see the red curve of D.7(e)), the focal length changes by an amount similar to that of an achromatic doublet. Taking the group delay dispersion into account results in a performance close to a triplet lens, as shown in the blue curve of Fig. D.7(e). The metalens-corrected spherical lens is now achromatic and diffraction-limited from 450 nm to 700 nm, as seen in Fig. D.7(f) with \( <0.075 \cdot \lambda \) root-mean-square wavefront error.
Figure D.7: Simulation results for aberration correction using a metalens. The refractive lens is a generic spherical lens available from Thorlabs Inc. The metalens corrector was designed with a frequency dependent phase profile, using the method described in the text. (a) A raytracing simulation of the refractive/metalens doublet at $\lambda = 530$ nm. The simulation was done using a commercial software OpticsStudio (Zemax Inc.). The layout of the refractive lens was obtained from Thorlabs’ website. (b) Focal spot intensities with and without the metalens. The spherical lens suffers spherical aberrations resulting in low Strehl ratio (black curve) away from the diffraction limit. (c) Phase profile across the center of the frequency-dependent metalens. The chosen wavelengths are shown in the legend; units are in nanometers. (d) The required group delay and group delay dispersion from the center to the edge of the metalens. (e) A comparison between relative focal length shifts for the refractive lens (orange), the metalens with engineered group delay only (red) and the metalens with simultaneously engineered group delay and group delay dispersion. (f) Root-mean-square wavefront error of the refractive/metalens doublet. The metalens provides group delay and group delay dispersion, showing the frequency-dependent phase profile in (c). The black dashed line shows a wavefront error of $0.075\lambda$, corresponding to a Strehl ratio of 0.8.

D.8 Group delay and group delay dispersion plot of nanofin elements

The group delay and group delay dispersions (GDD) achievable with our current library of nanofin elements in shown in Fig. D.8, together with the required values for the achromatic metalens...
mentioned in Section 5.2 and 5.3. It is worth noting that since only the relative group delay and GDD of the metalens need to be fulfilled, one can introduce two different offsets to these two quantities independently, which is equivalent to translating the data to achieve the best fit. Appropriate offsets are determined using the particle swarm method.

![Figure D.8](image)

**Figure D.8:** (a) Group delay (GD) and group delay dispersion (GDD) of elements (colored circles) versus that of the required GD and GDD (black circles) for realizing a metalens with \( n = 2 \). The coordinate of each purple circle represents the group delay and group delay dispersion of an element. The group delay and GDD were obtained by fitting the phase as a function of angular frequency using a quadratic polynomial for a bandwidth of 120 nm centered at 530 nm. Only the elements with R\(^2\) values larger than 0.99 are shown. Data after adding the offsets are shown by the green symbols. (b) and (c): Realized GD and GDD (green symbols) versus their required values (black lines).

**D.9 Required group delay and group delay dispersion for various camera angles**

The required group delay and group delay dispersions for different angles of the camera plane, with respect to the \( x \)-axis in Fig. 5.5, is shown in Fig. D.9. Note that for the camera plane perpendicular to the \( x \) axis, the dispersion required along the \( y \) axis of the meta-lens is minimized. This is important so that the size of the lens can be scaled up to match with the entrance slit size. Note also that since group delay and group delay dispersion are relative values, one can introduce offsets to our given library of nanostructures and search for the best fit.
**Figure D.9**: Group delay (horizontal axis) and group delay dispersion (vertical axis) required for the aberration-corrected off-axis meta-lens in the main text, along the x and y directions (blue and red symbols respectively) for (a)-(d) different angles of the camera plane. The scatter plots show the group delay, group delay dispersion and transmission efficiencies (color bar) of our entire library of coupled nano-fin elements.

**D.10 Simulated comparison of focal spots**

Figure D.10 shows the comparison, calculated via FDTD simulations, of a conventional Berry phase metalens and the aberration corrected metalens shown in Chapter 5.4. One can observe that significant aberrations occur for the Berry phase lens as the incident wavelength deviates from the design wavelength of 470 nm while the aberration corrected meta-lens remains diffraction-limited.
D.10 Simulated focal spots of the aberration corrected meta-lens (top panel) and Berry phase lens (bottom panel), at 470, 530, 590 and 660 nm wavelengths. Focal spots are false-colored by their respective wavelengths. Excellent agreement is observed compared to Fig. 5.7.

D.11 Higher order dispersion correction

Figure D.11 shows the dispersion values needed for the additional correcting metasurface in a doublet design, taking into account the 3rd order terms. This would lead to a more accurate fit of phase across the bandwidth of operation.

Figure D.11: (a) Fitted phase from nanofin elements (symbols) and required phase (line) as a function of frequency, compared to the case where the fitting was performed only up to 2nd order (GDD) terms. (b) GD and GDD values needed for a cut across the middle (along both x and y directions) of the correcting metasurface, which is similar to Fig. 5(c) in the main text; and (c) three-dimensional plot which includes the third order terms needed.
References


[68] V.-C. Su; C. H. Chu; G. Sun; D. P. Tsai, Advances in optical metasurfaces: fabrication and applications [Invited]. *Optics Express* 2018, 26 (10), 13148-13182.


