NOVEL OPTICAL METAMATERIALS, ABSORBERS, AND FILTERS
BASED ON PERIODIC NANOSTRUCTURES

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ABSTRACT

This research describes the design, fabrication, and characterization of novel optical metamaterials, absorbers, and filters based on periodic all-dielectric and metallo-dielectric nanostructures. With a properly chosen optical structure and its constituent materials, the geometry and dimensions were optimized to satisfy the user-specified criteria using a robust genetic algorithm. First, planar all-dielectric nanostructures were exploited to synthesize a mid-infrared (IR) filter and a mirror. An array of doubly periodic amorphous silicon (a-Si) blocks induced a guided mode resonance, resulting in a polarization insensitive and incidence angle tolerant single stop band at 3.0 μm. An effective medium approach was also applied to this planar all-dielectric nanostructure to produce a high efficiency mid-IR mirror centered at 3.3 μm. The fabricated filter and mirror structures were characterized using Fourier transform infrared (FTIR) spectroscopy to show excellent agreement with the predicted responses. Secondly, metallo-dielectric nanostructures were investigated to realize a zero index metamaterial (ZIM), a dispersion engineered metamaterial filter, and a metamaterial absorber. A free-standing optical ZIM, consisting of a gold-polyimide-gold tri-layer stack perforated by air holes, was synthesized at 1.55 μm with low absorption loss and a good impedance match to free space, verified through characterization using spectral holography. Based on the similar structure with minor modifications, a dispersion engineered broad band filter was demonstrated in the mid-IR wavelength range of 3-3.5 μm with a suppressed group delay. A conformal metamaterial absorber, patterned on a flexible polyimide and gold thin film stack, was also synthesized with a narrow band, polarization independent, incidence angle tolerant absorptivity centered at mid-IR wavelengths of 3.3 μm and 3.9 μm. The performance of the fabricated metamaterial filter and absorber was verified using FTIR, exhibiting excellent agreement with the simulated values.
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Chapter 1

Introduction

This chapter introduces the topic of the dissertation, which is the development of all-dielectric planar nanostructured filters and metamaterials that operate at infrared and optical wavelengths. A brief background for these two classes of devices is offered together with a discussion of the limitations and challenges of the current technologies and a summary of the research goals of the dissertation. The technical approach used in this research is also presented, and finally an overview of the subsequent chapters is provided.

1.1 Statement of Problem

Designed to interact with electromagnetic fields, optical materials or structures are widely used in a variety of scientific and engineering applications ranging from spectroscopy to solar cells at infrared (IR) and visible wavelengths [1-2]. In order to maximize performance, optical materials must be carefully selected to meet the specific requirements of any given application. When natural materials are insufficient for realizing certain functionalities, artificially engineered optical structures may offer a solution. By arranging structured materials in a random or orderly manner, it is possible to control the coupling between an incident light and a structured medium in order to produce the desired optical properties. During the last 20 years, with the aid of well-developed semiconductor fabrication methods, many useful photonic nanostructures have been proposed and realized, such as photonic bandgap materials [3-4] and metamaterials [5-6]. These revolutionary new concepts, in turn, have helped advance research into artificial optical
nanostructures, much of which has been undertaken with the purpose of providing an unprecedented degree of control over light propagation and light–matter interactions.

All optical nanostructures fall into one of two categories depending on the size of their features. Nanostructures in the first category have dimensions that are comparable to the wavelength of light such that diffraction is dominant and plays an important role in interacting with light. When the structure’s dielectric function has a periodic variation, photonic bandgaps, analogous to their electronic counterparts in solid state physics, are formed by Bragg diffraction [3-4]. All the structures are finite in size and possess interfaces to their surroundings so that it is possible to control the coupling of light into the modes within the structure. However, as the size of the components becomes smaller in comparison to the wavelength, the diffraction effect becomes weaker. The second category of nanostructures encompasses possesses dimensions that are much smaller than the wavelength of light, such that all the macroscopic optical responses of the nanostructures can be described based on the relative fractions of their constituents’ optical properties. Optical meta-atoms with prescribed electric and magnetic properties can be designed and arranged in a desired manner to create new artificial materials [6-8]. Nature provides only a limited set of materials on which it is possible to build novel optical materials or structures with customized electromagnetic and magnetic properties by engineering their electromagnetic scattering properties when illuminated by an incoming light wave.

Despite the diverse choice of optical nanostructures, building user-specified optical materials is not always straightforward. This is because it is difficult to balance multiple parameters or inputs such as electric and magnetic properties. These inputs are often interrelated; therefore, it is challenging to establish the optimal dimensions—those capable of satisfying all the user-defined requirements simultaneously—for an optical nanostructure during the design process. Most of the previously reported devices have relied solely on intuition-based design methodologies, and as a consequence they suffered from limited design flexibility and poor
performance. In customizing optical nanostructures, the proper optimization of parameters, such as geometries and dimensions, is critical.

In this research, we aim to overcome these central shortcomings of conventional design methodologies. Based on the optical nanostructures described, we propose a design, fabrication, and verification approach to creating various optical components that will be defined by their user-defined optical properties, such as scattering coefficients and effective medium parameters via robust genetic algorithm (GA) optimization [9]. Coupled with a full-wave electromagnetic solver, this heuristic searching algorithm can optimize optical nanostructures for user-specified optical functionalities. It is important to note that in order to reduce the structural constraints imposed on GA optimization we developed new fabrication processes and improved the current optical nanostructures, thus increasing the number of nanostructures appropriate for optical use. In addition, to ensure accurate synthesis we measured the optical properties of the constituent material candidates and incorporated them into the full-wave electromagnetic simulations conducted within the GA.

The optical nanostructures used here are broadly twofold. Firstly, to realize better-performing optical filters in the IR range, we studied all-dielectric planar structures because these are superior to metallic counterparts at higher frequencies. Secondly, we explored the concept of metamaterials to produce a variety of materials and devices in the IR and optical range such as absorbers, filters, and zero index materials. It is expected that these design methodologies will enrich optical filter applications and allow the exploration of optical functions with new optical properties. In the following subsections, these two categories of diffractive optical nanostructures and metamaterials are discussed with a brief background including their basic underlying physics. The limitations and challenges of these two areas are discussed as well, and then the goals of this research are defined.
1.1.1 Diffractive Optical Nanostructures

Over millions of years of evolution, biological organisms such as insects, fish, and birds developed diverse and complex multidimensional nanostructures that give rise to an assortment of striking photonic properties, including wavelength-specific reflection and transmission. However, it was not until the publication of two milestone papers on photonic crystals by Eli Yablonovitch and Sajeev John in 1987 that scientists started actively studying the underlying physics and applying the concept for building artificial optical crystals [3-4]. A photonic crystal is an artificially structured material with a periodic variation of dielectric function (Figure 1-1). This periodic variation in dielectric function results in photonic bandgaps that prevent the propagation of certain wavelengths of light in certain directions. Such phenomenon is commonly compared to the electronic bandgaps found in crystalline solids in which electrons are prevented from achieving certain energy levels due to periodic variations of the atomic electric potential over the lattice points.

![Example diagrams of simplified one-, two-, and three-dimensional photonic crystals.](image)

**Figure 1-1.** Example diagrams of simplified one-, two-, and three-dimensional photonic crystals. Two different colors represent materials with different optical constants. (a) One-dimensional, (b) Two-dimensional, and (c) Three-dimensional photonic crystals [10].
1.1.1.1 One-dimensional Structures

Before the publication of the two seminal papers, only low-dimensional photonic crystals, referred to as multi-layer dielectric stacks (i.e., Bragg mirrors [11]), had been widely studied in applications of highly reflective optical coatings and resonant cavities [12]. Nevertheless, the formation of one-dimensional photonic bandgaps from these crystals composed of layers of alternating high-dielectric and low-dielectric material is relevant to extending our understanding of two- and three-dimensional structures. Given its small dielectric constant variation ($\Delta \varepsilon$), a multilayer film with a periodicity of $d$ is depicted in Figure 1-2(b). When approaching zero, $\Delta \varepsilon$ is considered to be a uniform medium, leading the light wave to propagate at a speed of $c/\sqrt{\varepsilon}$ without any interruption. A slight difference in dielectric constants is sufficient to open the gap at the edge of the Brillouin zone (Figure 1-2(a)). The modes at $k=\pi/d$ are standing waves with a wavelength of $2d$. To illustrate why there is a frequency difference between these two modes, the electric field profile can be plotted in two ways without breaking the symmetry as overlaid in Figure 1-2(b). According to the electromagnetic variational theorem, the energy of the low-frequency mode is more concentrated in the high-$\varepsilon$ region, whereas the energy in the high-frequency mode is more concentrated in low-$\varepsilon$ region [10]. As a result, the mode below the gap has most of its power in $\varepsilon_2$, and the mode above the band gap has most of its power in $\varepsilon_1$. This difference in where the respective power of the two modes is concentrated raises the frequency a bit. In this simple example, there is only one reciprocal vector component to consider, but it is easy to think of similar situations in which two or three reciprocal vector components are involved in two-dimensional or three-dimensional photonic crystals.
Figure 1-2. Schematic illustration of bandgap formation in a one-dimensional photonic crystal. (a) Band diagram (b) Electric fields at the top of the air band and the bottom of the dielectric band are overlaid by multiple layers; the dark-blue areas have a higher dielectric constant than do the light-blue areas [10].

1.1.1.2 Two-dimensional and Three-dimensional Structures

To date, most demonstrations of photonic crystals have been conducted at relatively long wavelengths due to fabrication difficulties. The very first demonstration of a three-dimensional photonic crystal was conducted by Yablonovitch, who showed that an array of drilled holes forms an inverse diamond structure [13-14]. As there is no natural length scale in Maxwell’s equations, this result triggered attempts to use optical wavelengths to construct complete photonic bandgap structures in which inserting a defect would allow light to be localized at a single point. Efforts are still underway to construct various three-dimensional photonic crystals using state-of-the-art techniques such as layer-by-layer electron beam lithography, interference lithography, and self-assembly of microspheres in colloids.
Compared to their three-dimensional counterparts, two-dimensional photonic crystals can easily be fabricated by borrowing the conventional techniques used in the semiconductor industry. With less complexity in fabrication, two-dimensional photonic crystals still have many promising applications, —some of which have already been commercialized. One good example is photonic crystal fibers [15]. Also known as holey fibers, these have better properties than do optical fibers. They also have interesting photonic band gap effects and are currently a standard technique for supercontinuum generation [16]. Thin finite versions of two-dimensional photonic crystals, referred to as photonic crystal slabs, are also expected to be used in many areas. Since the structure of these crystals is comprised of a thin slab of semiconductor material with periodically arranged etched holes [17], it is feasible to integrate them into computer chips, thereby diversifying optical functionalities and improving optical data processing. This is allowed by total internal reflection, which guides light into the slab with diverse photonic crystal effects.

1.1.1.3 Photonic Crystal Slabs

In addition to the in-plane propagation properties of photonic crystal slabs, it is possible to use such devices to filter out-of-plane incident light, similar to the functionality previously demonstrated by one-dimensional resonant gratings [18-19]. This resonant phenomenon can be understood as a result of coupling between an incidence light and the guided modes of the slab determined by its periodic nature. When the following equation holds and thus satisfies the phase-matching condition, light can couple into the guided modes:

$$|k_0\sin \theta| = |k_{\text{para}} + \Lambda|$$  \hspace{1cm} (1-1)$$

where \(k_0\) is the wavevector of the incident light, \(\theta\) is the angle of incidence, \(k_{\text{para}}\) is the in-plane wavevector, and \(\Lambda\) is the reciprocal lattice vector. The quality factor (Q-factor) of this type of
resonance is usually very high; therefore, this resonance can be used as both narrow band and angular filters [20].

A simplified yet intuitive ray diagram describing this resonant coupling is shown in Figure 1-3. In order to reduce complexity, the structure considered here has only one-dimensional periodicity. When such a structure is illuminated by an incident light wave, some portion of the light is directly transmitted through the slab and the remaining portion is diffracted by the periodic structure. The diffracted portion is then guided into the structure to be rediffracted into the surroundings until it decays completely. The interference between the main transmission and the multiple diffracted rays at a given wavelength and incidence angle can be destructive due to phase mismatching, which leads to a guided mode resonance wherein no light is transmitted [19]. This process can be also explained using a band diagram from the photonic crystal and waveguide mode point of view (Figure 1-3).

Figure 1-3. Illustration of guided mode resonance using diffracted ray tracing [19].
In Figure 1-4, the band structure of a photonic crystal slab with infinitesimal one-dimensional line slots is plotted with the first TE band. It is nothing but the dispersion diagram of a blank dielectric slab waveguide. The only difference originating from the periodic nature is that all the dispersion curves can be folded into the first Brillouin zone by adding reciprocal lattice vectors, which allow modes to exist above the light line (i.e., leaky modes). Even with a normally incident light beam, the structure is accessible to the modes where the in-plane wavevector is zero (green circle). As long as the mode symmetry matches the polarization of the incident beam, resonant coupling can occur; and this coupling leads to an enhanced reflection peak and a reduced transmission dip in the measured spectra, which resembles a Fano resonance [21-26].

Figure 1-4. Band diagram (dispersion diagram) of a one-dimensional photonic crystal slab waveguide with infinitesimally small line slots, with the first TE band plotted.
1.1.1.4 All-dielectric Filters and Mirrors

Based on the underlying physics described in the previous section and the available nanofabrication techniques, various all-dielectric optical nanostructures can be realized. Among the possible optical functionalities, filters and mirrors may be among the most essential optical components. Similar optical filter or mirror functions can be achieved using different optical structures, provided that the working principle of the chosen optical structure is accessible to the range of targeted values. Various all-dielectric optical filter structures are available, each of which has pros and cons for specific applications.

One of the most popular all-dielectric filter structures is the multi-layer stack, which is shown in Figure 1-1(a). This structure has been widely utilized as a high-reflection coating by alternating quarter-wavelength-thick, high- and low-refractive index layers. Bandwidths can be adjusted by controlling the index contrast and slightly modifying the thicknesses to suit many applications [27]. Nonetheless, this structural configuration does have limitations. The number of alternating layers cannot be less than a certain number without compromising performance; thus the structure and hence the device to which it contributes can become quite thick. Finding two materials between which there is a high-index contrast that is not associated with noticeable absorption loss in the wavelengths of interest can also be a difficult task. The difficulty of distinguishing between two different polarizations of light is another possible challenge.

A less bulky but effective filter structure used in optical wavelengths is the one-dimensional resonant grating [28]. When the coupling between an incident light and a guided mode is controlled, a sharp resonance can be induced and used as a filter. This conventional one-dimensional structure, depicted in Figure 1-5(a), has a different filter response for each of two distinct polarizations of the incident field due to a lack of rotational symmetry. This filter response (i.e., guided mode resonance) is extremely sensitive to the angle of incidence, limiting
the applications to a collimated beam. Despite the weaknesses of one-dimensional gratings, efforts have been made to improve their performance by modulating periodicity and adjusting index contrast. With asymmetric or symmetric grating profiles, various designs for filters, high reflectors, polarization-insensitive devices, and polarizers have been proposed, opening the door to the possibility that all filter responses can be enhanced with another degree of freedom in design by incorporating rotational invariance in two dimensions [29].

Figure 1-5. Diagrams of (a) a 1D grating, and (b) a 2D grating.

By simply adding one more periodicity (Figure 1-5(b)), the limitations of one-dimensional gratings can be overcome without sacrificing their merits. Two-dimensional gratings were first theoretically investigated and their properties demonstrated by S. Peng in 1996 [30]. Since then, a number of studies have used different terminology, such as photonic crystal slab filters to refer to two-dimensional gratings [31]. Although previous filter functions and performances have been limited by simple unit cell designs and fabrication inaccuracies, two-dimensional resonant gratings have some clear advantages over one-dimensional counterparts and multi-layer stacks alike. That is, two-dimensional resonant gratings have polarization-insensitive responses and can be structured with only one or two layers for operation.
Considering their compactness and polarization insensitivity, two-dimensional resonant grating structures in this research are investigated in order to construct more effective optical filters. The researcher aims to overcome the current limitations of two-dimensional resonant grating structures pertaining specifically to filter functions and to general performance, including angular sensitivity. It is expected that performance can be optimized by optimizing the screen geometry that deviates from simple features such as air holes. Further, it is expected that through such optimization two-dimensional resonant gratings can become more competitive with a strong possibility of outperforming known functions or providing unexplored, useful responses.

1.1.2 Metamaterials

In 1968, Veselago published a paper that considered the theoretical case of a negative refractive index medium with several interesting phenomena [5], including the modified Snell’s law of refraction; however, this fictitious material did not receive further attention until Pendry delivered the phenomenally creative concept that a slab of negative index material could achieve perfect imaging of a source [6]. This influential theoretical account intrigued researchers from various disciplines, such that the field of metamaterials grew rapidly from that point on. The recent advent of transformation optics has stimulated even greater interest in metamaterials by offering the possibility of controlling the trajectory of light by spatially varying the index of refraction [32-33]. Various index values became necessary for specific applications with the control of both permittivity and permeability.
1.1.2.1 Artificial Electric Materials

Long before the recent proliferation of metamaterial research, artificial electric materials, referred to as artificial dielectrics, had already been widely investigated. In these studies, artificial dielectrics were developed at microwave frequencies in order to achieve desired permittivity values by mixing metals with dielectrics [34-35]. One of the most popular structures in artificial electric materials is the rodded medium in which either dielectric or metallic rods are arranged within a host medium in an orderly manner, resulting in an averaged dielectric constant of two different materials according to the respective filling fraction of each [36-38]. Shedding a new light on this idea, Pendry invented a similar structure [8], which led to the birth of a new field of study (i.e., metamaterials). In Figure 1-6(a), an example of the rodded medium is illustrated. In this case, for simplicity, a one-dimensional array of metallic wires is considered. The effective permittivity of this medium can be written using a Drude form with the damping ignored:

$$\varepsilon_{\text{eff}} = 1 - \frac{\omega_{p,\text{eff}}^2}{\omega(\omega + i\Gamma_{\text{eff}})} \approx 1 - \frac{\omega_{p,\text{eff}}^2}{\omega^2}$$

(1-2)

where $\omega_{p,\text{eff}}$ and $\Gamma_{\text{eff}}$ represent the effective plasma frequency and damping frequency of the rodded medium, respectively. Since the electrons are bounded only in the regions of wires in this composite medium, the effective electron density needs to be modified in the following form by taking the volume average:

$$N_{\text{eff}} = N \frac{\pi r^2}{a}$$

(1-3)

The effective mass of the electrons also needs to be reconsidered due to the self-inductance of the metallic wires. After a careful calculation as in [8] using the vector potential and momentum of electrons, the effective mass of the electrons in the wire medium can be obtained:
By replacing the new effective density and effective mass of electrons in the plasma frequency, \( \omega_{p,\text{eff}} \) can be expressed as

\[
\omega_{p,\text{eff}}^2 = \frac{\pi r^2}{a^2} \frac{m}{m_{\text{eff}}} \omega_p^2 = \frac{N_{\text{eff}} e^2}{\varepsilon_0 m_{\text{eff}}} = \frac{2\pi c_0^2}{a^2 \ln (a / r)}
\]  

(1-5)

As can be seen in the above equation, averaging the electrons over the volume of the medium lowers the overall density, and confining electrons to the thin wires enhances their mass. This results in a reduced plasma frequency, which allows the effective permittivity to change according to the demands at a certain frequency. In the region below the plasma frequency, the effective permittivity is negative, and in the region above the plasma frequency, the effective permittivity behaves as a transparent dielectric.

Other approaches to modifying the dielectric constant involve a periodically layered composite medium with two different constituent materials and randomly mixed metal–dielectric composites [39-43]. In particular, the layered medium has been widely studied for hyperlens applications [39-41]. When two different materials are layered alternately with specific thicknesses corresponding to the filling fraction depicted in Figure 1-6(b), the effective permittivity can be determined by applying a proper boundary condition according to the direction of the polarization of the electric field. Alternatively, the following expressions are also acquired by modifying the shape effect in Bruggeman’s effective medium theory [44-45]:

\[
\varepsilon_1 = \sum_i f_i \varepsilon_i, \quad \frac{1}{\varepsilon_\perp} = \sum_i \frac{f_i}{\varepsilon_i}
\]  

(1-6)
1.1.2.2 Artificial Magnetic Materials

In contrast to the negative values of electric permittivity readily available in noble metals at optical wavelengths, naturally occurring magnetic materials lose their properties rapidly as frequency increases even in the GHz range, such that accessing negative permeability at higher frequencies becomes more challenging [46]. As there is no atom capable of strongly interacting with the incident magnetic field at higher frequencies, this task can be solved by creating sub-wavelength artificial meta-atoms that are designed to couple with the magnetic field. This idea was first proposed by Pendry, who theorized that an array of split-ring resonators, as shown in Figure 1-7(a), would give an effective permeability through the magnetic resonance from each magnetic meta-atom (i.e., split-ring resonator) [7].
To induce the resonance in the purely inductive and non-resonant metallic rings, capacitive gaps are introduced. If the lumped element approximation can be applied using the equivalent circuit model, the resonance frequency range of each split ring resonator can be roughly estimated in the series RLC circuit (Figure 1-7(b)). The effective permeability is then acquired by calculating the magnetic momentum from the induced current [7]. By scaling down the structure, this geometry can be utilized at higher frequencies. However, due to fabrication difficulties, only planar structures have been investigated, which has limited the coupling efficiency when the structure is parallel to the incident magnetic field [47-48]. To make matters worse, the self-inductance of the electrons cannot be neglected at higher frequencies due to the finite electron density of the metals, which causes the resonance frequency to saturate [49].

The most widely used structure to achieve a magnetic resonance in the optical range is the paired nanostrip [50-53], which is illustrated in Figure 1–8(a). When the strip is aligned along the magnetic field, the resonance can be directly induced from the incident light with high coupling efficiency; thus, the limitations of the planar split ring resonator can be overcome. Combined with the non-resonant strips in the opposite direction of the magnetic field, the crossed
strips, known as a fishnet (Figure 1-8(b)), can control both the permittivity and permeability simultaneously, which allows the realization of negative index materials at optical wavelengths [54-56]. Since its introduction, this fishnet structure has served extensively as a basic geometry in synthesizing various kinds of optical metamaterials. Recently, a three-dimensional negative index metamaterial and a loss-compensated metamaterial have been demonstrated based on this geometry [57-58].

![Figure 1-8](image)

**Figure 1-8.** (a) Resonant magnetic nanostrips. (b) Fishnet geometry, combination of resonant strips and non-resonant strips.

### 1.1.2.3 Refractive Index Engineering

With the ability of controlling effective permittivity and permeability as explained in the previous subsections, various refractive-index values can be explored. This category of artificially engineered materials is not limited to negative values of the refractive index which have received tremendous attention since Pendry proposed the possibility of a perfect lens [6]. Compared with negative index metamaterials (NIMs), zero or low-index metamaterials (ZIMs or LIMs) have
received less interest in the literature. By engineering the electric and magnetic coupling through adjusting dimensions and geometries, values of refractive index between zero and one can also be achieved. In Figure 1-9, the field simulations of an infinite cylindrical line source embedded inside a slab and a square block of ZIM are shown. In this figure, the emerging electromagnetic waves from a ZIM (n~0) are collimated and parallel to the surfaces—and this configuration has recently become a subject of research interest aimed at increasing the directivity of an antenna embedded in such a medium [59-60].

![Figure 1-9](image)

**Figure 1-9.** Finite-element-method (FEM) 2D field simulation of an embedded current line source inside a slab of ZIM.

More complex index engineering became necessary when transformation optics was first analytically investigated in two seminal papers [33, 61]. Although the feasibility of various transformation devices such as cloaks and lenses has been demonstrated in other studies [61-63], a rigorous construction still requires exact spatial control of refractive indices. Furthermore, the
tool set of metamaterials needs to be completed with continuous values from negative, through zero, to positive in order to satisfy emerging transformation optics applications.

In addition to the fact that the less-studied zero- and low-index values require more investigation, current state-of-the-art metamaterials have several limitations. Thus far, most of refractive-index-engineered metamaterials suffer from a severe reflection loss caused by impedance mismatching to the surrounding medium (e.g., air in most cases). Only the product of permittivity and permeability ($\sqrt{\varepsilon\mu}$, index of refraction) has been explored, whereas the ratio ($\sqrt{\mu/\varepsilon}$, impedance) has been overlooked despite its importance in transmissive applications.

From the fabrication point of view, another source of imperfections, originating from the presence of a supportive substrate and the tapered sidewall angles in the layers, has introduced unwanted magneto-electric coupling in most previously studied structures. And, this unwanted coupling leads to broken-symmetry-induced bianisotropy in which electric permittivity and magnetic permeability are coupled with each other to describe the effective medium properties [64-65]. To make matters worse, this in turn degrades the transmission amplitude and increases the reflection loss caused by the deteriorated impedance mismatching.

Due to the difficulty of balancing the permittivity and permeability, engineering the values of the refractive index has been restricted within a narrow frequency band, limiting the operation of metamaterials in the broad range of frequencies. Extending the concept of index engineering to include dispersion engineering is essential for developing broadband applications. It is also expected that new types of devices or materials can be synthesized by controlling the dispersion curves of the effective medium parameters.

In order to overcome the stated drawbacks of current technologies and diversify the applications of metamaterials, in this research, we first explore the ZIMs and lessen or even obviate the structural limitations of the metamaterials by developing improved fabrication methods. Secondly, by engineering the dispersion curves of effective medium properties, we
investigate the possibility that metamaterials can be used in broadband filtering applications. Finally, for more practical device applications, metamaterial absorber structures are studied. The proposed metamaterial devices, together with the general synthesis approach, are expected to pave the way towards diversifying and improving metamaterials and consequently the uses to which they can be put.

1.2 Technical Approach

Synthesizing user-specified optical functionalities often necessitates fulfilling rather strict requirements. Some portion of these requirements can be readily satisfied by optimizing the geometries and dimensions of the optical nanostructures. However, a major bottleneck might exist in the fabrication constraints that limits the number of possible geometries of the optical nanostructures. When the design candidates are exempt from structural limitations, the possibility of achieving a better-performing design may increase. In the course of optimizing the optical nanostructures, theoretical calculations are also important to evaluate the design candidates and anticipate the scattering properties of the optimized structures. However, there should be a prerequisite to placing trust in theoretical expectations: the measured optical properties of the constituent materials must be incorporated into the calculation, otherwise the fabricated sample with the final design may not meet the user-specified criteria.

To overcome the stated limitations and enrich the optimization process of the optical nanostructures, in this research, we propose the following technical approach depicted in the flow diagram in Figure 1-10(a). First, the user-defined goals are set and the proper optical structure determined; this ensures that the targeted values are achievable based on the working principles of the chosen structure. The current limitations of the structure are then investigated and improved in order to enhance the optical properties of the optimized design when it reaches the
final stage. Since the optical properties of materials affect the coverage of optical responses from the designed structure, the appropriate choice of constituent materials should be made at this stage as well. The chosen constituent materials are then analyzed to obtain the optical properties for theoretical calculations. With these moderated fabrication constraints and measured optical properties of the constituent materials in place, the optimization process begins the search for the best-performing design.

Figure 1-10. (a) Process flow diagram of a GA synthesis. (b) Example of a unit cell of a periodic optical nanostructure.

The optimization algorithm used throughout this research is a GA, which is a heuristic search process inspired by the Darwinian theory of natural selection [9]. To optimize multiple parameters simultaneously, this algorithm relies on inheritance, mutation, selection, and crossover, mimicking the evolution process in nature. When a GA is coupled with a full-wave electromagnetic solver, the geometries and dimensions of the optical structures can be optimized for user-specified criteria. The process flow of a GA is described in the dashed blue square in Figure 1-10(a). To find an optimum solution to meet the user’s requests, the cost function is
defined to properly account for all the specific customized requirements. The searching process starts by randomly generating an initial population that includes a certain number of design members encoded in binary strings containing the structural information.

In Figure 1-10(b), a design example is shown for a dielectric unit cell subdivided into an 8×8 array of pixels. Each pixel is encoded as a “1” or “0” for the presence or absence of dielectric material. All other parameters including pitch size and the thickness of each layer are also encoded in the binary string to represent each member. The scattering coefficients of each population are evaluated using the measured optical properties of the constituent materials by an electromagnetic solver and compared to a user-defined cost function. Through this process of choosing the best-performing designs along with mutation and crossover, the next generation is produced for evaluation. This simple loop is repeated until the cost is minimized within a predetermined tolerance, thus achieving the desired electromagnetic properties. To avoid unrealistic designs in terms of fabrication, fabrication constraints are also enforced during the GA synthesis process.

The GA synthesized design is carefully fabricated following the pre-developed fabrication processes to satisfy the optimized dimensions. The optical properties of the fabricated nanostructure are then measured for characterization. In most cases, current existing measurement techniques are eligible to access the optical properties of the design. However, depending on the required measurement information, improvement of current techniques or the development of a new technique may be necessary to fully characterize the fabricated sample. Finally, the measured properties are compared to the theoretical predictions in order to ensure good agreement and confirm the performance of the synthesized design.
1.3 Overview

This dissertation introduces novel approaches for synthesizing a variety of all-dielectric filters and metamaterials for IR and optical wavelengths. This section provides an overview of the concepts covered in each of the following chapters along with a brief summary.

In Chapter 2, an all-dielectric grating structure was investigated for realizing a compact IR filter with a narrow bandwidth. To alleviate the polarization and angular sensitivity, a GA was employed to optimize a user-defined filter function based on a two-dimensional grating structure. The designed structure, composed of an array of subwavelength doubly periodic a-Si blocks on top of a polyimide membrane, has a polarization and angle tolerant response with a stop band centered at 3.0 µm. The optimized structure was carefully fabricated to reproduce the designed geometry and dimensions, and its polarization and angle tolerant filtering performance was verified by measuring its transmission and reflection using a Fourier transform infrared (FTIR) spectrometer.

Inspired by the work described in Chapter 2, a highly-efficient mirror function in the mid-IR range was explored in Chapter 3 by applying the effective medium theory to the two-dimensional gratings. From the scattering coefficients of a uniform slab of material, the proper effective medium conditions, giving rise to high reflection, were derived and incorporated into a GA coupled with a full-wave Maxwell solver to optimize the geometry and dimensions of the filter. The inverted effective medium parameters from the scattering coefficients of each design candidate were used for evaluation against high reflection mirror conditions (i.e., lossy ZIM). The optimized mirror design was fabricated in a small size sample (4 mm²) to confirm its reflection efficiency, and then a large scale sample (1 in.²) was produced with high throughput electron-beam lithography, demonstrating the possibility for its use in more practical coating applications.
Elevation and azimuth angle dependence were also investigated to be used with an unpolarized light beam with a non-negligible divergence.

Chapter 4 discusses the design, fabrication, and characterization of a free-standing optical ZIM that is symmetric in the direction of wave propagation. To overcome the limitations such as substrate and sidewall angle induced bianisotropy in the previously reported optical metamaterials, fabrication processes were optimized to produce a better performing metamaterial structure. With the aid of a GA, the structure was designed to meet the user-specified design criteria of near-zero refractive index with low absorption loss and a good impedance match to free space. The metamaterial consists of a gold-polyimide-gold tri-layer stack perforated by air holes in a fishnet geometry, resulting in a self-supportive and flexible metamaterial. The complex scattering coefficients of the fabricated ZIM was retrieved using spectral holography and inverted into effective medium parameters for verification.

Using this synthesis method, in Chapter 5, a free-standing broadband dispersion engineered flat-top band-pass optical metamaterial filter is presented, exhibiting a negative-zero-positive index behavior and reduced group delay over the 3.0μm ~ 3.5μm transmission band. Unlike previously reported narrow band index engineering, dispersions over the wide range of wavelengths were tailored for both the permittivity and permeability to control the scattering parameters. This was enabled by introducing structural perturbation in the conventional fishnet structure. Two additional square notches on each corner of the air holes induced two clear magnetic resonances at the desired wavelengths, thereby realizing the targeted filter function. The filter dimensions and geometry were optimized using a GA, and the filter was fabricated for characterization. Broadband filtering efficiency of the fabricated metamaterial was verified by measuring its transmission and reflection using FTIR.

By controlling dimensions and geometry of the subwavelength resonators, metamaterial concept can be further applied to artificial absorbing nanostructures. Loss is an undesirable factor
for transmissive applications, but it is useful when both reflection and transmission are required to be suppressed. Chapter 6 demonstrates a conformal metamaterial absorber with a narrow band, polarization-independent absorptivity centered at mid-IR wavelengths of 3.3 µm and 3.9 µm. The highly efficient nanoresonators were identified by using a genetic algorithm, exhibiting an effective electric and magnetic response that maximizes absorption in each wavelength band. The structure was patterned on a flexible Kapton and Au thin film substrate and characterized by collecting angle-resolved reflection using FTIR.

To summarize this work and propose possible directions of future research, in Chapter 7, the accomplishments and contributions in each chapter were listed to point out what has been done and what needs to be done. For realization of more sophisticated and better performing optical nanostructures, based on the guided mode resonance and dispersion engineering, required improvements and possible routes for synthesis were suggested for future work.
1.4 References


Chapter 2  

Planar All-dielectric Filter

This chapter presents the design and fabrication of a free-standing planar all-dielectric filter with a single transmission stop band centered at a mid-infrared wavelength of 3.0 μm. Using a robust genetic algorithm design method, polarization independent and incident angle tolerant filtering performance was investigated. The experimentally measured responses of the fabricated filter were verified by comparing with the simulated values. This was enabled through the collaboration of theoretical modeling, including electromagnetic full-wave simulations and GA synthesis, performed by Dr. Jeremy A. Bossard under the advisement of Dr. Douglas H. Werner.

2.1 Motivation

Periodic subwavelength nanostructures have been exploited to create new infrared (IR) and optical devices, materials, and coatings [1-12]. Their unique optical properties arise from the complex and often non-intuitive interaction of light with periodic arrays of subwavelength metal, semiconductor, and/or dielectric nanostructures. Previous device designs were based on the refractive index contrast [1], geometrical resonances [2], and guided resonances [3-6] produced by the nanostructure arrays to tailor the electromagnetic properties of the structure. A variety of IR filters having many different wavelength, polarization, and angle dependent responses have been proposed or demonstrated using two- and three-dimensional (2D and 3D) metallo-dielectric [7-8] and planar all-dielectric gratings [13], also referred to as photonic crystal slabs [5-6, 9-11].

The intrinsic loss of metals at IR and optical wavelengths limits the minimum bandwidth and absorption losses that can be realized for metallo-dielectric devices. All-dielectric structures can be used to overcome these limitations [14]. However, the majority of the dielectric filters that
have been reported are one-dimensional gratings with strong sensitivity to polarization and incidence angle [3]. More recently, 2D dielectric filters that have a polarization independent response have been fabricated by etching air holes in dielectric slabs [6] or by defining isolated amorphous silicon (a-Si) features on freestanding polyimide thin films [13]. The sharp guided resonances, which gave the narrow transmission or reflection bands, showed a strong divergence in transverse electric and magnetic (TE and TM) polarizations at oblique incidence angles.

In order to overcome the angular sensitivity of the guided mode resonance, a proper choice of geometries, dimensions, and constituent materials is required to control the coupling of light into the subwavelength nanostructure such that the user-defined requirements on properties such as the stop band position and the bandwidth are met. These competing requirements, on the other hand, often depend on one another, and thus must be balanced to achieve high performance optical devices. This is difficult to achieve using intuition-based design methodologies.

To overcome these limitations, in this research, a robust global optimization strategy was used to design an all-dielectric filter having a single transmission stop band centered at a mid-IR wavelength of 3.0 μm with transmittance less than $-25$ dB, a 10 dB bandwidth of 3.3%, and a high angular stability of up to 10º from normal for both polarizations. The significantly improved angular performance was achieved using a GA [15], which incorporated fabrication design rules, to optimize the filter geometry. The experimentally measured response of the fabricated filter was in strong agreement with that determined by full-wave electromagnetic simulation. In the following sections, the design, fabrication, and characterization of this planar all-dielectric filter are presented.
2.2 Electromagnetic Design Optimization

The planar all-dielectric filter consists of an inhomogeneous layer of doubly-periodic amorphous silicon ($a$-Si) nanostructures that are surrounded by air and supported by an ultra-thin polyimide substrate. The $a$-Si provides a large refractive index contrast to air and polyimide, whereas the polyimide film offers good mechanical stability and flexibility. For GA design optimization, the unit cell of the inhomogeneous top layer was divided into a $7 \times 7$ grid of pixels representing the blocks of air (‘0’) or $a$-Si (‘1’) as in Figure 2-1(a). The unit cell was constrained to have eight-fold symmetry to alleviate a polarization dependent response at normal incidence. It is expected that the TE and TM responses will diverge as the angle of incidence $\theta_i$ is increased from normal (Figure 2-5). Thus, the optimization was performed at the maximum oblique angle of incidence $\theta_m$, where a transmittance of less than $-10$ dB at the target stop band wavelength for both polarizations was desired over the range $0^\circ \leq \theta_i \leq \theta_m$. In other studies, it was found that this optimization technique is effective for $\theta_m$ up to $\sim 15^\circ$, beyond which several angles of incidence must be included to maintain high angular stability.

![Figure 2-1. Schematic illustration of the optimized unit cell](image)

(a) Pixelized unit cell assigned with binary numbers (b) Perspective view of the unit cell with dimensions.
The GA was used to optimize the all-dielectric unit cell to meet the filter specifications. For each candidate design, the scattering parameters were calculated by a full-wave periodic finite element-boundary integral (PFEBI) technique [16] and then evaluated against an ideal filter response to determine its cost given by,

\[
Cost = \frac{1}{N_{\text{pass}}} \sum_{N_{\text{pass}}} \left( \min \left( \left| T_{\text{TE,TM}} \right| - 0.95 \right) - 0.05 \right)^2 + \frac{2}{N_{\text{stop}}} \sum_{N_{\text{stop}}} \left( \max \left( \left| T_{\text{TE,TM}} \right| - 0.05 \right) - 0.05 \right)^2
\] (2-1)

where \(N_{\text{pass,stop}}\) are the number of pass and stop band frequencies and \(T_{\text{TE,TM}}\) are the TE and TM transmission coefficients. An ideal filter that satisfies this cost function would have a transmittance less than \(-25\) dB in the stop bands and greater than \(-0.45\) dB in the pass bands for both polarizations.

The first generation population members were created randomly and evaluated for their cost. The next generation was determined by mating the best performing candidates in the first generation and performing a mutation operator on a small percentage of offspring. The GA continued until it converged on an optimal solution that minimized the cost function. Importantly, nanofabrication design constraints were incorporated into the cost function to ensure that the optimized all-dielectric filter could be manufactured without modifying the nanostructure geometry. The measured optical constants of the a-Si and polyimide thin films were also included to properly account for material dispersion in the design.

The following filter properties were used as the input to the GA: (1) a single, polarization insensitive stop band condition (<\(-25\) dB) with high angular tolerance up to \(\theta_{\text{m}}=10^\circ\) was enforced from 2.97 to 3.03 \(\mu\)m, and (2) a high transmittance condition (>\(-0.45\) dB) was enforced from 2.50 to 2.83 \(\mu\)m and from 3.19 to 3.75 \(\mu\)m with a coarse wavelength sampling. The unit cell dimensions were limited to approximately half of the target wavelength to eliminate higher order propagating waves. The pixel size was constrained to be greater than 100 nm, and diagonal
connections between adjacent pixels were forbidden. The polyimide substrate thickness was allowed to vary from 0.5 to 1.0 µm and the a-Si from 0.2 to 0.5 µm. With these constraints, the GA evolved a planar all-dielectric filter that converged to satisfy the specified design criteria in a 16 member population over 33 generations. Figure 2-1(b) is an illustration of one unit cell of the optimized filter. In the 2D periodic array, the arrangement of pixels within the unit cell (1.72×1.72 µm²) form isolated a-Si features with a minimum air spacing between adjacent features equal to one pixel (246×246 nm²). The optimized thicknesses of the a-Si features and the polyimide substrate were 338 and 762 nm, respectively.

The simulated response of the optimized all-dielectric filter is plotted in Figure 2-2 for the TE and TM polarizations at normal and 10° off normal incidence and with the azimuth angle fixed at 0°. At normal incidence, a primary optimized transmission stop band with a transmittance of −25 dB and a 10 dB bandwidth of 3.3% occurs at the 3.0 µm target wavelength for both polarizations. For comparison, the 10 dB bandwidths of mid-IR metallo-dielectric stop band filters have been limited to 8~10% due to high metallic loss [8]. Although this filter was only optimized for minimum transmission, for reference, the peak reflection at the 3.0 µm wavelength was calculated to be −0.51 dB with an 11.0% 3 dB bandwidth. High performance all-dielectric mirrors could be designed by simultaneously maximizing the peak reflectance and minimizing the absorption. At 10° incidence, the TE center wavelength of the filter shifts slightly to 3.02 µm with a transmittance still less than −15 dB at 3.0 µm. The TM stop band does not shift between normal incidence and 10°. Further numerical analysis indicates that this filter maintains high angular stability for θm up to 20°. An additional narrow stop band centered at 2.61 µm was found between two adjacent wavelengths sampled during design optimization. In contrast to the primary stop band, at oblique incidence, the second band breaks into two weaker resonances that are different for TE and TM polarizations.
2.3 Principles of Operation

The planar all-dielectric filter properties can be understood by considering the electric field distribution of the mode at 3.0 μm determined by full-wave modeling. Figure 2-3(a) and 3(b) plot the electric field magnitude along the center of the a-Si features for both in-plane (x-y plane) and cross-section (x-z plane) views using a normally incident wave. The field is strongly confined within the high index a-Si and is asymmetric in the x and y directions, which is indicative of a guided resonant mode that couples to a normally incident wave [5]. At oblique incidence, additional weak guided resonances appear that do not couple to the normally incident wave.
because their field distributions are symmetric in both directions of periodicity. Resonances having extremely narrow bandwidth (<1%) can be achieved in dielectric gratings operating at a single incidence angle [4]. These results show that the GA can be used to optimize the tradeoff between bandwidth and angular tolerance by evolving structures that maintain strong field confinement over a wider range of angles.

**Figure 2-3.** Simulated electric field distribution (a) In-plane view (b) Cross-sectional view

### 2.4 Fabrication

The optimized design was fabricated to experimentally validate the filter performance following the procedure depicted in Figure 2-4. The polyimide substrate layer was deposited onto
an oxidized Si handle wafer by spin coating at 3300 rpm for 40 seconds and thermally curing the PI-2556 polyimide precursor. The polyimide precursor used in this experiment is HD Microsystems PI2556 diluted by 83% using HD Microsystems T9039 polyimide thinner. In order to imidize polyamic acid after the soft bake, the curing process was carried out in a nitrogen-purged oven by first heating at 150°C for 30 min, then at 250°C for two hours. On top of the polyimide film, an a-Si layer was deposited in a Trion ICP-PECVD system by introducing silane (SiH4) gas into the reaction chamber. During the deposition process, the substrate temperature was kept low at 300°C to prevent the polyimide film from glass transition. The thicknesses of the deposited films were measured by variable angle spectroscopic ellipsometry (J. A. Woollam BASE-160) and were confirmed to be within 5% of the designed values.

E-beam lithography was used for patterning the a-Si blocks. First a layer of positive E-beam resist (Nippon Zeon ZEP 7000A) was spun onto the wafer at 1000 rpm for 2 minutes. Following a soft bake at 180°C for 3 minutes, the resist was coated with 10 nm thick gold (Au) coating to prevent charging during E-beam exposure. The resist was then exposed in a Leica EBPG-5HR system to define the 2D square block array pattern in the resist. The dose of the E-beam current was adjusted to minimize blooming of pattern features due to proximity effects. Following exposure, the resist was developed in ZED 500 (Nippon Zeon) for 4 minutes. A 100 nm thick chromium (Cr) hard mask was deposited by evaporation and lifted-off in Microposit Remover 1165. The a-Si features were defined by reactive ion etching in a Trion Technologies Inductively Coupled Plasma system using chlorine (Cl₂) chemistry. The substrate bias power, coil power, and pressure were kept at 200 W, 50 W, and 2 mTorr, respectively, with the Cl₂ flow rate of 40 sccm. The hard mask was selectively etched using chromium Cr wet etchant (Transcene Cr etchant 1020) before the structure was removed from the Si handle wafer using a diluted buffered oxide etchant. The free-standing all-dielectric filter was mounted onto a testing frame for optical
characterization. The field emission scanning electron microscope (FESEM) image in Figure 2-5 shows an array of well-defined \( a \)-Si features with highly anisotropic sidewall profiles.

**Figure 2-4.** Fabrication process flow. (a) Deposit polyimide and \( a \)-Si layers (b) Spin E-beam resist with 10 nm Au coating (c) Expose and develop E-beam resist followed by Cr evaporation. (d) Lift-off process to define Cr mask (e) Reactive ion etching of \( a \)-Si to define features. (f) Remove the sample from the handle wafer by selectively etching the SiO\(_2\) underneath.
Figure 2-5. FESEM image of the fabricated planar all-dielectric filter structure. Inset: higher magnification image of the subwavelength $a$-Si nanostructures.

2.5 Characterization

Figure 2-6 shows the freestanding planar all-dielectric filter mounted on an aluminum holder for IR characterization. The filter transmittance was measured as a function of incidence angle using a Fourier transform IR spectrometer equipped with a liquid nitrogen cooled mercury cadmium telluride detector and an additional aperture in the beam path to reduce beam divergence. The transmission spectra, normalized to an air background, are plotted in Figure 2-7(a) to compare with theory.

The center wavelength of the primary optimized stop band for normal (TE and TM) and $10^\circ$ incidence (TM) was shifted from the design target of 3.00 $\mu$m to 2.93 $\mu$m, while still preserving the same Fano-shaped resonance, transmittance, bandwidth, and angular response.
Similarly, the 10° off-normal TE stop band found at 3.02 µm in the design was shifted by the same amount to 2.95 µm. A slightly larger discrepancy was noted for the unintended stop band at 2.61 µm, where the transmittance was increased from $-25$ to $-17$ dB for normal incidence (TE and TM). At 10°, the center wavelength of the TM resonances was maintained, but its transmission was less than $-4$ dB. The TE resonance splits into two stop bands at 2.61 µm and 2.66 µm, both with increased transmittance of $-9.5$ and $-14.5$ dB, respectively.

![Figure 2-6. Illustration of the angle-resolved transmission measurement setup for characterizing the mounted planar all-dielectric filter sample.](image)

The measured spectra confirm the strong agreement between the designed and fabricated planar all-dielectric filter properties with only a 2.3% difference in the measured center frequency of the primary stop band. The design geometry was adjusted to more precisely model the fabricated sample, and the modeled response is plotted in Figure 2-7(b). First, the polyimide substrate layer thickness was reduced to 680 nm. Second, 120 nm of polyimide was removed in the regions surrounding the $a$-Si features to account for overetching. With these adjustments, the
primary optimized stop band shifted to a center wavelength of 2.95 μm, which is within 1% of the measured value.

Figure 2-7. Transmission spectra for TE and TM polarizations at 0º and 10º. (a) Measured transmission spectra (b) Resimulated transmission spectra with modified dimensions of the fabricated planar all-dielectric filter.
2.6 Summary

A GA is employed to design a mid-IR planar all-dielectric filter composed of a doubly periodic array of \( a\)-Si nanostructures on an ultra-thin, freestanding polyimide substrate. The optimized design satisfied the user targeted mid-IR properties with a polarization insensitive stop band centered at 3.0 \( \mu \)m, a 10 dB bandwidth of 3.33\%, and an angular tolerance of up to 10\(^\circ\). The measured properties of the fabricated planar all-dielectric filter were within 2.3\% of the designed values, and improved to a less than 1\% difference when the actual fabricated layer thicknesses were included in the model. These results demonstrate that this integrated electromagnetic design optimization and nanofabrication approach provides a practical route to realizing dielectric filters with highly customized and diverse functions.
2.7 References

Chapter 3
All-dielectric Lossy ZIM Mirror

In this chapter, effective medium theory was applied to design a doubly periodic dielectric nanostructure grating that functions as a high-efficiency wavelength-selective mirror coating at the mid-infrared wavelength of 3.3μm. The geometry of the nanostructure grating was optimized using a genetic algorithm that was coupled with a full-wave electromagnetic solver to calculate the scattering parameters of the structure. By deriving the appropriate conditions that give rise to high reflection, the requirements of the effective medium parameters were properly described in the cost function used for the design optimization. The effective medium parameters were then retrieved from the scattering parameters using an inversion algorithm for evaluation. The design optimization was enabled through the collaboration of GA synthesis and electromagnetic simulations performed by Frank Namin under the guidance of Dr. Douglas H. Werner.

3.1 Background

There has been tremendous interest in designing periodic nanostructures to experimentally realize a wide variety of optical properties and device functionalities, including refractive index engineered coatings and materials [1-2]. In the infrared (IR) and the optical range, all-dielectric structures have been widely investigated due to their low absorption loss compared to their metallic counterparts, which is critical in high Q applications [3-10]. By exploiting
diffraction and interference phenomena, many useful all-dielectric photonic devices have been demonstrated, including as filters, polarizers, beam splitters, mirrors, and waveguides [6, 11-14].

Among the possible all-dielectric structures, planar structures such as one-dimensional gratings [13, 15-20] and photonic crystal slabs [3-5, 7-8], are attractive because of their compactness. A variety of advanced optical functions can be achieved by structuring one to two thin dielectric layers, which is not possible using conventional multilayer dielectric stacks. For example, sharp optical resonances can be induced by properly controlling the coupling between the incident light and the guided modes supported by the structure for various filtering applications [8]. Moreover, the periodicity can be extended to rotationally symmetric two-dimensional (2D) arrays of nanostructures to remove the polarization sensitivity of one-dimensional structures [3, 5-6, 15].

Traditionally periodic all-dielectric nanostructures have been analyzed by calculating the dispersion of the supported guided modes [7, 18, 20]. In these structures, the dispersion is strongly dependent on the specific properties of the nanostructure, including the component materials as well as the structure geometry and dimensions. For this reason, most previous device structures have been restricted to simple unit cell geometries, which limit the range of optical functions that can be accessed. Chapter 2 discussed using a genetic algorithm (GA) [21] optimization strategy to design a mid-IR stop-band filter with a measured response that met user-defined properties [6]. In this approach, multiple inputs can be fed into the design stage, and the optimum unit cell geometry is searched to minimize the cost function.

Optimizing the coupling between the incident light and the guided modes can be regarded as engineering the effective refractive index and impedance of the dielectric nanostructure. In particular, although the scattering parameters are governed by the coupling of light with a mode, the properties can also be modeled in terms of effective medium parameters for the case of
specular reflection and transmission. Thus, under these conditions, the nanostructure can be
considered as a uniform slab having specified effective medium parameters.

In this research, a nearly perfectly reflecting wavelength-specific mid-infrared (mid-IR)
mirror coating was designed, fabricated, and characterized. The geometry and dimensions of the
amorphous silicon (a-Si) subwavelength nanostructure coating was optimized by enforcing zero
index medium (ZIM) conditions. The optimized ZIM mirror had a single polarization-insensitive
reflection band at the mid-IR wavelength of 3.3μm with angular stability up to 15°, which was
enabled by the enforced lossy zero index medium (ZIM) conditions. The following sections of
this thesis chapter describe the design, fabrication, and characterization of this all-dielectric ZIM
mirror.

### 3.2 Design and Optimization

In order to design an all dielectric high reflection mirror in the mid-IR wavelength, a
guided mode resonance was exploited that allows exceptionally high reflection via a high Q
coupling between an incident wave and a leaky waveguide mode. This coupling can be expressed
with the effective index and effective impedance of a structured dielectric slab, provided that
there is no high order diffraction coupling from the periodic structure. High diffraction orders that
are not supported by a uniform slab of dielectric will be suppressed as long as the condition in Eq.
3-1 is met.

\[
\sqrt{\left( k_x + \frac{2\pi}{p}m \right)^2 + \left( k_y + \frac{2\pi}{p}n \right)^2} > k_0
\]  \hspace{1cm} (3-1)

where \(k_0\) is the wave vector of incident light, \(k_x\) and \(k_y\) are x and y components of \(k_0\), respectively,
\(p\) is the period of a 2D grating structure, and \(m\) and \(n\) are integer numbers. Assuming that the
index of refraction \((n)\) and normalized impedance \((Z)\) for a 2D grating structure with thickness \(l\)
are defined with suppressed high diffraction orders, the reflection coefficient can be written for normal incidence as given in Eq. 3-2.

\[ R = \frac{jZ \sin(n k \omega l) - j \sin(n k \omega l)/Z}{2 \cos(n k \omega l) + jZ \sin(n k \omega l) + j \sin(n k \omega l)/Z} \]  
(3-2)

To maximize reflection in Eq. 3-2, the numerator and the denominator need to be close in value to each other:

\[ \left| j \sin(n k \omega l) \left( Z - \frac{1}{Z} \right) \right| \approx \left| 2 \cos(n k \omega l) + j \sin(n k \omega l) \left( Z + \frac{1}{Z} \right) \right| \]  
(3-3)

There are two extreme ways of satisfying this requirement. The first case can be easily observed in most metals in the RF range (i.e. perfect electric conductor). When \( Z \) approaches zero, the \( 1/Z \) terms will dominate, leading to unity reflection. The second case occurs when \( 1/Z \) terms become negligible by an infinite value of \( Z \) (i.e., perfect magnetic conductor), which also satisfies unity reflection. For other cases, we now consider an index of refraction \( n = n' + jn'' \) with near-zero real part and large imaginary part (\( n' \approx 0 \) and \( n'' \ll 0 \)). Using the expansion formulas for sine and cosine functions with complex arguments, Eq.3-2 can be rewritten as in Eq. 3-4.

\[ R \approx \left| \frac{\sinh(n'' k \omega l) \left( Z - \frac{1}{Z} \right)}{2 \cosh(n'' k \omega l) + \sinh(n'' k \omega l) \left( Z + \frac{1}{Z} \right)} \right| \]  
(3-4)

For large negative arguments (\( x \ll 0 \)), \( \cosh x \) can be approximated to \( -\sinh x \), simplifying Eq. 3-4 to Eq. 3-5.

\[ |R| = \frac{Z + 1}{|Z - 1|} \]  
(3-5)

Considering complex impedance as in Eq. 3-6, impedance must be purely imaginary to achieve unity reflection under the above assumptions.
This is the case where index of refraction and impedance are purely imaginary (i.e., Re(n)→0, Im(n)→∞, Re(Z)→0). Such a lossy ZIM is here utilized to achieve a high reflection mirror.

The structure considered here is composed of a doubly-periodic amorphous silicon (a-Si) grating surrounded by air with a period small enough to suppress higher order diffraction modes. Silicon offers a comparably high index contrast to air, thus increasing the range of index modulation strength. For design optimization, the unit cell is divided into a 16×16 grid of pixels assigned with the value of either 1 (a-Si) or 0 (air) as shown in Figure 3-1(a). To alleviate polarization sensitivity, eight-fold rotational symmetry is enforced on the geometry. Along with the geometry of the unit cell, the unit cell size (periodicity), and thickness of the a-Si layer also have to be encoded into a binary string to completely describe the grating structure, and an initial population was randomly generated to begin the optimization process. The scattering parameters of each grating design is calculated using a periodic finite-element boundary-integral (PFEBI) code [21], and then its corresponding effective parameters (n, Z) are retrieved for evaluation using a well-established inversion algorithm [22]. The lossy ZIM conditions are described in the following cost function:

\[
\text{Cost} = |\text{Re}(n)| + |\text{Re}(Z)| - |\text{Im}(n)|
\]  

(3-7)

where both the real parts of the index of refraction and impedance are required to approach zero with a maximized imaginary part of the index of refraction when minimizing the cost. With the aid of a GA coupled with a PFEBI and an inversion algorithm, a best performing design is conversed upon after applying selection, crossover, and mutation over multiple generations. By evaluating the effective medium parameters of each design member against these lossy ZIM conditions, a
high-efficiency mirror can be optimized. In Figure 3-1, one unit cell of the GA optimized design is illustrated that has a high reflection band located at the mid-IR range of 3.3 \( \mu \text{m} \). The optimized doubly periodic structure has a unit cell size of 2.05 \( \mu \text{m} \) and is filled with 468 nm thick \( a \)-Si with patterned air holes.

\[ \text{(a) Pixelized unit cell assigned with binary numbers. (b) Perspective view of the unit cell.} \]

The dispersive effective refractive index and effective impedance of the optimized mirror structure are plotted in Figure 3-2(b) and (c). In the vicinity of the target wavelength of 3.3 \( \mu \text{m} \), there is an abrupt change in the real part of the refractive index with a huge dip in the imaginary part, in which a guided mode resonance occurs, whereas the impedance curves show comparably smooth transitions with the near-zero values in the real part. The optimized values at 3.3 \( \mu \text{m} \) are 0.072−5.410i and 0.001+0.794i for the effective refractive index and impedance, respectively, satisfying the desired lossy ZIM conditions.
Figure 3-2. Theoretical predictions of the optimized lossy ZIM mirror. (a) Simulated scattering parameters at normal incidence. (b) Retrieved complex effective refractive index values. (c) Retrieved complex effective impedance values.
The simulated scattering parameters of the optimized structure at normal incidence are shown in Figure 3-2(a). As expected from the effective medium parameters, near-unity reflection occurs at 3.3 µm with a comparably broad bandwidth. Although this mirror was only optimized for maximum reflection, owing to the negligible loss of a-Si in this wavelength range, the suppressed transmission at 3.3 µm is enough to be used as a stop band filter with an attenuation greater than 35 dB and a 6.6% 3 dB bandwidth.

3.3 Fabrication

The optimized mirror structure has isolated a-Si blocks located in the air holes, making the fabrication infeasible. To address this problem, we included a 500 µm thick fused silica substrate (Figure 3-1(b)) as a supportive layer that is transparent in this range of wavelengths with a reasonably small index contrast to air and a large index contrast to a-Si, which is critical requirement to support guided modes within the patterned a-Si layer. Fabrication of the optimized structure started with the deposition of a 468 nm thick a-Si layer on a cleaned fused silica substrate using a plasma enhanced chemical vapor deposition (PECVD) system (Applied Materials P-5000 cluster). To define doubly periodic a-Si geometry, electron-beam (E-beam) lithography (Leica EBPG-5HR) and anisotropic reactive ion etching (RIE) were utilized. After the thickness of the a-Si layer was confirmed with spectroscopic ellipsometry (J. A. Woollam BASE-160) to be within the tolerance of the designed value, a thin layer of E-beam resist (Nippon Zeon ZEP 520A) was applied by spin-casting and coated with a 10 nm Au layer by thermal evaporation to avoid charging effect during E-beam exposure. By carefully examining the dose arrays, a proper amount of dose was decided and used for large scale sample exposure. Following E-beam exposure and development of the resist, a 60 nm Cr layer was then evaporated and lifted off to form an etching mask. The pattern was transferred into the a-Si layer to define air
holes by highly anisotropic RIE (Tegal 6540) using chlorine (Cl$_2$) and argon (Ar) chemistry. Finally, the Cr mask layer was wet-etched and the samples were ready for characterization. The electron micrograph of the fabricated sample is shown in Figure 3-3 with highly anisotropic sidewalls.

For validating the performance of the optimized lossy ZIM mirror, two different sized samples were prepared. Firstly, a 4×4 mm$^2$ sample was fabricated using the above-mentioned process steps for characterization of the scattering parameters, and then another fabrication of a 1×1 in.$^2$ sample was conducted for uniformity mapping measurement in collaboration with Vistec Lithography Inc. using their high throughput E-beam lithography.

![Figure 3-3. FESEM image of the fabricated lossy ZIM mirror with an inset of magnified unit cell.](image)
3.4 Characterization

To account for the loading effect of the substrate, another theoretical calculation was conducted with the measured dispersive optical properties of fused silica in this range of wavelengths, which induced a slight reflection peak shift from 3.33 µm to 3.47 µm as shown in Figure 3-4. Despite this minor shift, the profile of the resonance was maintained, and the peak value was rather enhanced from 0.997 to 0.999 due to the fused silica substrate. The transmission amplitude was slightly increased from $2.71 \times 10^{-4}$ to $3.64 \times 10^{-4}$ but still negligible.

Figure 3-4. Simulated and measured transmission and reflection spectra of 4×4 mm² sample.
The mirror efficiency of the design was then validated by measuring the transmission and reflection of the fabricated 4×4 mm$^2$ mirror sample at normal incidence using a Fourier transform IR spectrometer (Bruker Optics IFS-66 main compartment) with a normal reflection setup composed of mirrors and a beam splitter depicted in Figure 3-5 that was built in-house. The measured transmission and reflection spectra were normalized to an air background and an Au mirror, respectively, and both were compared with the simulated scattering spectra in Figure 3-4. The center wavelength of the reflection resonance occurred at 3.48 µm with only 0.28% shift from the theoretical prediction, preserving the same Fano-shaped resonance with high reflection efficiency greater than 0.99. The position of transmission minimum was also shifted in the same manner with a reasonably low transmission value of 4.5×10$^{-3}$. The measured spectra confirm the strong agreement between the designed and fabricated lossy ZIM mirror except slight reflection decrease at band edges.

To demonstrate the possibility of this mirror for use in practical large area coating applications, we conducted uniformity measurement on the large size sample (1×1 in.$^2$). The

Figure 3-5. Illustration of the normal incidence reflection measurement setup for characterizing the mounted DFSS sample. M, mirror; L, lens; BS, beam splitter; A, absorbing block; S, sample.
sample area was divided into 9 sections as in Figure 3-6(a), and the transmission was collected from each section for amplitude mapping. In Figure 3-6(b), the measured transmission spectra were compared to each other along with the simulated transmission and reflection. Although the average resonance position was slightly shifted from 3.48 µm to 3.55 µm \( (i.e., \sim 2\%) \) due to the minor dimension difference from the small sample \( (4 \times 4 \text{ mm}^2) \), the resonance positions were maintained over the whole sample within 0.55\% from the center wavelength of 3.55 µm, thus confirming the uniformity of the fabricated sample. The measured transmission amplitudes at the resonance positions were suppressed to be in the range from \( 3.8 \times 10^{-3} \) to \( 4.8 \times 10^{-3} \) \( (i.e., -23 \sim -24 \text{ dB}) \), which in turn validates the high reflection efficiency at this wavelength owing to the negligible material absorption.

In addition to the uniformity measurement, the angular sensitivity of the fabricated mirror sample was tested. In most of practical filter and mirror applications, angular tolerance as well as polarization insensitivity is preferred for the operation with an unpolarized light beam with a non-negligible divergence. First, the azimuth angle dependence was investigated by measuring the transmission for a linearly-polarized normally-incident beam at three different polarization angles: \( \varphi = 0^{\circ}, 22^{\circ}, \text{and} 45^{\circ} \) (Figure 3-3). As expected from the eight-fold symmetry of the unit cell geometry, the spectral response of this mirror is almost independent to the polarization direction, which is shown in Figure 3-6(c). For these three polarization directions, the resonance positions remained the same at 3.55 µm with the average transmission amplitude of \( 4.8 \times 10^{-3} \) \( (i.e., -23 \text{ dB}) \). Secondly, the elevation angle dependence of the mirror was studied by measuring the transmissions for two distinct polarizations of TE and TM at various angles of incidence. The incident angles measured from normal ranged from \( 0^{\circ} \) to \( 20^{\circ} \) with a \( 5^{\circ} \) step, and the angle dependent transmission spectra are plotted in Figure 3-7(a) and (b) for TE and TM, respectively. Overall profiles around the resonance positions for both TE and TM polarizations share the same
asymmetric profile except the band edges in the TE spectra due to the additional coupling with other modes at oblique incidence angles. For tracking the amount of shift and amplitude change, the resonance position and transmission amplitude were plotted in Figure 3-7(c) with dual y-axes of wavelength and transmission versus the angle of incidence. The resonance position of the TE spectra was maintained up to 20° with less than 0.6% shift from the normal incidence, whereas the resonance position of the TM spectra gradually moved towards the shorter wavelengths after 10°. Nevertheless, the transmission amplitude of TM spectra stayed below $5.2 \times 10^{-3}$ ($i.e., -23\, \text{dB}$) up to 20°, validating the high reflection efficiency. The amplitude values of TE spectra are a bit higher than those of TM, but they are still below $2 \times 10^{-2}$ ($i.e., -17\, \text{dB}$), which gives a reasonable reflection efficiency of 0.98 when the absorption is ignored. Considering all the tendencies of the above-mentioned resonance behaviors, this mirror can be utilized up to 15° incidence without a noticeable degradation of reflection efficiency, and possibly can be used up to 20° when the requirements are not strict.
Figure 3-6. (a) Fabricated 1×1 in.² mirror sample with the description of divided sections for uniformity measurement. (b) Measured transmission spectra at each section. (c) Measured transmissions with different polarization directions: \( \varphi = 0^\circ, 22^\circ, \) and \( 45^\circ \)
Figure 3-7. Elevation angle dependent transmission measurement for (a) TE and (b) TM at various angles of incidence: 0° to 20° with a 5° step. (c) Resonance position shift and transmission amplitude change according to the angle of incidence.
3.5 Summary

Based on the effective medium approach, a high-efficiency mirror was successfully synthesized at the mid-IR wavelength of 3.3 µm. The mirror structure was optimized using a GA by evaluating the retrieved effective medium parameters from the scattering coefficients. It was demonstrated that the derived lossy ZIM conditions can be achieved to give high reflection originated from the guided mode resonance. The optimized structure was fabricated and characterized to show good agreement with the theoretical predictions. Angular tolerance, polarization insensitivity, and uniformity of the large scale sample were also demonstrated, thus verifying the possibility of this mirror to be used in more practical coating applications. It is expected that this design methodology further diversify the applications and functionalities of filters and mirrors based on the guided mode resonance.
3.6 References

Chapter 4

Zero Index Metamaterial

In this chapter, a free-standing optical zero-index metamaterial is presented that is symmetric in the direction of wave propagation. With the aid of a genetic algorithm, this structure is designed to meet the user-specified design criteria of near-zero refractive index with low absorption loss and a good impedance match to free space. The metamaterial consists of a gold-polyimide-gold tri-layer stack perforated by air holes in a fishnet geometry, resulting in a self-supportive and flexible metamaterial that serves as an important step towards the construction of 3D conformable transformation optical devices. This research was enabled through the collaboration with other research groups. Numerical simulations and GA synthesis were conducted by Zhi Hao Jiang under the advisement of Dr. Douglas H. Werner, and spectral holography measurements were performed by Dr. Qian Xu under the guidance of Dr. Zhiwen Liu.

4.1 Motivation

Introduced simultaneously by Pendry [1] and Leonhardt [2], transformation optics (sometimes referred to as transformation electromagnetic [3]) has revolutionized the way that physicists and engineers think about controlling electromagnetic waves. By choosing an appropriate spatial transformation and material index gradients, the behavior of light within a given region of space may be specified and controlled. A desired behavior might involve the exclusion of all light from a region of space, bending a beam without reflection, concentrating a
beam into a smaller region or splitting a single beam into multiple beams [3]. This technique requires point-by-point spatial control over both permittivity and permeability to achieve the desired electromagnetic wave trajectories. By incorporating metamaterial concepts into the transformation optics device, one can design an artificial material that satisfies the required 3D index profile.

However, constructing a designed transformation optics device using metamaterial building blocks is not as a rule straightforward due primarily to its overall geometrical complexity. As a consequence, only a few of the device demonstrations have been reported in the optical regime [4], with the majority of them being carried out at microwave frequencies [5-6]. Most conventional fabrication techniques for low frequency metamaterials involve the assembly of properly designed subwavelength units in a position-controlled manner to achieve the required spatially-dependent refractive index. As depicted in Figure 4-1, a stack of pre-defined metamaterial atomic layers with a desired refractive index variation within each layer as well as between layers can be applied even at optical wavelengths, provided that the spatially varying features of each functional layer are amenable to fabrication with currently available techniques and that the spacing and interaction between adjacent layers are properly taken into account.

New synthesis methods are required in order to diversify the possible applications of transformation optics and move beyond the current bottleneck, particularly with realizing optical devices. This includes developing design techniques that result in simplified material properties, constructing a library of metamaterial elements useful for refractive index engineering, and developing appropriate nanofabrication and assembly schemes. Among the refractive indices ranging from negative, through zero, to positive values higher than unity, the zero/low indices are probably the most useful ones as been employed in a large number of reported transformation optics devices such as the invisibility cloaks [4-8], optical illusion devices [9-10], flat Luneburg lenses [11-12], wave collimators [13-14], and so on.
The earliest investigations into the realization of zero/low index metamaterials (LIMs/ZIMs) used volumetric arrays of wires [15] or metallic meshes [16] to achieve a low effective permittivity \( (\varepsilon_{\text{eff}}) \) and a corresponding low effective index \( (n_{\text{eff}}) \). One can also reach the low \( n_{\text{eff}} \) using a magnetic resonance with the effective permeability \( (\mu_{\text{eff}}) \) approaching zero. However, neither approach is appropriate for transmissive applications due to the substantial reflection loss caused by an interfacial impedance mismatch. Other theoretical studies further suggested that these two cases can be combined to allow both the permittivity and permeability to approach zero at the same rate, thus providing a zero index and a matched impedance simultaneously [17-18] \( (i.e. n_{\text{eff}} = \sqrt{\varepsilon_{\text{eff}}\mu_{\text{eff}}} \approx 0 \) and \( Z_{\text{eff}}/Z_0 = \sqrt{\mu_{\text{eff}}/\varepsilon_{\text{eff}}} \approx 1 \), where \( Z_0 \) is the characteristic impedance of free-space). Such a ZIM requires balanced electric and magnetic responses within the wavelength band of interest, which in turn necessitates precise control over the subwavelength feature dimensions of the composite nanostructures, both in the design and fabrication aspects. Due to these difficulties, an impedance matched ZIM has not yet
been demonstrated, leaving an incomplete set of engineered refractive index values for optical transformation optics devices.

As a step towards completing the toolbox of metamaterials for transformation optics, in this research, a flexible free-standing low-loss polarization-insensitive optical ZIM with finely-tuned fishnet nanostructures was synthesized. The full set of scattering parameters were characterized using a spectral holography approach; the experimentally retrieved effective medium parameters of the fabricated ZIM were in strong agreement with theoretical predictions, simultaneously exhibiting a near-zero optical index and a matched impedance at 1.5 µm.

Importantly, unlike the current state-of-the-art optical refractive index engineered metamaterials, this ZIM does not suffer from the broken-symmetry induced bianisotropy introduced by the presence of the substrate [19] and the tapered sidewall angle in the fishnet’s features [20]. There are several compelling applications for such symmetric ZIMs, which include near-zero phase delay slabs and spatial filters. Moreover, this synthesis methodology can be applied, beyond merely a ZIM, to achieve low loss metamaterials with impedance matched to free space and index values anywhere between 0 and 1, thus providing the essential building blocks for constructing 3D conformal transformation optics devices, as well as 3D bulk-type flexible metamaterials. The following sections discuss the design, fabrication, and characterization of this ZIM.

**4.2 Electromagnetic Design Optimization**

In order to obtain a metamaterial design with an impedance-matched zero-index band that targets the telecommunication wavelength of 1.55 µm, a symmetric fishnet structure was employed consisting of two gold screens separated by a polyimide spacer with square air holes perforated through all three layers without a supporting substrate (Figure 4-5). The air holes were aligned in a doubly periodic arrangement with identical periodicity in both the x- and y-directions,
thus making the structure polarization insensitive to incoming radiation at normal incidence. With the combination of the resonant magnetic paired strips and non-resonant electric strips, the fishnet structure can be regarded as being comprised of an array of magnetic dipoles embedded inside a diluted metal, which yields a resonant $\mu_{\text{eff}}$ and Drude-like $\varepsilon_{\text{eff}}$. By carefully tailoring both the $\varepsilon_{\text{eff}}$ and $\mu_{\text{eff}}$ at the same wavelength, the desired impedance-matched ZIM can be obtained.

Due to the challenging design criteria for an impedance-matched ZIM, a powerful stochastic genetic algorithm (GA) [21] technique was employed to optimize the fishnet nanostructure to satisfy the performance goals defined above. The optimizer selected the geometric dimensions of the fishnet, including the unit cell size, air hole size, and layer thicknesses, in order to provide the best possible device performance. The measured frequency-dependent material properties and the applicable nano-fabrication constraints were incorporated into the optimizer to accurately model the material dispersion and to ensure that the resulting structure could be readily fabricated without further modification. For each candidate design in the GA optimization, the complex transmission and reflection coefficients were predicted using a full-wave periodic finite-element boundary-integral (PFEBI) solver [22]. The effective refractive index ($n_{\text{eff}}$) and normalized effective impedance ($Z_{\text{eff}}/Z_0$) were then inverted from the scattering parameters using a well-established retrieval method [23-25] and compared with an ideal ZIM response to determine the candidate’s cost defined by

$$\text{Cost} = |n_{\text{eff}} - n_{\text{tar}}|^2 + |Z_{\text{eff}}/Z_0 - Z_{\text{tar}}/Z_0|^2$$  \hspace{1cm} (4-1)$$

where $n_{\text{tar}}=0+0i$ is the desired effective index of refraction and $Z_{\text{tar}}/Z_0=1+0i$ is the target normalized effective impedance. The GA was able to minimize the cost and evolve a ZIM structure that meets all the design criteria specified in Eq. 4-1.
Figure 4-2. Optimized ZIM design. (a) Top view of fishnet geometry of the optimized structure. The red dotted square indicates the unit cell. (b) Diagram of a unit cell of the optimized ZIM design operating at 1.55 μm with the dimensions of w=365 nm, p=956 nm, and t=381 nm. The nanostructure consists of 39 nm gold (yellow) layers on top and bottom sandwiching a 303 nm polyimide (red) layer.

Figure 4-2 shows the geometry and dimensions of the optimized ZIM design. The width and length of the three-layered crossed strips in the unit cell are 365 nm and 956 nm, respectively. A 303 nm thick polyimide layer separates 39 nm thick top and bottom gold layers. The predicted transmission and reflection coefficients of this GA optimized design are shown in Figure 4-8(a) and (b). Because the effective impedance is well-matched to free space and the absorption loss in the metamaterial is low, the transmission amplitude remains remarkably high (~ 95%) around the target band at 1.55 μm. In addition, the transmission phase is in the vicinity of zero degrees and possesses a positive value (which is required, for example, in cloaking devices [2, 6, 18]), indicating a near-zero phase delay as an incident wave travels through the metamaterial. Considering the physical thickness of the structure, which is approximately λ/4, this near-zero absolute transmission phase provides confirmation that a near-zero length optical path can be achieved using the proposed metamaterial. The inverted effective material parameters are shown in Figure 4-9(a) and (b). A zero-index band with an extremely small extinction coefficient ($n_{eff} =$...
0.072+0.051i) that indicates low absorption losses in the structure is observable exactly at 1.55 μm following a negative index band at longer wavelengths. The near-unity normalized effective impedance with small imaginary part \( (Z_{\text{eff}}/Z_0=1.009-0.021i) \) substantiates the low reflection loss as exhibited in the amplitudes of the scattering parameters. To quantitatively evaluate the ZIM performance, two Figure-Of-Merits (FOMs) are defined specifically for the zero-index band as

\[
FOM_n = 1/|n_{\text{eff}}|, \quad FOM_z = 1/|Z_{\text{eff}} - 1| \tag{4-2}
\]

which evaluates how closely the effective refractive index approaches zero and how closely the effective impedance approaches unity. For this specific optimized design, the achieved FOMs have high values of 11.4 and 44.7, respectively, representing a new threshold in the state-of-the-art for ZIMs.

### 4.3 Principles of Operation

To acquire a clear understanding of the optical response of this ZIM, the current distributions on the top and bottom gold layers at 1.55 μm are plotted in Figure 4-3(a) and (b) with an incident electric field linearly polarized along the x-direction. An array of long gold metal strips, which is aligned to the x-axis along the incident electric field, provides a diluted Drude-type response [26] (Figure 4-10(a)). The anti-parallel currents induced by the incident magnetic field along the y-axis produce a resonant magnetic response, resulting in a Lorentzian line-shaped resonance in the effective permeability [27] (Figure 4-10(b)). By carefully tailoring both the \( \varepsilon_{\text{eff}} \) and \( \mu_{\text{eff}} \), they can be tuned to approach a near-zero value with \( \varepsilon_{\text{eff}} = 0.068+0.051i \) and \( \mu_{\text{eff}} = 0.072+0.049i \) at the same wavelength, leading to the proposed impedance-matched ZIM (Figure 4-9(a) and (b)). In such a way, a near-zero phase delay can be achieved which is clearly shown by
a snapshot of the field evolution in Figure 4-3(c). The electric fields on both top and bottom interfaces of the nanostructure, as well as inside the metamaterial, are almost identical in terms of both amplitude and phase.

**Figure 4-3.** Numerical simulations of current distribution on the Au screens at ZIM band. (a) Distribution of electric surface current (a) on the top Au layer and (b) the bottom Au layer. These anti-parallel currents induced by an incident magnetic field account for the resonant magnetic response in the ZIM. (c) Cross-section view of the snapshot of the evolving E-field. Almost identical field vectors before and after the medium are found to indicate near-zero phase delay with high transmission. The polarization of a normally incident beam is also indicated.
4.4 Fabrication

The designed ZIM structure is self-supportive with a perfectly vertical sidewall angle. To make this challenging fabrication possible and further applicable for constructing 3D transformation optics devices, the development of a compatible process is required. Unlike other previously reported fishnet-based optical metamaterials that were fabricated by a series of evaporations of metal and dielectric onto the electron-beam or interference lithography defined resist patterns, the following top-down route was chosen to improve the sidewall angles and overcome the well-known limitations of the lift-off process (i.e., thickness limit and inherent angle (~10°)).

The fabrication procedure began by first evaporating a 39 nm thick bottom gold layer onto a thermally oxidized silicon wafer, which served as a sacrificial handle substrate. The intermediate dielectric layer was deposited by spin-casting a polyimide precursor (HD Microsystem PI2556 resin diluted by 50% using HD Microsystem T9039 polyimide thinner) at 2400 rpm for 40 seconds. To form an imidized layer, the as-spun polyimide layer was cured by first heating at 150°C for 30 minutes and then at 250°C for 1 hour in a nitrogen-purged convection oven. The top gold layer was then evaporated to be 39 nm thick, which was identical to the bottom layer. Square air holes in the 3-layer stack were defined by reactive ion etching using the patterned silicon dioxide (SiO₂) masking layer. This was accomplished by first depositing a 200 nm thick SiO₂ layer on top of the 3-layer stack using an Applied Materials P-5000 Plasma Enhanced Chemical Vapor Deposition cluster system. The temperature was kept low at 150°C during the deposition to prevent thermal damage to the layers. A thin layer of positive electron-beam resist (ZEP 520A, by ZEON) was applied with a spin speed of 5000 rpm for 1 minute, followed by a soft bake at 180°C for 3 minutes. The exposure dose during electron-beam lithography (Leica EBPG-5HR) was adjusted to achieve a reasonable correspondence in
dimensions between the fabricated and designed unit cell. Following exposure, the resist was developed in n-Amyl Acetate solvent for 3 minutes to define the fishnet pattern. Reactive ion etching was then applied consecutively to the SiO$_2$ masking layer and 3-layer stack. Trifluoromethane (CF$_3$) 35 sccm and oxygen (O$_2$) 5 sccm were employed as etching gases for SiO$_2$ patterning in a Trion Technologies Inductively Coupled Plasma Etching system. The pressure, substrate bias power, and coil power were kept at 20 mTorr, 100 W, and 50 W, respectively. With the patterned SiO$_2$ mask, the top and bottom Au layers were etched with chlorine (Cl$_2$, 35 sccm) and argon (Ar, 5 sccm) chemistry in the same system. The pressure, substrate bias power, and coil power were maintained at 20 mTorr, 200 W, and 50 W, respectively. The polyimide spacer was etched using O$_2$ (90 sccm) in an Applied Materials Magnetically Enhanced Reactive Ion Etching system. The pressure, substrate power, and magnetic field were kept at 10 mTorr, 150 W, and 45 Gauss, respectively, with helium backside cooling. The process was completed by removing the remaining silicon dioxide masking layer using low-power fluorine-based plasma etching (Figure 4-4). The ZIM sample was removed from the handle substrate with diluted buffered oxide etch and mounted onto an aluminium metal frame for optical characterization. The active area of the ZIM is 3×3 mm$^2$, which is equivalent to 3100 × 3100 unit cells of the metamaterial (Figure 4-7(c)). High-magnification field emission scanning electron microscope (FE-SEM) images of the array show that the screen geometry was accurately reproduced during ZIM fabrication (Figure 4-5).

Importantly, this ZIM has an almost perfect sidewall profile (Figure 4-5) with nearly identical dimensions in the top and bottom layers (Figure 4-5), thus allowing the structure to be exempt from another source of bianisotropy caused by the noticeable sidewall angles which are present in most of the previously reported optical metamaterials [19-20]. Furthermore, only with these conditions satisfied can one use uncoupled $\varepsilon_{\text{eff}}$ and $\mu_{\text{eff}}$ to describe the effective medium properties. Due to the ductility and flexibility of its constituent materials, the fabricated structure
is pliant within limits, which is advantageous when considered in the context of a building block for conformal transformation optics devices (Figure 4-5).

**Figure 4-4.** Fabrication process flow. (a) Deposit Au, polyimide, Au and SiO$_2$ layers. (b) Spin E-beam resist. (c) Expose and develop E-beam resist followed by SiO2 reactive ion etching. (d) Reactive ion etching of multi-layer stack. (e) Reactive ion etching of the SiO2 mask layer. (f) Release the sample from the handle wafer by removing SiO$_2$ underneath.
Figure 4-5. FESEM images demonstrating the flexibility of the peeled-off ZIM sample. The insets show clean and vertical sidewalls in three layers.

As a comparison to the fishnet with a nearly-perfect sidewall angle shown in Figure 4-5, we also simulated a fishnet structure with the same dimension but having an 80° sidewall angle.
(which is a typical value found in most reported optical metamaterials [20, 28-31]. As shown in Figure 4-6, the transmission strength drops from ~95% to ~77% due to the deteriorated absorption loss and impedance matching. Also, the other drawback appears in the non-identical reflections that result when a normally incident wave illuminates from different sides (top and bottom) of the metamaterial. The non-zero difference (green curve) between these two reflection coefficients validates the existence of bianisotropy (i.e., magnetoelectric coupling), indicating that the transmitted wave has a polarization component orthogonal to that of the incident wave.

Figure 4-6. Simulation of fishnet with an imperfect sidewall angle. (a) The unit cell geometry of the fishnet with an imperfect sidewall angle. The dimensions are p=956 nm, w=365 nm, t=381 nm, and θ=80°. The structure consists of 39 nm gold (yellow) layers on top and bottom sandwiching a 303 nm polyimide (red) layer. (b) Simulated scattering parameter magnitudes. '1' denotes the top half space and '2' denotes the bottom half space. The ZIM band (around 1.54 μm) is indicated by the purple dashed line.
4.5 Characterization-Spectral Holography

To fully validate that the fabricated sample is a true ZIM, both the amplitude and phase information of the scattering parameters needs to be acquired for retrieval of the effective medium parameters. For this purpose, a spectral holography technique [32-33] was used to obtain its complex transmission and reflection coefficients. The schematic diagrams of the experimental setups for complex transmission and reflection coefficient measurements are shown in Figure 4-7(a) and (b). The transmission measurement was conducted using a Mach-Zehnder interferometer. As an input source, a white light supercontinuum, generated by coupling sub-nanosecond laser pulses into a nonlinear photonic crystal fiber, is sent to two arms of this interferometer. Interferograms with and without inserting the ZIM in the optical path of one arm are measured over a broad range of wavelengths (1.2-1.7 µm) by an optical spectrum analyzer. By taking the ratio of the reconstructed signal terms, the complex transmission $t$ can be calculated because it is the only different factor that changes when the ZIM is placed in the signal beam path.

The reflection coefficient was acquired by applying a Michelson interferometer. A mirror area used to produce a reference signal is fabricated beside the ZIM sample (Figure 4-7(c)) to assist in the measurement of the scattering coefficients. It has the same thickness as the ZIM sample thus allowing us to obtain the reflection interferograms from the mirror and sample surfaces, ideally at the same height level, by mechanically moving the entire aluminum frame in the transverse direction to the beam propagation by several millimeters. The reflection interferograms from the mirror and sample are collected, and then the complex reflection $r$ can be calculated using essentially the same method as employed for the transmission coefficient acquisition process.
Figure 4-7. Characterization of the fabricated ZIM sample. (a) Schematic of the Mach-Zehnder interferometer used for transmission measurement. (b) Schematic of the Michelson interferometer used for reflection measurement. M, mirror; L, lens; BS, beam splitter; OSA, optical spectrum analyzer. (c) Photograph of the peeled-off ZIM sample which is mounted on an aluminum frame for characterization. Mirror and sample areas are also indicated.

The detailed spectral holography procedure is as follows. A supercontinuum light source, which was fed into the interferometers for characterization, is generated by nonlinear processes that a pump pulse (JDS Uniphase NP-10620-100) experiences while traveling through a photonic crystal fiber (BlazePhotonics SC-5.0-1040) [34]. Transmission and reflection interferograms were
collected using an optical spectrum analyzer (Ando electric). The complex transmission \( (t = |t|e^{i\theta_t}) \) and reflection \( (r = |r|e^{i\theta_r}) \) coefficients of the fabricated ZIM sample were calculated from the measured data as follows. For the transmission measurement, the interferograms are given by

\[
E_T = |R + S|^2 = |R|^2 + |S|^2 + SR^* + RS^*
\]  \hspace{1cm} (4-3)

\[
E_T' = |R + St|^2 = |R|^2 + |St|^2 + StR^* + RS^*t^*
\]  \hspace{1cm} (4-4)

where \( E_T \) and \( E_T' \) are the transmission interferograms without and with the sample, respectively (Figure 4-11(a)). \( R \) represents the field amplitude of the reference beam in the frequency domain, while \( S \) stands for that of the signal beam. Figure 4-11(b) shows the inverse Fourier transforms (IFT) of these two interferograms. The center peak corresponds to the IFT of the first two terms on the right-hand sides of the Eq. 4-3 and Eq. 4-4 which are the individual spectra of the reference and the signal. The two sidebands are respective IFTs of the following two terms in Eq. 4-3 and Eq. 4-4. In these experiments, the signal beam path length was designed to be longer than that of the reference. As a result, the sidebands on the right represent the \( R^*S \) and \( R^*St \) terms. By taking the ratio of the Fourier transforms of these two sidebands, the complex transmission \( t \) can be obtained.

The measured reflection interferograms from the mirror and the sample are shown in Figure 4-12(a), and can be expressed as

\[
E_R = |R + Sm e^{i(\omega/c)DL}|^2 = |R|^2 + |Sm|^2 + Sme^{i(\omega/c)DL}R^* + RS^*m^*e^{-i(\omega/c)DL}
\]  \hspace{1cm} (4-5)
\[ E'_R = |R + Sr|^2 = |R|^2 + |Sr|^2 + SrR^* + RS^*r^* \] (4-6)

where \( E_R \) and \( E'_R \) are the reflection interferograms with the mirror and the sample, respectively, and \( m \) is the mirror reflection coefficient which is assumed to be unity. \( \Delta L \) is the beam path length difference between the two measurements due to necessary adjustment of the mirror position and orientation as aforementioned. The inverse Fourier transforms of the interferograms are shown in Figure 4-12(b). Following the same procedure as the transmission coefficient calculation, the complex reflection coefficient can be obtained. However, instead of the reflection coefficient \( r \), what we measured is \( re^{-(\omega/c)\Delta L} \). Note that the phase difference between transmission and reflection in the off-resonance region is \( \pi/2 \) under the assumption that it is lossless and possesses time reversal symmetry; \( \Delta L \) can be calculated and was found to be around 8 \( \mu \text{m} \).

The measured transmission and reflection coefficients are shown in Figure 4-8. Strong agreement was found between measurement and simulation with only a 0.05 \( \mu \text{m} \) wavelength shift (from 1.55 to 1.5 \( \mu \text{m} \)) in the resonance that is most likely caused by fabrication imperfections such as slight dimension mismatches. The measured transmission amplitude is around 95% near the ZIM band, thus confirming good impedance matching of the structure to free space, i.e. low reflection loss, as well as low absorption loss. Minor differences are observed in the reflection measurement, especially around the phase bump between 1.5 and 1.55 \( \mu \text{m} \). This effect is demonstrated because the reflected signal strength is close to the noise floor of the characterization system, and therefore this amplitude-sensitive behaviour cannot be clearly resolved by the detector. However, this difference does not affect the retrieval of the effective properties of the metamaterial as seen in Figure 4-9 and 10 where the effective refractive index, normalized impedance, permittivity, and permeability are displayed to show good agreement with the theoretical predictions. The ZIM band is slightly shifted to 1.5 \( \mu \text{m} \) with a complex index of
0.121+0.032i, exhibiting a high FOM of 8.08. The measured normalized effective impedance around this ZIM band has a value of 0.861−0.049i indicating a very close match to free space, which is only possible when both permittivity and permeability approach zero with a carefully chosen ratio as shown in Figure 4-10. In all, the measurements of the effective parameters not only qualitatively but also quantitatively agree well with the simulations, confirming an impedance-matched zero-index band around 1.5 μm, and thus validating the theoretical prediction of realizing near-zero optical path and depth using the proposed structure.
Figure 4-8. Simulated and measured scattering parameters. (a) Transmission and reflection amplitudes. (b) Transmission and reflection phases. Blue and red lines correspond to transmission and reflection, respectively.
Figure 4-9. Simulated and measured effective medium parameters. (a) Effective refractive index. (b) Normalized effective impedance. Blue and red lines correspond to real part and imaginary part, respectively.
Figure 4-10. Simulated and measured effective medium parameters. (a) Effective permittivity. (b) Effective permeability. Blue and red lines correspond to real part and imaginary part, respectively.
Figure 4-11. Transmission interferograms acquired using spectral holography. (a) Measured transmission interferograms. (b) Inverse Fourier transforms. Blue and red lines correspond to with and without placing the ZIM, respectively.
Figure 4-12. Reflection interferograms acquired using spectral holography. (a) Measured reflection interferograms. (b) Inverse Fourier transforms. Blue and red lines correspond to with and without placing the ZIM, respectively.
4.6 Summary

A polarization-insensitive impedance-matched free-standing conformal ZIM was demonstrated at optical wavelengths with near perfect transmission properties. The nanostructure, composed of a three layer metallo-dielectric fishnet with a vertical sidewall angle, was fabricated and released from the sacrificial wafer forming a truly symmetric metamaterial in the direction of wave propagation. A spectral holography approach was adopted to provide full characterization of this optical metamaterial. The strong agreement between measurement and theory verifies the low-loss near-zero index and highly transmissive nature of the device. This ZIM, together with the general synthesis approach introduced here, not only paves the way to novel substrate-free optical devices such as beam collimators and near-zero phase delay slabs but also represents a critically important step closer to the possibility of assembling 3D conformable transformation optics devices and substrate-free bulk-type optical metamaterials with such building blocks.
4.7 References

Chapter 5
Dispersion Engineered Metamaterial Filter

This chapter presents a free-standing broadband dispersion engineered flat-top band-pass optical metamaterial filter with a negative-zero-positive index behavior and reduced group delay over the 3.0 μm ~ 3.5 μm transmission band. Both the permittivity and permeability were tailored to control the scattering parameters by applying structural perturbations to the basic fishnet structure. The filter dimensions and geometry were optimized with the aid of a genetic algorithm (GA) coupled with a full-wave electromagnetic solver, and the performance of the fabricated metamaterial was verified by measuring its transmission and reflection using FTIR. This proof-of-concept metamaterial filter demonstrates the possibility of engineering the metamaterial dispersion in the optical range, which is essential for developing broadband applications. This research was enabled through the collaboration of GA synthesis and electromagnetic simulations performed by Zhi Hao Jiang under the guidance of Dr. Douglas H. Werner.

5.1 Motivation

The advent of metamaterials and their unique effective medium properties such as negative [1-2], zero/low [3], and high [4-5] indices of refraction has revolutionized the way scientists and engineers manipulate the propagation and radiation of electromagnetic waves. Over the last decade, this nascent field has witnessed rapid development with the emergence of a wide variety of novel electromagnetic devices, ranging from the early perfect lenses, which provide
sub-wavelength imaging resolution beyond the diffraction limitation [6-8], to invisibility cloaks, which hide objects from being detected [9-11], as well as other devices such as ultrathin perfect metamaterial absorbers [12], artificial mirrors [13], electromagnetic induced transparency [14-15], flat wave collimating lenses [16], and so on. The functionalities of metamaterials rely on their effective medium properties [17] arising from the sub-wavelength inclusions aligned in a periodic lattice. However, it is well-known that, due to the resonating nature of these inclusions, the responses of the metamaterials are inherently dispersive (i.e., frequency dependent refractive index and group delay), resulting in signal distortion and limited operational bandwidths that are inappropriate for broadband applications.

In recent years, efforts have been directed at overcoming these disadvantages. One possible approach is to design metamaterials at frequencies away from the resonance band in order to avoid strong dispersion. Such a method allows metamaterial devices to possess broader bandwidths and has been demonstrated by ground plane cloaks [18-19], Luneburg lens [20-21], broadband artificial mirrors [22], etc. Nonetheless, these particular devices made use of only positive refractive index values, thus forfeiting the other range of possible values, i.e. negative and zero indices which are also accessible by metamaterials.

An alternative method to avoiding the dispersive band, referred to as dispersion engineering [23-25], actually turns the shortcomings of dispersion into assets that can be used to realize new functionalities or improve existing devices’ performances by tailoring the metamaterial properties to specific device needs. Dispersion engineering has been applied to several planar guided-wave devices that have been demonstrated in the microwave range using transmission-line metamaterials with customized phase responses, including broadband power dividers [26], broadband rat-race couplers [27], impulse devices [28], and so on. The dispersion engineering approach has also been employed for radiated-wave applications such as a backfire-to-endfire leaky-wave antenna [29], and a soft-metahorn antenna [30].
In the optical range, such dispersion profile tailoring technique has been considered to achieve new types of planar metamaterials analogous to electromagnetic induced transparency [14-15], and slow and/or fast light devices [31]. These optical devices have very narrow bandwidths due to the sharp Fano resonances they utilize. Therefore, it would also be highly beneficial to bring the broadband dispersion engineering concept into the optical wavelength in order to provide more comprehensive control over the metamaterial properties and facilitate the design of broadband devices. Currently, due primarily to the limitations of available constitutive materials and fabrication techniques, broadband dispersion engineering, which tailors both the refractive index and group delay profiles over a wide wavelength band, is largely unexplored and offers an excellent avenue for diversifying optical functionalities.

In this research, broadband dispersion engineering was investigated to synthesize a new type of optical metamaterial. Particularly, an optically-thin flat-top band pass metamaterial filter was designed, with a negative-zero-positive index behavior and reduced group delay over the 3.0μm ~ 3.5 μm transmission band. Tailoring of both the permittivity and permeability profiles was enabled by carefully loading sub-wavelength nano-notches to the metal-dielectric-metal fishnet structure. Fabrication and measurement of the meta-filter confirms its broadband, low loss performance. This proof-of-concept example illustrates the possibilities of controlling and utilizing metamaterial dispersive properties to achieve broadband devices. In the following sections, the proposed concept of dispersion engineered filters, design procedure of this proof-of-concept device, and its fabrication and characterization are presented.
5.2 Electromagnetic Design Optimization

5.2.1 Dispersion Engineered Filter

Band pass filters are devices which allow signals to transmit through within a certain wavelength range and block the waves outside the transmission window (Figure 5-1(a)). Within the pass band, a flat-top transmission band with a constant group delay is desired to ensure the transmitted signal has minimal distortion in both frequency and time domains. The most conventional technique to construct optical band pass filters is stacking evaporated thin films onto substrates that are transparent at wavelengths of interest or combining multiple birefringence half-wave plates [32].

At the mid- and near-infrared (IR) wavelength range, however, these devices are too thick to be integrated into potential nano- or micro-scale systems and require a number of different kinds of materials or components. Other approaches such as patterned frequency selective surfaces on substrates and metal films [33] with periodic cross-shaped air-holes [34] have also been demonstrated in the far- and mid-IR wavelengths. These approaches lend themselves to optically thin structures and more design flexibility, but are limited in a narrow and sharp transmission window. Recently, several thin band pass filters have been reported in the terahertz range using the resonating properties of metallo-dielectric metamaterials or complementary metamaterials [35-37].

In order to verify the possibility and usefulness of dispersion engineering, a band pass filter in the optical wavelength is here proposed by tailoring the effective medium properties of metamaterials, which enables to control both refractive index and group delay simultaneously. From this perspective, the desired effective medium properties are displayed in Figure 5-1(b), showing a Drude-like permittivity and a permeability with two Lorentz-shaped resonances.
located on both sides of the plasma wavelength of the permittivity. Such a material has a gradually changing index \( n_{\text{eff}} = \sqrt{\varepsilon_{\text{eff}}\mu_{\text{eff}}} \) from negative to positive values. Between the negative unity index and positive unity index wavelengths, the permittivity and permeability are balanced such that the effective impedance is well matched to free space throughout this band. A flat-top pass band can thus be achieved. Outside of this band, the unbalanced permittivity and permeability produce a mismatched impedance that blocks the transmission of waves.

Importantly, in addition to adjusting the values of permittivity and permeability, their slopes must also be carefully controlled in order to compensate the dispersive index, therefore alleviating group delay fluctuations within the balanced band (i.e., the pass band). Considering a uniform slab made of such a postulated effective medium with a thickness of around 0.15\( \lambda \), the scattering parameter magnitudes and the group index can be calculated as plotted in Figure 5-1(c), showing a flattened pass band and a near-constant group delay between \( \lambda_n \) and \( \lambda_p \). In this theoretical calculation, loss is not included in the simulation. Nevertheless, loss is an important factor in the implementation of such an effective material using available materials and geometries.
Figure 5-1. Theoretical concept of band pass filters enabled by matching permittivity and permeability. (a) Ideal response of a band pass filter with a flat-top transmission window and a constant group delay within the desired window. (b) Real parts of the dispersive permittivity, permeability and refractive index profile of a hypothetical material. (c) The scattering parameter amplitudes (in dB scale) and group delay ($\tau_g$) of a slab (with a thickness of $0.15\lambda$) made of such a material.
Chapter 6
Summary and Future Work

6.1 Summary

The goal of this research was to realize user-specified optical functionalities based on the all-dielectric and metallo-dielectric nanostructures for IR and optical wavelengths. In order to attain the strict requirements, the proper optical nanostructure was first determined to ensure that the desired optical responses were achievable. With the chosen optical nanostructure, the structural limitations were identified and improved by developing compatible fabrication methods, which alleviated potential performance degradation caused by the structural imperfections. The constituent materials were carefully decided to maximize the device performance, and their optical constants were measured to be incorporated into a full-wave electromagnetic solver. With the aid of a GA, the interrelated specific requirements were satisfied by optimizing the dimensions and geometry of the optical nanostructure. The GA synthesized design was then fabricated and characterized to confirm that the desired goals were achieved. The optical structures exploited in this work were two-fold: planar all-dielectric nanostructures and metallo-dielectric metamaterials.

In chapters 2 and 3, planar all-dielectric nanostructures, capable of inducing guided mode resonances, were investigated to design compact mid-IR filters and mirrors. To overcome the intrinsic loss of metals at IR and optical wavelengths that limits the minimum bandwidth and absorption losses, all-dielectric nanostructures were used. Chapter 2 presented a mid-IR planar all-dielectric filter composed of a doubly periodic array of $a$-Si nanostructures on an ultra-thin, freestanding polyimide substrate. The GA optimized filter design satisfied the targeted mid-IR filtering performance with a polarization and angle tolerant stop band centered at $3.0 \, \mu\text{m}$ and a 10
dB bandwidth of 3.33%. The FTIR measured angular stability of the fabricated filter was maintained up to 10°, and the resonance position was within 2.3% of the designed values.

Based on the guided mode resonance, in chapter 3, a high-efficiency mirror was synthesized at the mid-IR wavelength of 3.3 µm using a doubly periodic array of a-Si nanostructures on a fused silica substrate. The effective medium approach was applied to derive high reflection conditions and incorporated into a GA coupled with a full-wave electromagnetic solver to evaluate the retrieved effective medium parameters using the calculated scattering coefficients. The derived lossy ZIM conditions led to high reflection originating from the guided mode resonance. The optimized lossy ZIM mirror structure was then fabricated and characterized using FTIR to show good agreement with the theoretical predictions. The measured mirror efficiency was maintained for both TE and TM polarizations up to 15° of incidence with 99% reflection and a shift of the resonance position less than 1% from the normal incidence. This new approach is expected to further diversify the applications and functionalities of filters and mirrors based on the guided mode resonance.

The concept of metamaterials was also explored in chapter 4, 5, and 6 to realize various IR and optical functionalities. First, a free-standing ZIM, designed at the telecommunication wavelength of 1.55 µm, was demonstrated in chapter 4. To eliminate the bianisotropy problems and substrate loading effects, compatible fabrication processes were developed to employ a better performing structure that is symmetric in the direction of wave propagation. The designed ZIM consisted of a gold-polyimide-gold 3-layer stack perforated by square air holes, resulting a self-supportive and flexible metamaterial. The effective medium properties of the fabricated ZIM were verified by spectral holography. The measured interferograms were used to calculate the scattering coefficients which in turn were inverted into the effective medium parameters for verification. The ZIM band was slightly shifted to 1.5 µm with a complex index of 0.121+0.032i and a normalized effective impedance of 0.861–0.049i indicating a very close match to free space
(i.e., 95% transmission). This ZIM, along with the developed fabrication methods, greatly expands the metamaterial playground, allowing the synthesis of various conformable metamaterials that can be used as the building block of 3D transformation optical devices as well as 3D flexible metamaterials.

Band-limited index engineering was further extended to dispersion engineering producing a broad band metamaterial filter in chapter 5. Over the wide range of wavelengths, the permittivity and permeability were tailored to shape a pass band filter function. Structural modification was made to the conventional fishnet geometry, allowing the control over the position and strength of the two magnetic resonances. The GA optimized metamaterial filter exhibited a flattened transmission band from 3.0 μm to 3.5 μm with a negative-zero-positive index behavior and reduced group delay of 12 fs. The filter structure was composed of a gold-polyimide-gold 3-layer stack perforated by notch-loaded square air holes. To confirm its filtering performance, the structure was fabricated and characterized using FTIR. The measured pass band was slightly widened to the range from 2.95 μm to 3.05 μm with an in-band insertion loss smaller than 1 dB and a transmission variation of 0.6 dB. The transmission in the stop bands was suppressed to be around −10 dB. This proof-of-concept metamaterial filter represents a big step towards dispersion engineering in the optical range that is promising to be used to realize highly customized and diverse metamaterial functionalities, let alone broadband applications.

In addition to the index and dispersion engineering, in chapter 6, electric and magnetic resonances were utilized to produce a polarization insensitive, dual-band metamaterial absorber in the mid-infrared at 3.3 μm and 3.9 μm. The GA identified H-shaped nanoresonator array induced polarization-insensitive high absorptivity greater than 90% over a ±50º incidence-angle range. The nanoresonator array was patterned on a Kapton layer backed by a thin gold film, thus resulting in a conformal and substrate-insensitive absorbing film. The fabricated metamaterial absorber was characterized by conducting angle-resolved reflection measurement at multiple
angles. The two absorption peaks remained above 90% for an incidence angle up to 50° for both polarizations, with the −10 dB bandwidth of both bands exhibiting a maximum 0.06 μm broadening compared to the simulated results. It was also verified by full-wave simulation that the absorption efficiency of this metamaterial absorber maintains when integrated onto curved surfaces of arbitrary materials, making it attractive for advanced multi-spectral coatings that suppress the reflection and transmission.

6.2 Recommendations for Future Work

6.2.1 Dispersion Engineered Guided Mode Resonances

Stemming from the work on the guide mode resonance (GMR) in this research, there are several interesting directions for future projects. One possible direction for GMR is to produce broad band filters and mirrors. Although the GMR has been widely used to produce an extremely narrow single stop band filter [1-2], it can also be exploited to be used in broad band applications [3]. A GMR structure is a slab waveguide that is eligible to hold many guided modes inside. Due to the presence of the periodicity, those modes can couple into surroundings to induce GMRs [2]. As expected from this origin, the position of a resonance relies on the dispersion of the guided mode. If many GMRs are gathered around the wavelength range of interest, a broad band filter can be realized [3]. The periodic geometry and dimensions of the structure can be optimized for tailoring dispersions of the modes [3]. Doubly periodic structures may have more flexibility than one-dimensional counterparts when the complexity in designing can be overcome by a GA.

Another possible application for the GMR is to control the group velocity of light [4]. Especially, slowing down light has many promising applications such as enhanced nonlinear
optical phenomena and optical delay lines [5]. The speed of the signal with finite time duration is determined by the group velocity, which can be engineered by the transmission phase dispersion. When properly designed, GMR elements are eligible to open a transmission window with the desired phase dispersion giving rise to a group delay [4]. The concept of dispersion engineering is not limited only to the control of light propagation. This can be further extended to produce various optical components by sculpting the amplitude as well as the phase in the broad range of wavelengths.

In order to diversify the applications, structural modifications can be made to the GMR structures. Adding more GMR layers may increase the number of possible functionalities through the control of coupling [6]. Each waveguide layer has its own guided modes, and these modes are coupled in between layers when the separation distance becomes small enough. In this case, the whole layers act as one unit having more complex mode profiles due to evanescent mode coupling, which may be applicable to various dispersions [6]. When the distance between GMR layers is far enough to ignore evanescent mode coupling, the cavity-like responses, originating primarily from the propagating waves, become dominant and can be used for tunable filters by varying the distance [7]. The compactness of GMR structures allows further stacking to produce more useful optical components [4].

### 6.2.2 Dispersion Engineered and Loss Compensated Metamaterials

Based on the various metamaterial structures synthesized in this work, several aspects need to be considered more in terms of metamaterial diversity and performance. As an extension of dispersion engineering conducted in chapter 5, various dispersion functions can be explored to diversify the applications of metamaterials. Recently reported electromagnetically induced transparency metamaterials fall in this category and promise to be useful for slow light
applications [8-9]. With the aid of a GA, the performance can be improved and optimized for bandwidth, group velocity, and loss. As long as the geometry and dimensions of the structure are feasible with the current state-of-the-art fabrication techniques, structural modification can be further pursued to have more control over the magnetic and electric resonances, producing more complex profiles in the dispersion curves. More sophisticated user-specified dispersion engineered metamaterials can be investigated.

For transmissive applications, loss is an important factor to be addressed. In this research, a GA was employed to minimize the reflection and absorption losses from the metamaterial structure by adjusting the geometry and dimensions. By removing the supporting substrate, a better impedance matching was achieved, suppressing the reflection loss. However, inherent metallic resistive loss becomes nonnegligible when many layers are stacked to form bulk-type metamaterials [10]. Recently, there have been attempts to compensate this loss by incorporating gain medium such as quantum dots [11], single quantum wells [12], and dyes [13]. Pumping this type of medium, embedded inside the metamaterial, with a bit higher energy, appears to be increased light intensity at the desired wavelength where the loss is severe, but it has not been demonstrated yet that the response is stable under steady-state conditions. Another possible approach to tackle this loss problem may be the synthesis of low loss materials [14]. There are a couple of things to be considered as an alternative for metals. To reach a negative permittivity, the carrier concentration must be high enough and controllable to access various values. The low mobility and interband transition cause undesirable losses that might limit the applications. Possible candidates to replace silver and gold in the visible range include transparent conducting oxides that can be highly doped to control the carrier concentration with high interband transition energy.

Another interesting direction for optical metamaterials is to realize tunable and reconfigurable structures, which extends the usefulness of metamaterials tremendously in more
practical applications such as filters and modulators. Dynamic tuning can be done in many ways, but it mainly relies on the changes of material properties. In THz range, a phase modulator and a frequency agile metamaterial were demonstrated by optical pumping to control the electron density in the semiconductors [15-17]. Electrical tuning was also used in the visible range to obtain refractive index changes in the accumulation layer of a gold-silicon dioxide-indium tin oxide stack under applied field [18]. Drastic changes of the refractive index are also available when phase change materials are incorporated into the metamaterial structures. For example, chalcogenide glasses [19] such as Germanium Antimony Tellurium [20], used in rewritable optical discs and phase-change memory applications, have a wide tuning range for optical properties by changing the crystalline phase from amorphous to crystalline [21].

In order to regard metamaterials as a material, the bulk-type properties are required. Most previously reported optical metamaterials, including work in this research, could be considered as metafilms or metasurfaces as opposed to materials due primarily to the difficulties of fabricating thick, three-dimensional structures. Recently, there have been a few demonstrations of three-dimensional metamaterials in the optical range [10, 22-24]. However, constructing isotropic three-dimensional structures still requires more precise and complex control over geometry and dimensions, which has only been theoretically realized so far [25-26]. Breakthrough fabrication methods or synthesis techniques need to be developed to solve this long-lasting problem along with a loss issue that becomes a more crucial factor in realizing thicker metamaterials.

6.3 References


### 6.3.1 Design of the Structure

For a proof-of-concept demonstration, a flat-top band pass metamaterial filter with a pass band between 3 µm and 3.5 µm within the 2.5 µm to 4 µm window was designed. The structure for this metamaterial consists of a tri-layer metal-dielectric-metal stack perforated with doubly periodic notch-loaded square air holes in all three layers was employed. The infinite metallic strip array along the incident electric field is primarily responsible for the effective permittivity profile, whereas the array of parallel metal slab pairs to the incident magnetic field mainly determines the effective permeability profile, as will be shown below. The added eight notches in the air holes provide additional design flexibility in tailoring both the effective permittivity and permeability profiles, which are also eligible to induce a higher order magnetic resonance without including any additional major structural modifications.

Achieving the aforementioned dispersive properties within a broad wavelength range, however, is still a sophisticated task, which requires fine adjustment of the geometric dimensions. To facilitate the design process, a genetic algorithm (GA) [38] optimization technique was employed here, coupled with a full-wave electromagnetic solver and an inversion algorithm to optimize the meta-filter structure for the desired effective medium parameter profiles, including the unit cell size, notch-loaded air hole size, and layer thicknesses. To ensure responses
Insensitive to the polarization of normally incident plane waves, an eight-fold rotational symmetry was enforced to the structure in the plane perpendicular to the incident wave vector. Importantly, the measured wavelength-dependent material properties and appropriate nanofabrication constraints were also incorporated into the optimization algorithm to simultaneously account for the constituent material dispersions and avoid unfeasible candidate structures [33].

For each candidate design in the GA optimization, the complex scattering parameters were calculated using Ansoft High Frequency Structure Simulator (HFSS) with periodic boundary conditions assigned on the lateral walls of the simulation domain to approximate a plane wave incident on the structure at normal incidence. The effective medium parameters ($\varepsilon_{\text{eff}}$, $\mu_{\text{eff}}$) within the target wavelength range were then retrieved from the simulated scattering parameters using an inversion algorithm [39] and compared with the target effective medium profile requirements to determine each candidate’s Cost defined by

$$\text{Cost} = \text{Cost}_1 + \text{Cost}_2$$

where $\varepsilon_{\text{tar},i}$ and $\mu_{\text{tar},i}$ are targeted permittivity and permeability values, respectively, and $\tau_{g,\text{mean}}$ is a desired group delay at each frequency point ‘$i$’. $\text{Cost}_1$ minimizes the difference between the targeted and optimized values for permittivity and permeability in the passband and increases the impedance mismatch at the stop band frequencies to ensure high reflection. $\text{Cost}_2$ minimizes the difference between the desired and optimized group delays. The GA evolved until it converged to a best performing design with a sufficiently low cost value.

The single unit cell geometry and dimensions of the GA evolved design are shown in Figure 6-2. Four square air holes, located at each corner of the unit cell with an edge length of
990 nm, are loaded with two orthogonal 198×198 nm² small square notches at the corner towards the center of the unit cell. The total thickness of the tri-layer stack is 510 nm (i.e., only about 1/6λ at 3 μm), thus promising to be easily integrated into nano- or micro-scale infrared/optical systems.

Figure 6-1. Diagrams of the optimized metamaterial band pass filter design. (a) Top view of the unit cell. (b) Perspective view of the unit geometry. The dimensions are p=2113 nm, w=1123 nm, g=198 nm, t=30 nm, and d=450 nm, respectively.

The simulated scattering profile of the designed metamaterial filter exhibit a pass band between 3 μm and 3.5 μm with a small insertion loss less than 1.1 dB (Figure 6-3(a), top). Notably, this pass band has a flattened top with a variation less than 0.4 dB within the 3 μm-3.5 μm window. This is important in maintaining the profile of transmitted light intensity in the frequency domain. Outside the pass band, the transmission is suppressed below −10 dB, indicating more than 90% of the incident light is blocked by the metamaterial slab. A sharp roll-off can be seen between the pass band and the stop band with a roll-off of ~93 dB/μm on the shorter wavelength side and a roll-off of ~101 dB/μm on the longer wavelength side. As theoretically expected, this band pass property originates from the effective medium properties of
the metamaterial (Figure 6-3(b), and Figure 6-4(a) and (b), top). Interestingly, the index goes from negative unity to positive unity across the pass band with a small imaginary part less than 0.15 (Figure 6-3(b), top), ensuring low absorption loss in this range.

The group delay of this metamaterial filter, which is inherently dispersive, can be calculated according to

$$
\tau_g = \frac{L}{v_g} = \frac{L(\text{Re}(n) + \omega \frac{d \text{Re}(n)}{d\omega})}{c_0}
$$

where $c_0$ is the speed of light in free space and $L$ is the total thickness of the metamaterial. Owing to the careful manipulation on the profile of the effective medium parameters throughout the pass band, in addition to the high transmission, the slope of the effective refractive index is well controlled such that the group delay within the pass band shows only a small variation (Figure 6-3(b), top). Between 3 µm and 3.5 µm, the group delay ranges from 15 fs to 27 fs (i.e., a variation range of 12 fs). Considering its broad bandwidth, this suppressed group delay variation is reasonably low compared to the previously reported metamaterials [40-41] in the optical range. Further suppression can be achieved, provided with more geometrical flexibility and more stringent requirements described in the cost function during the optimization procedure.
Figure 6-2. Simulation comparisons between optimized structure with (top) and without notches (bottom) illuminated by normally incident radiation. (a) Simulated scattering parameter amplitudes. (b) Retrieved effective index of refraction ($n_{eff}$).
Figure 6-3. Simulation comparisons between optimized structure with (top) and without notches (bottom) illuminated by normally incident radiation. (a) Retrieved effective permeability ($\mu_{\text{eff}}$). (b) Retrieved effective permittivity ($\varepsilon_{\text{eff}}$).
### 6.3.2 Tailoring Permittivity and Permeability Profiles

The formation of the gradual index transition and the low loss flat-top pass band can be better understood by examining the effective permittivity and permeability. The effective permittivity (Figure 6-4(b), top), with a Drude-like response, varies from negative unity to positive unity in the wavelength range of interest, with two little anti-resonances just on the edges of the pass band caused by the corresponding magnetic resonance modes. The effective permeability (Figure 6-4(a), top) shows two resonant modes; one main resonance at 3.7 µm and a weaker second one at 2.85 µm. Within the pass band, the permeability also changes from negative unity to positive unity with a sharper slope in the longer wavelength side. The main resonance has been widely exploited in the literature as a prerequisite for the formation of a negative index band. However, the second resonance mode occurring at a shorter wavelength has often been overlooked, because it usually has a weaker strength than the main mode and its excitation, sensitive to the geometry and dimensions, is not always guaranteed. In this design, the effective excitation of this second resonance arises from the structural perturbation, providing the possibility of shaping the dispersive profile of permeability in the shorter wavelength region.

Physically, as the current distributions on the top and bottom metal layers show in Figure 6-5(a) and (b), both of the two magnetic resonance modes are generally attributed to the anti-parallel currents. These currents are driven by the incident magnetic field, which further induce another magnetic field either enhancing or reducing the total magnetic field strength and result in Lorentz-shaped permeability resonances. For the main resonance at 3.7 µm, the currents are flowing primarily in the x direction, with a phase difference of 180° between the central area and the end areas in the x direction. This results in the thin rectangular charge accumulation/depletion regions near the boundary of the center square with different current phases. The magnetic field distributions showing a resonating dipole mode (Figure 6-5(c) and (d), left) are in close
correspondence with the current distributions. The fields are pointing primarily to the $y$ direction and exhibit a 180° phase difference in the central area compared to the other areas.

However, at 2.85 $\mu$m where the second magnetic resonance mode occurs, the mode pattern is found to be different from that of the main mode. The current distribution in the center square turns out to be more complicated, as the in-phase pattern is no longer maintained, while the outer regions exhibit no phase difference. Interestingly, the charge accumulating/depleting areas shrink down to two point-like small regions, further leading to a curl-like distribution of the magnetic fields in the corresponding areas (Figure 6-5(c) and (d), right). Accordingly, due to these changes in the current distribution, two quadrupole modes can be identified near the top and bottom edges of the central area in the magnetic field distribution. When compared to the main mode at 3.7 $\mu$m, these quadrupole modes are weaker in terms of the magnetic field intensity, thus resulting in a weaker resonance in the permeability with a lower quality factor as seen in Figure 6-4(a). It should be noted that these two magnetic resonance modes, especially the oft-overlooked second one, are critical to the realization of the proposed filter function since they give a chance for the permeability to catch up at a similar rate with the inevitably increasing permittivity. Therefore, a matched impedance is possible to maintain the high transmission even in the shorter wavelength range.
Figure 6-4. Magnetic field and current distributions of the metamaterial band pass filter at two magnetic resonance modes. (a) Current distribution on the top Au layer at 3.7 µm (left) and 2.85 µm (right). (b) Current distribution on the bottom Au layer at 3.7 µm (left) and 2.85 µm (right). (c) 3D tilted view of magnetic field distribution in the structure at 3.7 µm (left) and 2.85 µm (right). (d) Top view of magnetic field distribution in the structure at 3.7 µm (left) and 2.85 µm (right).
6.3.3 Role of Nano-Notches

In order to appreciate the role of notches in tailoring both the permittivity and permeability, a notch-free structure was simulated to be compared with the designed filter structure. For a full comparison, the scattering parameter amplitudes and effective medium parameters are plotted in Figure 6-3 and 4 (bottom parts), informing the difference in terms of device performance as well as the underlying physics. As can be seen in the permeability profiles (Figure 6-4(a) bottom), the main magnetic resonance at 3.7 µm with the absence of the notches is almost saturated, exhibiting a huge amount of loss that is unfavorable in any transmissive applications. The second magnetic resonance, attributed to a decrease of the capacitance and an increase of the inductance due to the absence of the notches, is shifted from 2.85 µm to 2.6 µm with a small increase of the quality factor.

The capacitance decrease and inductance increase also give rise to a sharper slope and a shorter effective plasma wavelength in the effective permittivity along with a more noticeable anti-resonance at the wavelength where the main magnetic resonance occurs. The unbalanced permittivity and permeability result in a refractive index with an evanescent mode in the wavelength range of interest, i.e. dominating imaginary part over the real part. Another adverse consequence of this unbalance is a poor impedance match, leading to the near-unity reflection within the targeted pass band range. This notch-free metamaterial performs as a nice mirror rather than a good transmitter.

To realize a high performing metamaterial, the dimensions of these notches need to be carefully tuned and precisely fabricated, since they have large impacts on the electromagnetic properties of the device. The dispersion tailoring capability of these notches and the sensitivity of the device to feature dimensions are closely interrelated, and therefore both of them are required to be well balanced to optimize the trade-off. Although a more complicated structure with a better
control over the resonance profiles may offer increased flexibility in shaping the dispersive permittivity and permeability, as a proof-of-concept device, these notches are considered here as the perturbation elements and optimized to achieve the proposed filter function. The entire structure can be readily implemented by current nano-fabrication technology due to the minor modifications to the conventional fishnet geometry.

6.4 Fabrication

As pointed out recently [42] and shown in the previous chapter, the substrate breaks the vertical symmetry of the entire structure, thus bringing unwanted additional biaxiality (i.e., magneto-electric coupling). This in turn induces nonnegligible orthogonal polarization components in the transmission. To make matters worse, the substrate also causes more reflection loss due to the impedance mismatch on the interfaces of the metamaterial.

To overcome these disadvantages of having a supportive substrate, the optimized metamaterial pass band filter was fabricated to be free-standing by standard electron beam (E-beam) lithography and reactive ion etching (RIE), followed by a peeling-off process to release the structure from the sacrificial wafer (see more fabrication details in Chapter 4). The fabrication procedure began by first depositing the tri-layer stack composed of a 30 nm thick bottom gold layer, a 450 nm thick polyimide layer, and a 30 nm thick top gold layer. A 100 nm thick silicon dioxide (SiO$_2$) layer, grown on top of the tri-layer stack by plasma enhanced chemical vapour deposition (PECVD), was then etched using RIE with a patterned E-beam resist as an etching mask that was defined by E-beam lithography. Finally, the etched pattern in the SiO$_2$ layer was transferred into the layers using consecutive RIE processes defining nano-scale notch-loaded air holes. The fabricated structure was removed from the sacrificial wafer by etching the underlying SiO$_2$ layer and mounted on an aluminium test frame for characterization.
As the scanning electron micrograph (SEM) image captured from the top of the sample shows in Figure 6-4(a), the structure was almost identically reproduced from the design with a fabrication error less than 2% in dimensions. Importantly, the dimensions of the critical notches are around 200 nm, only 2 nm away from the original design. The vertical sidewalls (Figure 6-4(b)) also ensure identical dimensions for all three layers of the sample, preventing another source of bianisotropy (i.e., sidewall angle induced magneto-electric coupling), which is present in most of the previously demonstrated optical metallo-dielectric metamaterials.
Figure 6-5. FESEM images of the fabricated metamaterial filter (a) Top view of the fabricated metamaterial filter structure. The inset shows a magnified view of the structure. (c) Tilted view of the fabricated metamaterial filter structure to show the side walls and its free-standing nature.
6.5 Characterization

The fabricated sample was characterized using a Fourier transform IR (FTIR) spectrometer (Bruker Optics IFS-66 main compartment) equipped with a liquid nitrogen cooled mercury cadmium telluride (MCT) detector. The transmission and reflection were measured at normal incidence angle. For the reflection measurement, a custom optical setup composed of mirrors and a beam splitter was mounted in the main compartment of the FTIR to collect the specular reflection at normal incidence. The absolute reflection amplitude was then determined by referencing the measured values to the reflectivity of a Au mirror, and the absolute transmission amplitude was normalized to an air background.

The measured scattering parameter amplitudes shown in Figure 6-6 are found in strong agreement with simulated predictions. The experimentally measured pass band is slightly widened, extending to the range from 2.95 μm to 3.05 μm with an in-band insertion loss smaller than 1 dB. This broadening is caused by the lower quality factor of the resonances within the band, as seen in the reflection curve. This slight decrease in quality factor also leads to smaller roll-off speed on both sides of the pass band, around 76 dB/μm and 91 dB/μm on the short and long wavelength sides, respectively. The transmission window has a flattened shape with a transmission variation of 0.6 dB. The transmission outside the pass band is maintained below −10 dB at longer wavelengths and slightly larger than −10 dB in the shorter wavelength region, but overall the amplitude is still below −8.3 dB, i.e. less than 15% of the incident light is passed through.
The amplitudes of transmission and reflection of both simulation (top) and FTIR measurement (bottom) are plotted for comparison.

**Figure 6-6.** Scattering parameter measurement of the fabricated metamaterial band pass filter. The amplitudes of transmission and reflection of both simulation (top) and FTIR measurement (bottom) are plotted for comparison.

### 6.6 Summary

The concept of dispersion engineering was applied to synthesize a metamaterial pass band filter for broad band application in the mid-IR wavelength range from 2.5 µm to 4 µm. The optimized polarization-insensitive impedance-matched free-standing meta-filter exhibits a flattened-top pass band from 3.0 µm to 3.5 µm with a suppressed group delay variation less than
This was achieved by tailoring both the permittivity and permeability over the wide range of wavelengths to control over the slopes as well as the values, enabling the ability of shaping the scattering parameters according to the proposed filter function. The GA optimized filter structure was fabricated for characterization, and FTIR scattering measurements confirmed its filtering performance. It is expected that the demonstration of this proof-of-concept meta-filter extends the concept of index engineering to dispersion engineering in the optical range for diversification of the optical functionalities.
6.7 References

Chapter 7

Metamaterial Absorber

This chapter demonstrates a conformal metamaterial absorber with a narrow band, polarization-independent absorptivity of >90% over a wide ±50° angular range centered at mid-infrared wavelengths of 3.3 μm and 3.9 μm. The highly efficient dual-band metamaterial was realized by using a genetic algorithm to identify an array of H-shaped nanoresonators with an effective electric and magnetic response that maximizes absorption in each wavelength band when patterned on a flexible Kapton and Au thin film substrate stack. This conformal metamaterial absorber maintains its absorption properties when integrated onto curved surfaces of arbitrary materials, making it attractive for advanced multi-spectral coatings that modify the infrared signature of the protected surface. Modeling work presented here, including field simulations and GA synthesis was performed by Zhi Hao Jiang under the advisement of Dr. Douglas H. Werner.

7.1 Motivation

Optical metamaterials [1-2] are composite structures composed of nanoscale resonators organized to give a highly customized electric and magnetic response over a specific wavelength range of interest. Most of the revolutionary new optical devices that are being enabled by the unique effective medium properties of the metamaterial require a negative, zero, or graded refractive index with extremely low absorption loss. Several prominent device examples that
exploit these properties include perfect lenses [3-4], flat collimating lenses [5], and invisibility cloaks [6-7]. Another far less studied class of metamaterials relies on creating structures that are tailored for complete absorption of the incident light in one or more wavelength bands independent of polarization and incidence angle [8-9]. Thus metamaterial absorber (MMA) designs must balance the electric and magnetic resonances within each wavelength band to simultaneously minimize reflection and transmission, and hence maximize absorption, for both the incident electric and magnetic fields. The availability of single and multi-band MMAs could provide significant performance improvements for diverse applications including microwave-to-infrared signature reduction [8-11], bio-chemical spectroscopy [12-19], thermal imaging [20-22], and solar energy conversion.

Rapid progress has been made in demonstrating MMAs that operate in the microwave and terahertz (up to 1.6 THz) regimes [8-17]. A number of single-band device designs were reported with measured polarization independent absorptivity greater than 90% for angles of incidence from 0° to 60° at frequencies up to 1.6 THz [10, 12]. More advanced multi-band terahertz MMAs with up to three well defined absorption bands have also been designed and fabricated [14-16]. Currently, the best performing dual-band MMA has a maximum absorptivity of 85% at 1.41 THz and 94% at 3.02 THz measured at a 30° incidence angle [15]. Several of the terahertz devices incorporated a flexible Kapton film as the dielectric spacer to enable their use in conformal coatings [12, 14]. A new type of absorber with a 2D omnidirectional broadband response [11] was designed using transformation electromagnetics approaches [23].

Fewer high-performance infrared-to-visible MMAs have been experimentally verified because the aggressively scaled nanoresonators needed to reach these shorter wavelengths impose even stricter constraints on the metamaterial design and fabrication process [18, 22]. Recently, an angularly-tolerant single-band MMA designed for near-infrared operation at 1.6 μm was implemented by patterning a periodic array of simple circular Au features on a planar magnesium
difluoride and Au thin film stack [18]. The absorptivity of this device measured at normal incidence using unpolarized light was 99%. In the mid-infrared (mid-IR) at 6 µm, a MMA design composed of an array of nanoscale Au crosses on a planar alumina and Au thin film stack achieved a near-unity absorptivity of 97% at normal incidence [22]. In contrast, multi-band MMAs require nanostructures with more sophisticated geometries to induce an additional electric and/or magnetic resonance for each absorption band, which forces additional restrictions on the design. Moreover, mechanically flexible constituent materials such as Kapton must be integrated to realize MMA coatings for curved surfaces. This added complexity has hindered advances in multi-band infrared MMAs despite the need for future coatings and devices with strong multi-spectral absorption properties in the aforementioned applications.

In this research, to address the current limitations of MMAs, a conformal MMA was synthesized to have two nearly perfect, narrow absorption bands centered at mid-infrared wavelengths of 3.3 µm and 3.9 µm. A genetic algorithm (GA) was used to identify an array of H-shaped nanoresonators on a flexible Kapton and Au thin film stack that excite the appropriate electric and magnetic resonances for strong absorption in each band. Measurements of a fabricated MMA are in strong agreement with theoretical predictions, showing polarization-independent absorptivity greater than 90% over a ±50º angular range in both of the targeted wavelength bands. Full wave simulations that illuminate a curved metal surface protected with this conformal MMA coating confirm that nearly all of the reflected light is eliminated within the two absorption bands, which is in sharp contrast to the unprotected surface. This work represents a significant step toward realizing high efficiency, multi-band MMA coatings to modify the signature of highly reflective curved surfaces within the 3 to 5 µm atmospheric window. The following sections discuss design, fabrication, and characterization of this MMA.
7.2 Electromagnetic Design Optimization

Our dual band mid-infrared MMA employs a three-layer metallo-dielectric stack composed of two gold (Au) layers—a doubly periodic array of electrically isolated nanoresonators at the top and a solid ground plane at the bottom—separated by a thin dielectric layer. Kapton was chosen for our dielectric layer because it is a highly durable and flexible polymer that can easily conform to the topography of most practical curved surfaces. The array of Au nanostructures on the top screen create a resonant electric response, while the Au ground plane functions together with the top screen to produce strong coupling to the magnetic component of the incident light radiation. The continuous Au ground plane, which is thicker than the penetration depth of light in the mid-infrared wavelength regime, prevents transmission of incident radiation through the structure. Therefore, strong absorption is achieved by minimizing the in-band reflection. Importantly, the Au ground plane also decouples the electromagnetic properties of the MMA coating from the surface it protects, allowing integration onto curved surfaces of arbitrary materials.

The specific goal was to design a periodic array of nanoresonators that gives two polarization independent absorption bands centered at 3.3 μm (90 THz) and 3.9 μm (77 THz) with absorptivity greater than 90%, i.e., at least 10 dB attenuation, over a ±50° field-of-view. The two closely spaced absorption band wavelengths were selected arbitrarily within the 3 to 5 μm atmospheric window for this proof-of-concept design. To achieve such dual-band performance using the described three-layer structure, a robust GA [24] coupled with a full-wave electromagnetic solver was employed to optimize the geometry and dimensions of the structure to best satisfy the user-defined requirements. For each of the starting candidate nanoresonator designs, the unit cell of the periodically patterned top Au layer was divided into a 14 × 14 grid of pixels that were randomly assigned a binary value corresponding to the presence “1” or absence
“0” of Au on the pixel. The Au pattern was further constrained to possess eight-fold symmetry to achieve a polarization independent absorber response. During the optimization, the unit cell size and the Kapton thickness were allowed to vary over a prescribed range, where the unit cell size was restricted to be smaller than half-wavelength to suppress higher order diffractions. To accurately account for the material dispersion in the design, the measured optical constants of the Au and Kapton thin film were used in the optimization. Furthermore, nanofabrication design constraints were incorporated to ensure that the optimized structure could be easily fabricated without modifying the nanoresonator geometry [25-26].

During the GA evolution, the wavelength-dependent scattering parameters of each candidate design was calculated using the Ansoft High Frequency Structure Simulator (HFSS) full-wave finite-element solver with appropriate boundary conditions assigned to approximate a TEM wave incident on the structure at different angles. The absorptivity was calculated by

\[ A_{TE,TM} = 1 - T_{TE,TM} - R_{TE,TM}, \]

where \( R_{TE,TM} = |S_{11}|^2 \) and \( T_{TE,TM} = |S_{21}|^2 \) represent the TE and TM reflectivity and transmittance, respectively. The absorptivity was evaluated against an ideal dual-band absorber response to determine its cost, given by:

\[ \text{Cost} = \sum_{\lambda} \sum_{\theta_i} [(1 - A_{\theta_i,TE}) + (1 - A_{\theta_i,TM})] \tag{7-1} \]

where \( \lambda \) is the wavelength of the target bands (3.3 \( \mu \)m, 3.9 \( \mu \)m) and \( \theta_i \) is the desired angle of incidence range (0° to 50°). The GA evolved the top Au screen nanoresonator geometry, unit cell size, and Kapton thickness until it converged to a sufficiently low cost solution, i.e. the optimized design was achieved.

The diagram of the GA-optimized dual-band MMA design is displayed in Figure 7-1, including its geometry and dimensions. The top Au screen is made up of a doubly periodic array of orthogonal stub-loaded H-shaped (SLH) nanoresonators identified by the GA, which have a
central connecting bar that is 630 nm long and 210 nm wide and two arms that are 840 nm long and 105 nm wide. Each arm is offset from the edge of the central bar by 105 nm and the arms are separated from one another by 210 nm. The total thickness of the three-layer structure is 200 nm, less than 1/15 of the operating wavelength. This design satisfies the nanofabrication design constraints imposed during design optimization, which ensures that the exact structure can be realized experimentally to minimize discrepancies that would degrade the resonant electromagnetic properties compared to theory.

Figure 7-1. Diagrams of the optimized dual-band MMA coating. (a) Top view of a single unit cell with dimensions $a=1475$ nm, $g=840$ nm, $w=210$ nm, $c=105$ nm, $w=210$ nm, and $h=315$ nm. (b) Perspective view of a unit cell with thickness $d=200$ nm (Au: 50 nm, Kapton: 100 nm).

The simulated reflectivity of the dual-band MMA at normal incidence as a function of wavelength is shown in Figure 7-2 for both polarizations. Transmission (not shown) is zero due to the Au ground plane. Two strong absorption bands are clearly resolved at the target wavelengths of 3.3 μm and 3.9 μm. Both bands have a $-10$ dB bandwidth of $\sim0.1$ μm with a maximum absorptivity of 94.7% or $-0.24$ dB (reflectivity of 6.3% and $-12$ dB) at 3.3 μm and of 99.6% or
−0.02 dB (reflectivity of 0.4% and −24 dB) at 3.9 μm. Additional reflectivity data simulated at oblique incidences are plotted in Figure 7-3(a) and Figure 7-4(a). The calculated angular dispersion of the absorption for both polarizations is shown in Figure 7-5(a). The two vertical red strips demonstrate that the two absorption peaks remain centered at 3.3 μm and 3.9 μm over a broad range of incidence angles for both polarizations. The absorptivity in both bands remains >91% over a wide field-of-view of ±50° due to the efficient excitation of both electric and magnetic resonances. Further investigation shows that this MMA still achieves absorptivity >60% for TE polarization and >85% for TM polarization in both bands at an incident full-angle of 160° (Figure 7-4(a)). Moreover, in the course of design optimization, we observed a design tradeoff between the absorbing efficiency and the desired number of absorption bands, which is similar to the findings in other studies [27-28]. In spite of these tradeoffs, the optimized dual-band MMA coating achieves all of the original performance goals.

**Figure 7-2.** Simulated and measured reflectivity of the MMA coating under TE (left) and TM (right) incident radiation at normal incidence.
Figure 7-3. Simulated and measured reflectivity at various angles of incidence (0°, 15°, 25°, 35°, 45°, 50°). (a) Simulated TE (left) and TM (right) reflectivity. (b) Measured TE (left) and TM (right) reflectivity. The two vertical dashed lines indicate the wavelengths of minimum reflectivity.
Figure 7-4. Simulated reflectivity and reflection phase at various angles (a) TE (left) and TM (right) reflectivity at various angles of incidence (60°, 70°, 80°). (b) TE (left) and TM (right) reflection phase at various angles of incidence (0°, 15°, 25°, 35°, 45°, 50°). The two vertical dashed lines indicate the wavelengths of minimum reflectivity.
Figure 7-5. The angular peak dispersion of the MMA absorption spectra. (a) Contour plot of simulated absorptivity as a function of wavelength and angle of incidence under TE (left) and TM (right) incident radiation. (b) Contour plot of measured absorptivity as a function of wavelength and angle of incidence under TE (left) and TM (right) incident radiation.
7.3 Principles of Operation

To understand the contribution of each H-shaped Au nanoresonator segment to the electric and magnetic response, the current distributions of the MMA at the center wavelength in each absorption band under a normally incident illumination with the electric-field linearly polarized in the x-direction are shown in Figure 7-6. For the first absorption band at 3.3 µm, the currents are primarily concentrated on the two arms of the SLH nanoresonator and the corresponding area of the ground plane underneath the arms (Figure 7-6(a)). Similar to the so-called fishnet [29] and paired nanorod [30] structures, the antiparallel currents give rise to a magnetic resonance. In this band, the strength of the magnetic resonance overwhelms the electric response, and thus the strong absorption can be attributed to the nearly pure magnetic response of the structure. This dominant magnetic response indicates that an artificial magnetic conducting (AMC) condition [31] is satisfied, as confirmed by the zero reflection phase (Figure 7-4(b)) that is maintained regardless of the angle of incidence.

For the second band at 3.9 µm, the current is mainly confined on the top Au layer and is concentrated in the connecting bar and the two arms of the SLH nanostructures, which behave as electric LC resonators (Figure 7-6(b)). The inductance is introduced by the middle connecting bar, while the gaps between the two arms provide the capacitance. Therefore, the electric resonance is driven when the incident electric field is parallel to the arms. In addition, a magnetic resonance is also introduced by the antiparallel currents on the connecting bar and the corresponding area on the bottom Au ground plane. The combined contribution of both of these electric and magnetic resonances produces the strong absorption at 3.9 µm, consistent with the −90º reflection phase that is shown in Figure 7-4(b).
Figure 7-6. Finite-element method simulations of the unit cell current distributions at normal incidence. (a) 3D view of the current distribution in the top Au nanoresonator layer (top) and bottom Au ground plane layer (middle) for the first absorption band at 3.3 µm. The bottom plot shows the cross-sectional view of the current distribution. (b) 3D view of the current distribution in the top Au nanoresonator layer (top) and bottom Au ground plane layer (middle) for the second absorption band at 3.9 µm. The bottom plot shows the cross-sectional view of the current distribution.
7.4 Curved Surface with a MMA Coating

To investigate the performance of this conformal dual-band MMA for real-world applications such as infrared/optical signature modification, a 3D full-wave simulation was conducted on the MMA coating a highly reflective curved surface. As the schematics in Figure 7-7 illustrate, the field pattern of a curved Au ground plane is compared with that of the same ground plane protected by the MMA when illuminated by an obliquely incident beam with a finite beamwidth. The insets show the 3D views of the structures for each case. For the simulation of the MMA-coated curved metal ground plane surface, the HFSS finite element solver was employed. In the simulation domain, a strip of the MMA with a width of one unit cell in the $x$ direction and a length of twenty-six unit cells was considered. To mimic a 1D infinite structure, perfect electric conducting (PEC) boundary conditions were assigned to the front and back walls in the $x$ direction for $TE$ polarization and perfect magnetic conducting (PMC) boundary conditions for $TM$ polarization. A waveguide was connected to the semicircular air box as a feed for a finite width beam impinging obliquely on the structure. Dispersive optical constants measured by variable angle spectroscopic ellipsometry were also included to accurately model the Au and Kapton layers.

Without the MMA coating, the curved ground plane creates strong reflections in multiple directions for each band as shown in Figure 7-8(a). The field snapshots show that standing wave patterns arise due to the interference between the incident and reflected waves. However, when the MMA covers the curved surface, reflection is nearly eliminated for both polarizations in both bands, as displayed in Figure 7-8(b) and (c). A slight reflection remains near the horizon, which is caused by the portion of the curved MMA that is illuminated by waves at near-grazing incidence angles. This can be attributed to the reduction in absorptivity that occurs at large incidence angles, particularly for the $TE$ polarization (Figure 7-4(a)). The considerable reduction in
reflection achieved by the MMA coating confirms that its performance is retained when it protects a highly reflective curved surface. In addition, as is evident from Figure 7-8(b) and (c), the field is strongly confined within the thin functional layer (only ~1/15λ) of the MMA indicating a high absorptivity-over-thickness efficiency that is superior to conventional (non-metamaterial) absorbers. Moreover, no fields are observed behind the solid Au ground plane; accordingly, the performance will not be affected by the object that the MMA covers, making it useful as a coating on various metal and dielectric surfaces.

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**Figure 7-7.** (a) Schematics of a curved metal surface (50 nm Au) producing strong reflection in multiple directions when illuminated by an incident beam. (b) Schematics of a MMA coated curved metal surface (50 nm Au) with significantly reduced reflection when illuminated by an incident beam. The inset shows a 3D view of the MMA coated curved metal surface.
Figure 7-8. Simulation of the electric field magnitude for the curved metal surfaces at 3.3 µm (left) and 3.9 µm (right). (a) Without MMA coating. (b) With the MMA coating under TE incident radiation. (c) With the MMA coating under TM incident radiation.
7.5 Fabrication

Nanofabrication of the GA-optimized MMA began by thermally evaporating a 50 nm Au layer on a clean Si wafer. The intermediate Kapton layer was then deposited on top of this bottom Au layer by spin coating the wafer with the polyimide precursor (HD Microsystems PI2556:T9039 = 1:2) at 4000 rpm for 40 seconds. The as-spun polyimide film was soft baked at 120°C for one minute and then cured in a N₂-purged convection oven at 250°C for one hour in order to complete the imidization through elimination of the solvent. These conditions also ensured that the bottom Au layer was free from pore formation originating from migration and coalescence, which is critical to completely suppress the transmission.

The top Au nanoresonator array was defined using electron-beam lithography and a metal lift-off process. A layer of electron-beam resist (Nippon ZEP 520A) was spun onto the Kapton layer at 5000 rpm for 1 minute, followed by a soft bake at 180°C for 3 minutes. The amount of exposure dose during electron-beam lithography (Leica EBPG-5HR) was adjusted to achieve a reasonable correspondence in dimensions between the designed and fabricated MMA unit cell. Following exposure, the resist was developed in n-Amyl Acetate solvent for 3 minutes to define the H-shaped nanoresonator pattern. A 50 nm Au film was then evaporated and lifted-off by dissolving the resist (MicroChem NANO™ REMOVER PG). The field emission scanning electron microscope (FESEM) image in Figure 7-10(a) shows an array of well-defined nanoresonators. The three-layer metallo-dielectric MMA structure was removed from the handle substrate to demonstrate its mechanical flexibility and durability (Figure 7-10(b)). Since there is no abrupt nano-scale or micro-scale variation in this curved sample, the nanoresonators maintain good adhesion to the Kapton layer.
Figure 7-9. FESEM images of the fabricated H-shaped nanoresonator array. (a) Top view of the fabricated structure. The dashed yellow square indicates the unit cell. (b) Fabricated conformal MMA coating showing its mechanical flexibility and durability.
7.6 Characterization

The MMA reflectivity was measured using a Fourier transform IR (FTIR) spectrometer (Bruker Optics IFS-66 main compartment) equipped with a liquid nitrogen cooled mercury cadmium telluride (MCT) detector. For near normal incidence (<1° off normal) measurements, a custom optical setup composed of mirrors and a beam splitter was constructed in the main sample compartment of the FTIR to direct the reflected IR signal to the MCT detector. For oblique incidence measurements, the sample was mounted on a variable angle specular reflection accessory (Thermo spectra-tech Model 500) to collect the angle-resolved reflectivity data in 10° increments to a maximum angle of 50°. In each case, the absolute reflectivity was determined by referencing the measured values to the reflectivity of a Au mirror.

The measured reflectivity of the fabricated MMA at normal incidence is shown in Figure 7-2. The wavelengths corresponding to the maximum normal incidence reflectivity shifted by less than 3% from the simulated values, changing from 3.30 μm to 3.40 μm in the first band and from 3.90 μm to 3.85 μm in the second band. At these wavelengths, the measured reflectivity for both polarizations was 3.2% (−15 dB) and 4.0% (−14 dB), respectively. This represents a decrease in reflectivity from 6.3% to 3.2% (3.1% difference) in the first band and an increase from 0.4% to 4.0% (3.6% difference) in the second band. Additional reflectivity data measured at oblique incidences can be found in Figure 7-3(b). The absorptivity of the fabricated MMA, calculated from the measured reflectivity, is shown in Figure 7-5(b), as a function of both the wavelength and the angle of incidence. The two absorption peaks of the MMA remain above 90% for an incidence angle up to 50° for both polarizations, with the −10 dB bandwidth of both bands exhibiting a maximum 0.06 μm broadening compared to the simulated results.

The measured results confirm that the optical properties of the nanofabricated MMA coating agree well with the simulations for all of the initial design parameters. The sources of the
small differences between experiment and theory were identified through an in-depth study of the fabricated structures by FESEM. Simulations reveal that the primary cause of the wavelength shift is the slight rounding at the ends of the patterned Au nanoresonators, which changes the effective electrical length of the central connecting bar and the two arms. Two primary factors contribute to the degradation in bandwidth. First, the slightly tapered sidewall profile (~80°) of the evaporated Au nanoresonators results in some degradation in the quality factor of the resonances. Second, variations in the Au nanoresonator dimensions across the large area MMA (~5%) can give slightly shifted absorption bands, resulting in a broadened integrated line shape of the absorption peaks from the large area structure. Further optimization of the Au nanoresonator fabrication processes is expected to produce MMA coatings with even better correspondence to theory.

7.7 Summary

A polarization insensitive, dual-band MMA was shown with measured absorptivity greater than 90% over a ±50° incidence-angle range in the mid-infrared at 3.3 μm and 3.9 μm. Electromagnetic simulations confirm that the absorption properties of the metamaterial are due to the electric and/or magnetic resonances excited by the periodic array of H-shaped nanoresonators patterned on a Kapton and Au thin film stack in both wavelength bands. The highly conformal nature of the MMA coating was demonstrated experimentally, and 3D full-wave simulations demonstrate that the absorbing properties are maintained when it is placed on a highly reflective curved surface. Because the performance of the MMA is independent of the material it protects, it is promising for a variety of applications, including modifying the mid-infrared signature of curved metal surfaces.
7.8 References


Appendix: Analysis and Synthesis Methods

Theoretical calculations are always important in the course of designing optical nanostructures as well as in the stage of post-processing the measured data for verification. In this section, a periodic finite element-boundary integral (PFEBI) method, which is a full-wave electromagnetic analysis technique that was widely used in this research, is introduced along with a genetic algorithm (GA) optimization method. GA optimized optical metamaterials are fabricated and characterized using an inversion algorithm, which retrieves effective medium parameters from the measured scattering coefficients.

A.1 Periodic Finite Element Boundary Integral Method

A PFEBI method can be used to calculate the scattering parameters of a periodic optical structure. This hybrid method is the combination of the finite element method (FEM) and the boundary integral method (also referred as the method of moments), enabling efficient numerical simulations of inhomogeneous material layers [1]. This method starts with considering a unit cell of the periodic optical structure. Owing to the periodicity, the numerical calculation is only necessary within a unit cell when proper boundary conditions are applied, which saves the time and resources. In Figure A-1, a unit cell of the periodic structure is described with four faces (F₁, F₂, F₃, F₄) and periodicities of pₓ and pᵧ. A standard FEM [1] procedure performs the field calculation inside the unit cell, and the computation domain is terminated in the z-direction by applying the mixed potential integral equation (MPIE) [2] on the top and bottom boundaries of the unit cell. Using these constraints, the pertinent finite element function is written as
\[-jk_0Z_0 \int_S \left( n \times W \right) \cdot H_{nm} \, ds = \int_S \left[ \frac{1}{\mu} \left( \nabla \times W \right) \cdot \left( \nabla \times E \right) - k_0^2 \varepsilon_r W \cdot E \right] dv \]

\[-2k_0^2 \int_S \int_S G_p \left( r, r' \right) \left( n \times W \right) \cdot \left( n \times E \right) ds \, ds \]

\[-2k_0^2 \int_S G \left( r, r' \right) \nabla \cdot \left( \nabla \times W \right) \nabla \cdot \left( \nabla \times E \right) ds \, ds \]

where \( G_p(\vec{r}, \vec{r}') \) is the periodic Green’s function, \( S \) represents the top and bottom surfaces of the unit cell, \( k_0 \) and \( Z_0 \) are the wave number and impedance of the free-space, respectively, \( \vec{W} \) is the weighting function, and \( V \) represents the volume of the unit cell. In order to relate the fields on the opposing sides of the faces, the following phase boundary condition can be applied.

\[ e_{F_a/F_{a'}} = e_{F_b/F_{b'}} \exp \left( -j(k_0 \sin \theta \cos \phi D_z + k_0 \sin \theta \sin \phi D_y) \right) \]

where \( e_{F_a/F_{a'}} \) represents the unknown field at an edge of the faces, and \( \theta \) and \( \phi \) are the angles of the incident field. By discretizing and calculating the unit cell volume integrals in Eq. A-1, the coefficients of the basis functions can be acquired. The electric field on the top and bottom faces of the unit cell is then produced using the basis functions, which in turn is used to calculate the scattering coefficients of the structure.

**Figure A-1.** Unit cell of the periodic optical structure with sub-gridded meshes using FEM.
A.2 Genetic Algorithm Optimization

A GA is a heuristic search algorithm that is inspired by natural evolution [3]. To optimize multiple parameters simultaneously, this algorithm relies on inheritance, mutation, selection, and crossover, mimicking the evolution process in nature. When the GA is coupled with a full-wave electromagnetic solver such as PFEBI, the geometries and dimensions of optical structures can be optimized for user-specified criteria. The process flow of a GA is described in Figure A-2(a). To find an optimum solution to meet the requirements, first of all, the cost function needs to be defined to properly account for all the constraints. The searching process begins with the random generation of an initial population of design members, which are represented by binary strings that contain the structural information.

![Flow diagram of a GA](image)

**Figure A-2.** (a) Flow diagram of a GA. (b) Example of a unit cell of a metallo-dielectric structure.

In Figure A-2(b), a design example is shown for a metallo-dielectric unit cell that is subdivided into an 8×8 array of pixels. Each pixel is encoded as a “1” or “0” for the presence or
absence of metal. All other parameters including pitch size, thickness of each layer are also encoded into binary string to represent each member. The scattering coefficients of each population member are then evaluated by an electromagnetic solver and compared to a user-defined cost function. By choosing best performing designs and applying mutation and crossover, the next generation is produced for evaluation. This simple loop iterates until the cost is minimized to within the design tolerance, and the desired EM properties are achieved. To avoid unrealistic designs in terms of fabrication, fabrication constraints can be enforced during the GA synthesis process. These fabrication constraints remove single isolated pixels or diagonal connections during the cost evaluation of each member.

A.3 Effective Medium Parameter Retrieval

For a fabricated index engineered metamaterial, characterization is crucial to verifying its effective optical properties. In most of cases, experimental observables are restricted to the scattering parameters. Sometimes only the intensity parts are accessible due to the limited capability of the available measurement methods. Nonetheless, the optical properties of the metamaterial are still attainable because the scattering parameters are always governed by the optical properties of the medium. A standard procedure for confirming the optical properties of the fabricated metamaterial is to compare the extracted effective optical parameters from the simulation and the experiment [4], which include the electric permittivity $\varepsilon$, magnetic permeability $\mu$, refractive index $n$, and impedance $Z$. Assuming that a slab of metamaterial with thickness of $d$ is surrounded by air under normal incidence as depicted in Figure A-3(a), both the transmission coefficient $t$ and reflection coefficient $r$ can be expressed in terms of refractive index $n$ and impedance $Z$ when the effective medium approach is valid (Figure A-3(b)): 
\[ t' = \left[ \cos(nkd) - \frac{i}{2} \left( Z + \frac{1}{Z} \right) \sin(nkd) \right]^{-1}, \quad t' = e^{ad} \]  
(A-3)

\[ r = \left[ -\frac{i}{2} \left( Z - \frac{1}{Z} \right) \sin(nkd) \right] t' \]  
(A-4)

To obtain the explicit expressions for effective \( n \) and \( Z \), the above equations can be inverted as follows:

\[ \cos(nkd) = \frac{1 - r^2 + t^2}{2t} \]  
(A-5)

\[ Z = \pm \sqrt{\frac{(1 + r)^2 - t^2}{(1 - r)^2 - t^2}} \]  
(A-6)

With \( n \) and \( Z \) available, the effective permittivity \( \varepsilon \) and permeability \( \mu \) can be readily calculated according to the simple relationship:

\[ \varepsilon = \frac{n}{Z} \]  
(A-7)

\[ \mu = nZ \]  
(A-8)

However, due to the multi-valued characteristics in trigonometric and square root functions in the expressions for \( n \) and \( Z \), additional constraints are required to solve for \( \varepsilon \) and \( \mu \). Since any passive material should satisfy the causality condition, the imaginary parts of \( n \) and the real part of \( Z \) should be positive. Under this condition, the signs in Eq. A-6, A-9, and A-10 can be decided.

\[ n'' = \pm \frac{1}{kd} \text{Im} \left[ \cos^{-1} \left( \frac{1 - r^2 + t^2}{2t} \right) \right] \]  
(A-9)

\[ n' = \pm \frac{1}{kd} \text{Re} \left[ \cos^{-1} \left( \frac{1 - r^2 + t^2}{2t} \right) \right] + \frac{2 \pi m}{kd} \]  
(A-10)

Furthermore, the inverted \( n' \) is multivalued with many possible roots given by the integer \( m \) as seen in Eq. A-10. In order to resolve the root \( m \), the retrieval process begins far away from
the metamaterial resonances, where \( n \) can be selected that gives reasonable values for \( \varepsilon \) and \( \mu \) in the low frequency limit. Then, the index of refraction is traced to the wavelength of interest, maintaining continuity in \( n' \) across wavelengths. The branches for different values of \( m \) can be clearly separated when the thickness of the slab is much smaller than the wavelength.

**Figure A-3.** (a) Slab of metamaterial. (b) Slab of effective medium.

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### A.4 References

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