TOP-DOWN AND BOTTOM-UP INTEGRATION OF ENGINEERED NANOSTRUCTURES FOR METAMATERIALS

A Dissertation in

Electrical Engineering

by

Lan Lin

© 2016 Lan Lin

Submitted in Partial Fulfillment of the Requirements for the Degree of

Doctor of Philosophy

May 2016
The dissertation of Lan Lin was reviewed and approved* by the following:

Theresa S. Mayer  
Distinguish Professor of Electrical Engineering and Material Science and Engineering  
Associate Dean for Research and Innovation  
Dissertation Advisor  
Chair of Committee

Douglas H. Werner  
Professor of Electrical Engineering

Zhiwen Liu  
Professor of Electrical Engineering

Lasse Jensen  
Associate Professor of Chemistry

Kultegin Aydin  
Professor of Electrical Engineering  
Head of the Department of Electrical Engineering

*Signatures are on file in the Graduate School
ABSTRACT

The emergence of metamaterials has expanded the understanding of materials and their associated functionalities beyond the scope of naturally occurring substances. By properly engineering the electric and magnetic resonances of meta-atoms, which have a length scale smaller than the incident wavelength, novel artificial materials can be constructed. This research investigates novel optical metamaterials that are designed and fabricated with optimized subwavelength nanostructures and properly chosen constituent materials to meet user-specified criteria. Both top-down and bottom-up integration techniques were developed to explore the palette of materials and nanostructures that can be accessed. First, precisely defined subwavelength nanostructures fabricated using conventional top-down techniques were exploited to synthesize a metamaterial absorber, metamaterial waveplates, and a dielectric magnetic mirror. A single-layer metal nanostructure array that had been optimized by genetic algorithm was demonstrated to achieve near-ideal absorptivity from 1.77 μm to 4.86 μm and over incidence angles up to 45°. The performance of the fabricated absorber was verified using Fourier transform infrared (FTIR) spectroscopy. A half-wave plate and a quarter-wave plate that exhibit polarization conversion ratios and reflectivity greater than 92% from 640 nm to 1290 nm with incident angles up to 40° were investigated. A Mie resonance-based amorphous silicon nanoresonator array was designed, resulting in a lossless magnetic mirror at 1 μm with near unity reflectivity and zero reflection phase. The fabricated waveplates and magnetic mirror were characterized with custom-built measurement setups, and the results show excellent agreement with the design targets. Secondly, an alternative hybrid top-down and bottom-up integrated technique was developed to increase the range of materials and nanostructures beyond those available when using conventional top-down techniques and to synthesize optical metamaterials with reconfigurable structures/functionalities. A directed self-assembly strategy was investigated using an applied electric field to deterministically assemble...
pre-synthesized nanoparticles into predefined locations and to modulate the patterned structures. Based on this approach, reconfigurable two-dimensional ordered gold nanowire lattices with AC frequency-dependent lattice spacing were demonstrated. Electric field simulations and electric force calculations were performed to understand the formation of the lattices. Finally, a simplified electrical equivalent resistor and capacitor (RC) model was developed to explain the observed frequency dependent periodicity change.
TABLE OF CONTENTS

List of Figures ........................................................................................................................................... vii

Acknowledgements ....................................................................................................................................... xii

Chapter 1 Introduction ................................................................................................................................. 1
  1.1 Motivation and Background .................................................................................................................... 1
  1.2 Overview ............................................................................................................................................... 11
  1.3 References ........................................................................................................................................... 14

Chapter 2 Near-Ideal Optical Metamaterial Absorber with Super-Octave Bandwidth ......................... 17
  2.1 Background and Motivation .................................................................................................................... 17
  2.2 Design and Optimization ....................................................................................................................... 19
    2.2.1 Electromagnetic Optimization Methodology .................................................................................. 19
    2.2.2 Optimized Subwavelength Nanostructure Array for Broadband Absorption .............................. 23
    2.2.3 Resonant Electromagnetic Properties .......................................................................................... 25
  2.3 Nanofabrication and Optical Measurements .......................................................................................... 28
  2.4 Selection of Metal for Broadband Absorption ...................................................................................... 34
  2.5 Summary ............................................................................................................................................... 37
  2.6 References ........................................................................................................................................... 38

Chapter 3 Broadband and Wide Field-of-view Plasmonic Metasurface-enabled Waveplates ............... 40
  3.1 Background and Motivation .................................................................................................................... 40
  3.2 Principle of Reflective Plasmonic Metasurface-based Waveplate ....................................................... 42
    3.2.1 Structure and Operation of Anisotropic Metasurface .................................................................. 42
    3.2.2 Anisotropic Optical Response of the Metasurface Nanorod Building Blocks .............................. 44
  3.3 Optimized Broadband and Wide-angle Half-wave Plate ...................................................................... 48
    3.3.1 Design and Optimization of a Half-wave Plate ........................................................................... 48
    3.3.2 Experimental Realization of the Half-wave Plate ....................................................................... 52
    3.3.3 Optical Characterization of the Half-wave Plate ....................................................................... 53
  3.4 Demonstration of a Metasurface-based Quarter-wave Plate ................................................................. 55
  3.5 Summary ............................................................................................................................................... 58
  3.6 References ........................................................................................................................................... 59

Chapter 4 Dielectric Nanoresonator-Based Lossless Optical Perfect Magnetic Mirror with
Near-Zero Reflection Phase ....................................................................................................................... 62
  4.1 Background and Motivation .................................................................................................................... 62
  4.2 Electromagnetic Design and Simulation ............................................................................................... 64
  4.3 Fabrication and Characterization .......................................................................................................... 70
Chapter 5 Formation and Frequency Response of Two-Dimensional Nanowire Lattices......77

5.1 Motivation and Background.......................................................................................77
5.2 Assembly Experiment Setup......................................................................................79
5.3 Nanowire Lattice Formation and Frequency Response ............................................80
  5.3.1 Lattice Formation ............................................................................................ 80
  5.3.2 Frequency Dependent Lattice Restructuring ...................................................84
5.4 Summary ....................................................................................................................89
5.5 References..................................................................................................................90

Chapter 6 Summary and Future Work .....................................................................................93

6.1 Summary ....................................................................................................................93
6.2 Recommendations for Future Work...........................................................................95
  6.2.1 Light-Induced Tunable Assembly of a-Si:H Nanodisks .......................................95
  6.2.2 Frequency-Dependent Reconfigurable Assembly of a-Si Nanodisks .............100
  6.2.3 Nanoparticle Assembly with Predefined Structures ........................................103
6.3 References..................................................................................................................108

Appendix A Electromagnetic Design Optimization........................................................109
  A.1 Genetic Algorithm (GA) ................................................................................... 109
  A.2 Covariance Matrix Adaptation Evolution Strategies (CMAES) .......................111

Appendix B Assembly Electrode Fabrication.................................................................113

Appendix C Nanoparticle Fabrication and Characterization ............................................114
  C.1 PECVD aSi:H Particle Fabrication:................................................................. 114
  C.2 Effective Conductivity Characterization Approach...........................................116

Appendix D Anisotropic Lossy Dielectric Particle Rotation............................................119
LIST OF FIGURES

Figure 1-1. Flow diagram of the design optimization process..................................................3

Figure 1-2. (a) Schematic of unit cell. (b) FESEM image of the filter. (c) FTIR measurement and simulation result 20........................................................................4

Figure 1-3. Schematic of the DEP force. .................................................................................7

Figure 1-4. (a) Schematic of the parallel electrodes with an applied electric voltage. (b) Uniformly spaced Rh nanowire array 60, (c) Force distribution calculated by MST with COMSOL and MATLAB. .................................................................10

Figure 1-5. Diagram of the components to complete a directed-self-assembly system............11

Figure 2-1. Diagram of the MMA. (a) Cross-sectional view of the four-layer EBG-based broadband absorber. (b) Schematic of the Pd-based MMA structure optimized to have broadband absorption from 2µm to 5µm in the mid-IR. (c) Top view of the nanostructure unit........................................................................................................20

Figure 2-2. Simulation of angular dispersion of the MMA absorption spectra. (a) Normal incidence absorptivity for unpolarized, TE and TM illumination. Contour plot of absorptivity as a function of wavelength and incident angle from normal up to grazing under (b) unpolarized (c) TE and (d) TM illumination.........................................................24

Figure 2-3. Top view of finite-element method simulations of the current distributions in the top Pd screen for normal incidence at 1.50 µm and peak wavelengths of 2.13 µm, 3.00 µm, 3.70 µm, and 5.05 µm.................................................................26

Figure 2-4. Cross-section view of finite-element method simulations of the current distributions for normal incidence at 1.50 µm and peak wavelengths of 2.13 µm, 3.00 µm, 3.70 µm, and 5.05 µm..............................................................................................27

Figure 2-5. FESEM image of the nanofabricated nanostructures..............................................29

Figure 2-6. (a) Illustration of normal incidence reflection measurement setup. M: mirror, BS: beam splitter, S: sample. (b) Measured absorptivity for unpolarized, TE and TM illumination at normal incidence and compared with simulation result ..................31

Figure 2-7. (a) Illustration of off-normal incidence reflection measurement setup for characterizing the fabricated sample. Contour plot of measured absorptivity as a function of wavelength and angle of incident from normal up to 55º incidence under (b) unpolarize (c) TE and (d) TM illumination.................................................................32

Figure 2-8. (a) Top: FESEM image of unit cell for the fabricated sample of Pd based MMA, Scale bar is 200 nm. Bottom: Unit cell with 10 nm increase in Pd features and corner rounding to match the nanofabricated structure in the FESEM image. (b) Simulation at normal incidence under unpolarized illumination to study the sensitivity
of the MMA to nanostructure feature size changes and polyimide thickness variations.

Figure 2-9. (a) Diagram of the optimized Au-based MMA structure. (b) Top left: top view of one unit cell of the design. Bottom left: FESEM image of a unit cell of the fabricated structure. Scale bar is 200 nm. Right: Low-magnification FESEM image of the same structure. Scale bar is 600 nm. (c) Simulation and measurements for Au-based MMA under unpolarized illumination at normal incidence. The average Au thicknesses of the Au nanostructures determined by atomic force microscopy measurements are 10 nm, 11 nm, and 12 nm. (d) Simulation and measurements for Pd-based MMAs with Pd nanostructure thicknesses of 30 nm and 40 nm under unpolarized illumination at normal incidence.

Figure 2-10. Simulation of the Au based MMA at normal incidence under unpolarized illumination with Au screen thicknesses ranging from 10 nm to 14 nm to study the sensitivity of the MMA to nanostructure Au film thickness variations.

Figure 3-1. Schematics of the proposed plasmonic metasurface-based waveplates. (a) 3D view of the multilayer structure model including an anisotropic homogeneous metasurface located at a distance of \( t_m/2 \) above the SiO\(_2\) layer. The metasurface is characterized by electric and magnetic surface polarizability tensors \( \chi E \) and \( \chi M \). Optically thin strongly-coupled nanorod array is used to realize the metasurface. (b) Interference model for evaluating the optical response of the multilayer structure based on the surface polarizability tensor parameters of the metasurface.

Figure 3-2. Anisotropic scattering of single and coupled nanorod pair. (a) Schematics of an incident \( x \)- and \( y \)-polarized wave at normal incidence illuminating a single or a coupled nanorod. The dimensions are \( a_x = 250, a_y = 75, t_m = 30, g = 20 \) (all in nm). (b) Scattering field magnitudes (normalized) and phases of the single (top) and twin (bottom) nanorod. (c) Electric field distribution on the single nanorod at 930 and 550 nm and electric field distribution on the coupled nanorod at 1125 and 550 nm. (d) Dispersion of \( \Delta \phi \) showing the impact of nanorod length (\( a_x \)), nanorod aspect ratio (\( a_y/a_x \)), and gap size (\( g \)).

Figure 3-3. Broadband and wide-angle plasmonic metasurface-based half-wave plate. (a) Tilted 3D view of the metasurface-based half-wave plate. An s-polarized wave incident from an angle of \( (\theta_i, \varphi_i=135^\circ) \) is converted into a p-polarized wave upon reflection. Inset shows the side view of the nanostructure and light path. (b) Unit cell configuration of the optimized half-wave plate. The dimensions are \( a_x = 210, a_y = 70, p_x = 252, p_y = 252, t_m = 42, t_d = 114, t_m2 = 100 \) (all in nm). (c) Top-view FESEM image of a portion of the fabricated nanostructure showing the nanorod array. Scale bar: 400 nm. The inset shows the magnified top view of two by three unit cells. Scale bar: 100 nm. (d) Theoretically predicted and experimentally measured polarization conversion ratio (PCR) and reflection magnitude (Refl. Mag.) as a function of wavelength at different angles of incidence \( (4^\circ, 20^\circ, 40^\circ) \). (e) Theoretically predicted and experimentally measured polarization state in the plane perpendicular to the wave vector at 700, 900, and 1150 nm for different angles of incidence \( (4^\circ, 20^\circ, 40^\circ) \).
Figure 3-4. Schematic figure of the custom-built optical setup used for measuring the reflection magnitude and polarization of reflected light...............................55

Figure 3-5. Broadband and wide-angle plasmonic metasurface-based quarter-wave plate. (a) Tilted 3D view of the metasurface-based quarter-wave plate. A circularly-polarized wave incident from an angle of $(\theta_i, \phi_i=0^\circ)$ is converted into a linearly-polarized wave upon reflection. Inset shows the polarization of the reflected and incident waves in the plane perpendicular to the wave vector. The angle between the electric field and the plane of incidence of the reflected light is $45^\circ$. (b) Unit cell configuration of the optimized quarter-wave plate. The dimensions are $a_x = 180$, $a_y = 90$, $p_x = 240$, $p_y = 282$, $t_m = 40$, $t_d = 150$, $t_{m2} = 100$ (all in nm). (c) Top-view FESEM image of a portion of the fabricated nanostructure showing the nanorod array. Scale bar: 400 nm. The inset shows the magnified top view of two by three unit cells. Scale bar: 100 nm. (d) Theoretically predicted and experimentally measured polarization conversion ratio (PCR) and reflection magnitude (Refl. Mag.) as a function of wavelength at different angles of incidence (4°, 20°, 40°). (e) Theoretically predicted and experimentally measured polarization state in the plane perpendicular to the wave vector at 700, 900, and 1150 nm for different angles of incidence (4°, 20°, 40°). ..........57

Figure 4-1. (a) Unit cell configuration of the designed dielectric magnetic mirror. The length and width of the cross-shaped resonator are 431 nm and 116 nm, respectively. The $a$-Si thickness $t_d$ is 366 nm. (b) Tilted view of FESEM image for the fabricated all-dielectric magnetic mirror showing a well-defined $a$-Si nanoresonator array. Inset image shows the magnified unit cell. Scale bar: 100 nm. ........................................65

Figure 4-2. (a) Simulated (top red) and measured (bottom blue) reflection amplitude of the cross-shaped $a$-Si nanostructure. (b) Simulated (top red) and measured (bottom blue) reflection phase of the cross-shaped $a$-Si nanostructure. (c) Simulated (top red) and measured (bottom blue) transmission amplitude spectrum. (d) Simulated (top red) and measured (bottom blue) transmission phase spectrum........................................66

Figure 4-3. (a) Magnetic field (left) and electric field (right) distributions in the $a$-Si nanoresonator shown in Figure 4-1 (a) at the electric resonant wavelength of 1.14 $\mu$m. (b) Magnetic field (left) and electric field (right) distributions in the $a$-Si nanoresonator at the magnetic resonant wavelength of 0.99 $\mu$m.................................67

Figure 4-4. (a) Simulation study of reflection amplitude (top) and phase (bottom) shift with different $a$-Si nanoresonator thickness ($t_d$). (b) Reflection amplitude (top) and phase (bottom) shift with different $a$-Si resonator length ($L$). (c) Reflection amplitude (top) and phase (bottom) shift with different $a$-Si resonator width ($w$)...............................................69

Figure 4-5. Schematic of the spectral holography setup for (a) reflection measurement, and (b) transmission measurement configurations. BS: beam splitter, PCF: photonic crystal fiber, OSA: optical spectrum analyzer. .............................................73

Figure 5-1. (a) Side and top view representation of coplanar electrode setup, showing stem/bulb (S/B) geometry and optical micrograph of electrode gap (transmitted light; intensities in this image were inverted such that electrodes appear bright for clarity). (b) Transmission electron micrograph image of Au nanowires........................................79
Figure 5-2. Nanowire assembly between coplanar electrodes depicting the process of lattice formation (stem region). Sequential images taken at the same assembly location of the Au nanowires, as applied field strength was increased over ca. 13 min time period. Particles first organized along electrode edges with increased voltage. They then began to form bridges and eventually a lattice when frequency was increased. .................. 81

Figure 5-3. (a-c) Solid Au nanowires assembled within a stem/bulb electrode at constant field conditions (800 V/cm, 400 kHz, for the stem region). Arrow points to a chain of nanowires as it funnels from the bulb to the stem region of the electrode gap in less than two seconds. (d) Simulation showing field gradient at the top surface of the patterned stem/bulb electrodes using experimental bias conditions. Red indicates highest field gradient and blue indicates lowest. The field gradient is highest in the stem region. ...................................................................................................................... 83

Figure 5-4. (Top) Cartoons of solid Au nanowires illustrating induced dipole interactions between neighboring particles. (Bottom) Three-dimensional simulation showing charge density within solid Au nanowires in a non-uniform electric field. Electric field: (340 V/cm at 900 kHz, nanowire overlap length set at 0.75 µm. ........................... 84

Figure 5-5. Optical micrographs showing the change in particle overlap of Au solid nanowires at (a) 100 kHz and (b) 900 kHz with constant voltage (340 V/cm). As frequency increases, the overlap of neighboring particles decreases, creating a more open lattice. ...................................................................................................................... 85

Figure 5-6. Simulation of the electric force (x-component) on the Au nanowires in an AC electric field (340 V/cm, 100 or 900 kHz) as a function of Au overlap with vertical neighbor, d (µm). The top and bottom wires in each simulation are fixed while the left and right wires are simultaneously moved in the x-direction. The stable nanowire array configuration occurs when the electric force = 0. ............................................................ 87

Figure 5-7. Equivalent RC circuit diagram shows coupling between neighboring nanowires in both x-and y-directions. ...................................................................................................................... 88

Figure 6-1. (a) FESEM image of a-Si:H nanoparticles. (b) Illustration of a light-induced conductivity change. ........................................................ 101

Figure 6-2. Plot of the real parts of the Clausius-Mossotti factor as a function of frequency for a-Si:H and a-Si nanoparticles with different conductivity dispersed in EG. .......... 97

Figure 6-3. (a)-(c) Sequentially increased, reduced, and increased illumination intensity to selectively choose the assembly particle type at 500 kHz, 10^4 V/cm. (d) Selectively assembled single-particle-type 2D crystal within the mixed particle solution under low illumination intensity. (e) 2D crystal with mixed particle type is formed by increasing the a-Si:H nanoparticle conductivity, and both types of particles exhibit positive DEP. ............................................................ 99

Figure 6-4. FESEM image of fabricated a-Si nanodisks. ...................................................................................................................... 101
Figure 6-5. Electron-beam evaporated $\alpha$-Si particle orientation and frequency dependence with the same applied electric field of $10^4$ V/cm. (a) At 400 kHz (b) at 3 MHz. .............. 102

Figure 6-6. (a) Schematic figure of the assembly setup. (b) FESEM image of Si micro-ring with a radius of 10 $\mu$m, width of 500 nm and thickness of 230 nm. (c) Simulated electric field gradient distribution in the cross-sectional view. (d) Simulated electric field gradient distribution from the top view. ........................................................................ 104

Figure 6-7. (a-b) 1$\mu$m-diameter Ag-coated SiO$_2$ particle with applied electric field of 830 V/cm at two different frequencies. (c) 1$\mu$m-diameter polystyrene particle with applied electric field of 500 V/cm. ............................................................................................... 105

Figure 6-8. (a) FESEM image of a coupled Si micro-rings structure with a radius of 10 $\mu$m, width of 500 nm, thickness of 230 nm, and a spacing between two micro-rings of 200 nm. (b-c) 1 $\mu$m-diameter Ag-coated SiO$_2$ particles with an applied electric field of 417 V/cm at 20 kHz and 3 kHz, respectively. (d) 1 $\mu$m-diameter polystyrene particles with an applied electric field of 500 V/cm at 5 kHz. ................................................................................. 106

Figure A-1. Example of a unit cell. .......................................................................................... 109

Figure A-2. Flow chart of GA. ................................................................................................. 110

Figure A-3. Flow chart of CMA-ES$^2$. .................................................................................... 112

Figure C-1. Experiment observation of positive DEP (PDEP, left column) and negative DEP (NDEP, right column). (a) Stem/Bulb electrode with aSi:H particles. (b) Interdigitated electrode with aSi:H particles. (c) Quadruple electrode with polystyrene particles. ................................................................. 117
ACKNOWLEDGEMENTS

Attending graduate school was a major life transition for me from which I gained many valuable experiences. I would like to take this opportunity to thank many people who helped me go through this process and make my life at Penn State memorable. First and foremost, I would like to express my sincerest gratitude to my advisor, Dr. Theresa S. Mayer, for offering me this great opportunity to work on this research and collaborate with wonderful people. This work would not be possible without her support and guidance. Her academia vision and invaluable advice inspire me to explore new fields and have fun in science. I also want to thank Dr. Douglas H. Werner, Dr. Zhiwen Liu and Dr. Lasse Jensen for serving on my committee and providing valuable advice throughout my study and research.

Special thanks to my collaborators, Dr. Seokho Yun, Dr. Jeremy A. Bossard, Dr. Zhihao Jiang, Dr. Ding Ma, Sarah J. Boehm and Dr. Christine D. Keating for all the insightful discussions and joint efforts that we have made together on MRSEC projects. Many thanks to my labmates and the nanofab staff, Dr. Yu Yuwen, Dr. Xiahua Zhong, Dr. Wenchong Hu, Dr. Mengwei Kuo, Dr. Scott Levin, Dr. Oren Gall, Liu Liu, Xuexue Guo, Donna Deng, Daniel Schulman, Andrew Swisher, Dr. Bangzhi Liu, Dr. Chad Eichfeld, Shane Miller, Guy Lavallee, Michael Labella, Kathy Gehoski, and Bill Drawl for their kindness and help in both research and life.

I want to thank my parents and parents-in-law for their understanding, patience, constant support and encouragement. Last but not least, I would like to thank my husband, Yanhui Zhao, for his love and company during all these years.
Chapter 1

Introduction

The research presented in this dissertation focuses on the development of metamaterials for infrared and optical wavelength applications utilizing top-down and bottom-up techniques. This chapter provides a brief background on metamaterials along with a discussion of the limitation and challenges of current technologies. Finally, an overview of the subsequent chapters is included.

1.1 Motivation and Background

Optical metamaterials are a new class of artificially engineered composite materials composed of electromagnetic nanostructures with length scales much smaller than the wavelength of the incident light. The effective permittivity and effective permeability of such metamaterials can be tailored through proper design of the artificial subwavelength meta-atoms that couple with the incident electromagnetic field \(^1\text{-}^4\). These metamaterials can exhibit extraordinary optical properties that do not exist in naturally occurring materials, including zero and negative refractive indices \(^5\text{-}^{12}\). These properties can be used to create new optical devices such as optical absorbers, subwavelength imagers, and cloaks \(^13\text{-}^{19}\). Alternatively, a gradient (positive) index can also be created by combining two or more non-resonant subwavelength nanostructures. In this case, the effective index of each region of the material is determined by the volume fraction of each of the constituents.

The design of a metamaterial that provides a desired electromagnetic response is achieved by selecting the correct electromagnetic nanostructure geometry and the proper component
materials. For instance, an optically thin mid-infrared filter with a broad transmission band can be realized by adding deep-subwavelength nano-notches to a metallodielectric fishnet structure, and ±30 nm (equivalent to ~λ/100) deviation of the nano-notch length from the optimized value would result in a significant reduction in the average pass-band transmission from 82% to 64% \(^{20}\); a mid-infrared perfect absorber can be achieved by tri-layer disks composed of metal-dielectric-metal, and replacing the dielectric layer of Ge to GaAs in this stack would lead to a absorption peak shift from 7.0 \(\mu\)m to 8.5 \(\mu\)m \(^{21}\). Therefore, fulfilling all the user-specified criteria requires optimized nanostructure designs. In addition, discrepancies between simulations and experiments due to overlooked fabrication constraints and inaccurate properties of constituent materials need to be minimized to achieve robust designs \(^{22-25}\).

To address the stated issues and enrich the optimization process of the nanostructures to meet the customized optical response, an iterative design optimization strategy, as depicted in Figure 1-1, is used in this research to construct metamaterials with user-specified optical functionalities. Prior to beginning the design process, new nanofabrication and materials integration techniques must be developed, and optical properties of the constituent materials must be characterized. Using these factors as constraints, the nanostructure design is optimized using optimization algorithms, such as Genetic Algorithm (GA), Covariance Matrix Adaptation Evolution Strategy (CMA-ES), as described within the dashed blue box in Figure 1-1. The initial candidates are randomly generated and evaluated by Full-wave Electromagnetic Analysis with a cost function that defines the user-specified requirements. If the optimal design is not reached, a new generation of candidates will be generated based on the optimization algorithm. The optimization process is repeated until the cost function is minimized. Then, the sample with optimized dimensions is fabricated with the pre-developed technique and characterized by optical
measurement. Finally, the measured property is compared with the initial goal and the simulation results to verify the performance of the design. If the measured result does not match the initial goal, further improvements of the fabrication technology are needed.

An example of a metamaterial that was produced using this iterative design strategy is shown in Figure 1-2. A broadband low-loss optical metamaterial filter with high transmission over the wavelength range from 3.0 µm to 3.5 µm and high rejection outside this band was demonstrated based on a dispersion engineering method\textsuperscript{20}. Metamaterials with nanostructures composed of metals and dielectrics have been widely investigated due to their novel optical responses. However, the intrinsic dispersive optical properties of these materials limit their operational bandwidth to a narrow region. Recently, dispersion engineering has been used to design metamaterials with broad bandwidths in radio frequencies \textsuperscript{26}. By tailoring the dispersive and resonant response of the metamaterial based on its functionality, the dispersive band could be

Figure 1-1. Flow diagram of the design optimization process.
avoided. The optimized design of this metamaterial band-pass filter is illustrated in Figure 1-2 (a). Multiple parameters, including the thickness of each layer, and period of the unit cell, dimensions of the notches, were considered and tuned to achieve the target performance with the aid of a Genetic Algorithm (GA) $^{27}$ (see Appendix A-1) coupled with a full-wave electromagnetic solver. The subwavelength air-hole pattern was defined by electron-beam lithography followed by Inductively Coupled Plasma Reactive Ion Etching (ICP-RIE) through a deposited, three-layer Au-Polyimide-Au stack. The fabrication result was verified by Field Emission Scanning Electron Microscopy (FESEM), and the scattering properties were measured by Fourier Transform Infrared (FTIR) spectroscopy at normal incidence, as shown in Figure 1-2 (b) and (c). Figure 1-2 (c) confirms that the fabricated device meets the design specifications, functioning as a filter with high transmission and low reflection within the 3.0 µm to 3.5 µm band. This IR metamaterial filter was
properly designed by this iterative strategy and precisely fabricated using the conventional top-down methods.

Metamaterials composed of metal and dielectric multilayer nanostructures as described above have been investigated intensively over the past decade. However, the inherent absorptive loss of metal at optical frequencies and the difficulties of fabricating complex geometries at nanoscale limit the prospect of fully three-dimensional metamaterials. Recently, optical metamaterials containing two-dimensional layered nanostructures, instead of three-dimensional bulk nanostructures, have attracted much attention due to their reduced dimensionality and potential to address issues such as fabrication complexity and integration challenges. Plasmonic and dielectric metamaterials with single-layer nanostructures have been studied to control propagation and modify the amplitude and phase of electromagnetic waves. In Chapters 2, 3 and 4, single-layer subwavelength nanostructures are optimized to meet three desired optical responses, respectively. Chapter 2 demonstrates a broadband absorber with near-ideal absorptivity over ±45° incident angles at mid-infrared wavelengths, using a single metallic screen layer; in Chapter 3, wave-plates with above 92% polarization conversion ratio and reflectivity from visible to near-infrared wavelength ranges are achieved with strongly coupled nanorod arrays; Chapter 4 presents a perfect magnetic mirror with near-zero reflection phase at 1µm that is investigated by a cross-shaped a-Si nanoresonator array.

Metamaterials with tailored geometries can be precisely fabricated by conventional top-down approaches. Specifically, the submicron feature geometries were defined using electron-beam lithography, and these patterns were transferred onto the deposited metal and dielectric materials using reactive ion etching or lift-off process. However, the range of materials and nanostructures that can be realized using these approaches is limited. For instance, nano-spheres are usually approximated by nano-disks to fabricate, and it is difficult to integrate core-shell
structures through this technique. Therefore, new integration techniques must be developed to increase the palette of materials and nanostructures that can be experimentally accessed. Recently, Stebe et al. proposed particle assembly as a novel approach to overcome these limitations 47. Although self-assembled optical metamaterials have been reported 48-53, this approach solely forms a monolayer with simple particle structures or short-range ordering and provides less freedom to control the position and the orientation of the particles over macroscopic areas. This lack of control is problematic because both position and orientation are important factors in the construction of optical metamaterial devices. Here, a deterministic assembly method was developed to position nanoparticles. This bottom-up technique can be used to manipulate nanoparticles over large areas with fast response times, providing the potential to construct tunable and/or anisotropic optical metamaterial devices.

The use of electric field gradients and forces to deterministically assemble arrays of presynthesized subwavelength resonators that are registered to predefined features on a substrate provides a unique opportunity to construct complex optical structures that are not accessible using conventional top-down fabrication methods. The behavior of particles in solution is subject to the combination of gravitational forces and Brownian motion, as well as electrokinetic forces if the solution is subject to an electric field. The electrokinetic forces include contributions from several different factors, for instance, dielectrophoresis (DEP), electro-osmosis (EO), and electro-thermal (ET) effects. EO effects caused by the surface charge movement under the electric field usually occur at low frequency, and ET effects are typically limited to solutions with high electrolyte conductivities or high-frequency electric fields 54. Here, only DEP is considered because both EO and ET are related to the solution flow, which lacks particle/material selectivity, and proper electric field frequencies are chosen in this research to minimize these electro-hydrodynamic effects.

DEP occurs when neutral particles in solution are subjected to a non-uniform electric field 54-57. The particles and the solution will be polarized in the electric field, and the origin of the DEP
force can be understood from a simple model of a nanowire in a non-uniform electric field, as shown in Figure 1-3. The induced charge on the neutral, polarized nanowire is assumed as $q_+$ and $q_-$ on the two ends, respectively, and the net force on the nanowire can be expressed as:

$$\vec{F}(\vec{r}) = q_+ \vec{E}(\vec{r} + \frac{1}{2} \frac{\vec{p}}{|\vec{p}|}) + q_- \vec{E}(\vec{r} - \frac{1}{2} \frac{\vec{p}}{|\vec{p}|}) \tag{1-1}$$

where $l$ is the length of the nanowire, $\frac{\vec{p}}{|\vec{p}|}$ is a unit vector pointing along the axis of the wire, and $\vec{r}$ represents the spatial coordinates of the center point of the nanowire. As shown in Figure 1-3,
the imbalanced electric field forces exerted on the two ends of the electric dipole will drive the particle either to the region of the highest or the lowest electric field strength. When the length of nanowire is much smaller compared to the varying electric field, Equation (1-1) can be expressed as:\(^54\):

\[
\vec{F} = C \varepsilon_1 \text{Re}(K_f) \nabla |\vec{E}|^2
\]  

(1-2)

where \(C\) is a geometry related constant, \(\varepsilon_1\) is the real part of the permittivity of the solution, and \(K_f\), which is referred to as the Clausius-Mossotti factor, is the polarization factor that depends on the complex permittivity of the particle and the solution. \(K_f\) and the complex permittivity can be expressed as:

\[
K_f = \frac{\varepsilon_2 - \varepsilon_1}{\varepsilon_2 + 2\varepsilon_1}
\]  

(1-3a)

\[
\varepsilon_i = \varepsilon_i + j\frac{\sigma_i}{\omega}
\]  

(1-3b)

where \(\varepsilon_2\) is the complex permittivity of the particles, and \(\varepsilon_1\) is complex permittivity of the surrounding medium. If the particle is more polarizable than the surrounding medium, positive DEP will occur, and the particle will move to the highest \(\nabla |\vec{E}|^2\) region. In contrast, if the particles are less polarizable than the solution, negative DEP will occur, and the particles will move toward the lowest \(\nabla |\vec{E}|^2\) region.

A more accurate force calculation can be done by integrating Maxwell stress tensor (MST) on the surface of the object \(^58,59\). The MST and electrostatic force per unit volume can be expressed as:
\[ \sigma_{ij} = \varepsilon E_i E_j - \frac{1}{2} \varepsilon E^2 \delta_{ij} \]  

(1-4a)

\[ \vec{f} = \nabla \cdot \sigma \]  

(1-4b)

where \( \sigma_{ij} \) is the Maxwell stress tensor, \( \varepsilon \) is the material permittivity, \( E \) is the electric field, \( \delta_{ij} \) is Kronecker’s delta, and \( \vec{f} \) is force per unit volume. According to the divergence theorem, the total force acting on the particle can be obtained by integrating the \( \vec{f} \) over the volume or MST over the surface of the object:

\[ \vec{F} = \iiint \vec{f} \, dv = \iint \sigma \, ds \]  

(1-5)

Figure 1-4 (c) is the force distribution calculated by MST integration in COMSOL Multiphysics of a metallic nanowire in a non-uniform electric field generated by a parallel electrode structure, illustrated in Figure 1-4 (a). In this example, a voltage of 2V is applied across a 6 \( \mu \)m electrode gap, and the metallic nanowire is 10 \( \mu \)m long with a diameter of 250 nm. Here, the maximum force exists near the gap where the nanoparticles assemble, which matches the experimental result, as shown in Figure 1-4 (b).
Figure 1-4. (a) Schematic of the parallel electrodes with an applied electric voltage. (b) Uniformly spaced Rh nanowire array. (c) Force distribution calculated by MST with COMSOL and MATLAB.
Figure 1-5 summarizes the components needed to complete a directed self-assembly (DSA) system. According to this diagram, the required components are a non-uniform electric field determined by a predefined pattern, a way to apply the electric field, and a colloid containing the dispersed particles and a solution, as highlighted by the red box with a solid line in Figure 1-5. The variety of materials and geometric types of particles and the designed distribution of the non-uniform electric field, as highlighted by the dashed yellow lines, provide a versatile approach to achieve complex and reconfigurable optical systems. In Chapter 5, metallic nanowires dispersed in water with an in-plane applied electric field and stem bulb electrodes are demonstrated to achieve a tunable two-dimensional metallic lattice using this DSA system.

1.2 Overview

This dissertation introduces novel approaches to create a variety of metamaterials for IR and optical wavelength applications. This section provides an overview of the concepts covered in each of the following chapters, along with a brief summary.
In Chapter 2, a metamaterial absorber with near-ideal absorption over a super-octave bandwidth was demonstrated with an optimized single-layer Pd nanostructure array. An average 98% absorptivity was achieved between 1.77 and 4.81μm over a wide ±45° field-of-view by exciting multiple electromagnetic resonances generated by the nanostructure elements. Both normal and off-normal performance of the design were verified by fabricating the nanostructures and measuring its optical response with a Fourier transform infrared (FTIR) spectrometer. The design with an Au-based metamaterial absorber was also investigated and compared. The results revealed that Pd produces a design that was robust against process variation and provided fabrication reproducibility.

Chapter 3 discusses plasmonic metasurface-based nanostructures that achieve high polarization conversion efficiency with designed phase modulation for two orthogonally polarized incident light over a broad bandwidth and wide field-of-view. These anisotropic optical responses were obtained by utilizing strongly coupled nanorod resonators and enhanced by the interference of light through the multilayer Au/SiO₂/Au structure at subwavelength scale. Both half-wave and quarter-wave plates were demonstrated from 640 to 1290 nm and characterized by collecting the reflection and status of polarization with a custom-built optical setup.

Chapter 4 presents a lossless all-dielectric metamaterial composite with an α-Si nanoresonator array to obtain a perfect magnetic conducting surface and achieve 99.5% reflectivity with near-zero reflection phase at 1μm. This was achieved by exciting the primarily magnetic TM₀₁δ resonant mode in a double periodic array of cross-shaped α-Si nanoresonators. The designed optical magnetic mirror was carefully fabricated to reproduce the designed pattern, and its performance was verified by measuring its reflectivity amplitude and phase with spectral holography.

As an alternative to top-down fabrication, in Chapter 5 a bottom-up technique with electric-field assisted, directed self-assembly was developed and utilized to form reconfigurable two-dimensional nanowire lattices. A dense 2D lattice with “running bond” brickwork-like patterns
formed by thin SiO$_2$-coated solid Au wires with the long axis aligned parallel to the electric field spanning the electrodes was investigated, and the lattice periodicity could be controlled with the applied AC frequency. Electric field simulations were performed to understand the formation of the lattice and its tunability dependence on the assembly conditions.

Chapter 6 summarizes the accomplishments and contributions in each chapter and proposes possible directions of future research. Based on the electric-field assisted DSA technique, several assembly systems are proposed with preliminary results with the aim to realize more sophisticated and higher-performance optical devices.
1.3 References

Chapter 2

Near-Ideal Optical Metamaterial Absorber with Super-Octave Bandwidth

This chapter presents the design, fabrication and characterization of a broadband, polarization-insensitive metamaterial with greater than 98% measured average absorptivity that is maintained over a wide ±45° field-of-view for mid-infrared wavelengths between 1.77 and 4.81 µm. The nearly ideal absorption is realized by using a genetic algorithm (GA) to identify the geometry of a single-layer metal nanostructure array that excites multiple overlapping electric resonances with high optical loss across greater than an octave bandwidth. The response is optimized by substituting palladium for gold to increase the infrared metallic loss and by introducing a dielectric superstrate to suppress reflection over the entire band. This research was enabled through the collaboration of theoretical modeling, including electromagnetic full-wave simulations and GA synthesis, performed by Dr. Jeremy A. Bossard under the guidance of Dr. Douglas H. Werner.

2.1 Background and Motivation

Electromagnetic absorbers have wide applicability across the radio frequency (RF) and optical regimes, including such diverse uses as signature control, spectroscopy, solar cell enhancement, thermal imaging, and emissivity control. Among the electromagnetic structures that have been investigated, metamaterials composed of nanoscale resonators with customized electric and/or magnetic response have emerged as compelling candidates for optical-wavelength absorbers, with recent efforts focused on mid-infrared (IR) structures having multiband and wide field-of-view (FOV) properties. Nearly ideal black-body absorbers have been
demonstrated for the visible and mid-infrared (IR) spectral regimes using non-resonant nanomaterial structures, including metal-dielectric composites and vertically aligned single-wall carbon nanotubes. However, these solutions offer limited flexibility to tailor the wavelength range, polarization, and/or angular response of the broadband absorptivity. Metamaterial design approaches can be used to create broadband optical absorbers with such advanced functionalities, but progress toward this goal has been limited by the narrow bandwidth of the individual EM resonator elements and the challenge of impedance matching over a wide spectral range.

Electromagnetic absorbers rely on electric or magnetic losses in the constituent material to convert an electromagnetic wave into heat. One promising design approach considers the entire metamaterial to be an effective medium and uses nanoscale resonator elements to introduce large imaginary parts in the effective permittivity and/or permeability dispersion of the homogenized medium. The imaginary dispersive properties result in high absorption over a narrow band around resonance as the wave passes through the medium. Multiple closely spaced resonances can be realized by incorporating discrete resonators of different sizes into a single unit cell period. The number of different resonator types that can be patterned at dimensions compatible with IR operation is limited, which restricts the absorption bandwidth that can be obtained. It has also been proposed that bandwidth can be increased by integrating many nanoscale resonators into a unit cell by stacking them on top of each other. However, fabricating multiple aligned nanoscale features is complex and time consuming, and the proposed structures are only strongly absorbing for a single polarization.

Metamaterial absorbers (MMAs) based on electromagnetic band-gap (EBG) surfaces offer a compelling alternative with more straightforward nanofabrication. EBG absorbers employ a single lossy metallic screen backed by dielectric and ground layers to form a resonant cavity that couples with incident electromagnetic waves. Narrow single-band and multi-band THz and IR EBG MMAs have been demonstrated with near unity absorption, polarization independence, and
Efforts have been made to extend this design concept to create IR absorbers with wide and controlled bandwidths by combining resonators of different sizes in the same metallic screen, but achieving high absorption over an octave bandwidth in the IR has so far remained an elusive goal.

In this research, an EBG-based MMA that provides near-ideal spectral absorptivity across a broad mid-IR band with well-defined band-edge wavelengths is investigated. A genetic algorithm is employed to identify a single-layer nanostructured metallic screen geometry that supports multiple overlapping resonances to cover greater than an octave bandwidth. A dielectric superstrate layer is introduced to improve the impedance match to free space, thereby minimizing reflectivity and maximizing absorptivity across the band. The traditional gold (Au) metal is replaced by palladium (Pd) to broaden the MMA bandwidth and to improve fabrication reproducibility. Measurements of the optimized EBG-MMA show strong agreement with simulations, demonstrating a 98% average spectral absorptivity over a predefined 1.77 µm to 4.81 µm wavelength band and ±45º angular range. This work represents a significant step toward realizing practical optical MMAs that provide near-unity broadband absorption over a specified wavelength range, enabling the realization of customized IR black bodies and sources.

2.2 Design and Optimization

2.2.1 Electromagnetic Optimization Methodology

The structure for the broad-band EBG-based MMA consists of a four-layer metallodielectric stack as shown in Figure 2-1 (a). The bottom three layers of the structure create the resonant electromagnetic cavity of the absorber, which is composed of a doubly periodic array of metal nanostructures patterned on a thin dielectric layer that is backed by a solid metal ground
plane. A second dielectric layer is included on top of the structure to provide an impedance match between the metallic nanostructures and free space. This three-layer resonant electromagnetic cavity can be optimized to produce multiple overlapping resonances over a broad bandwidth in the near-to-mid IR range. In addition, the metal ground plane prevents light transmission through the MMA, thereby causing the incident light to be either reflected or absorbed. At each electric or magnetic resonance, high currents are generated in different regions of the metallic nanostructure array, resulting in strong absorption due to metallic loss.

Figure 2-1. Diagram of the MMA. (a) Cross-sectional view of the four-layer EBG-based broadband absorber. (b) Schematic of the Pd-based MMA structure optimized to have broadband absorption from 2µm to 5µm in the mid-IR. (c) Top view of the nanostructure unit.

A robust GA optimizer is employed to identify a metallic nanostructure array that simultaneously maximizes absorption over a broad bandwidth while also giving a wide-FOV and polarization-independent response. The GA is a stochastic optimizer that evolves an initial population of randomly generated candidate nanostructures based on the principles of natural selection. Each population member is a binary string, called a chromosome, which contains the
encoded design parameters. This optimization approach affords great flexibility to access complex and non-intuitive nanostructure features that fall well outside the bounds of conventional resonant elements such as crosses and rings.\textsuperscript{2-3,9} The design methodology exploits this flexibility to find elements that can support multiple resonances across the wavelength range of interest, thereby enabling MMAs with extremely wide absorption bandwidths.

A MMA with a minimum absorptivity of greater than 90% over an angular range of ±40° and more than an octave of bandwidth across the 2 to 5 µm mid-IR spectral range was demonstrated. For this work, structures were optimized using two different noble metals with low reactivity, Au and Pd, for both the ground plane and patterned nanoresonator layer. Polyimide was selected for both dielectric layers because of its non-dispersive optical properties over the target wavelength range. The optical properties of all of the constituent materials used for GA optimization and electromagnetic simulation were characterized by variable angle spectroscopic ellipsometry (J.A. Woollam IR-VASE). Most of the previous single- and multi-band mid-IR absorbers were based on Au resonant structures;\textsuperscript{19,21,26,33} thus Au was investigated for broadband MMAs as well. Recently, a single-band absorber with exposed Pd nanostructures was used for highly sensitive hydrogen gas detection due to the hydrogen absorption properties of Pd metal.\textsuperscript{35} Compared to the more commonly used Au metal, Pd with higher optical loss provides significant advantages for robust and reproducible fabrication of broadband EBG-based MMA. Consequently, the optimized absorber design that satisfied the target properties incorporated Pd rather than Au resonant elements. The nanofabricated absorbers were characterized in nitrogen (N\textsubscript{2}), which mitigated optical property changes due to environmental exposure. Practical applications would be enabled by replacing the polyimide layers with an alternate dielectric such as silicon nitride (Si\textsubscript{3}N\textsubscript{4}), which forms an excellent diffusion barrier against hydrogen, oxygen, and moisture.\textsuperscript{36} In addition, the costly noble metals used in this work could be replaced with less expensive metals having similar optical properties, such as nickel (Ni), lead (Pb), or tungsten (W).
The GA evolved all of the adjustable design parameters within predefined ranges to identify a four-layer MMA that meets the target optical properties, including the thickness of each layer, the unit cell period, and the patterned metal feature geometry. The metal ground plane layer thickness was fixed at 100 nm, which is several times the skin depth for both Pd and Au. The MMA was optimized by encoding one unit cell of the nanostructure array as a 15×15 binary grid, where “1” represents a pixel with a metal feature and “0’ represents a pixel without a metal feature. The unit cell was forced to have 8-fold mirrored symmetry, which provides polarization independence. This also greatly reduces the computation time required to identify a structure that satisfies the target optical properties because only one triangular fold must be optimized by the GA.

In each optimization cycle, the optical properties of all candidate MMA unit cells in the population were evaluated with a full-wave electromagnetic solver and compared to the target properties. More specifically, the reflectance and transmittance at a set of test wavelengths and angles were calculated using a full-wave finite element-boundary integral (FEBI) technique. Because the metal ground layer is thick enough to fully attenuate the wave, the transmittance is zero and the absorptivity is the difference between unity and the reflectance. The deviation between the properties of the candidate unit cell and the target is evaluated according to the following Cost function:

\[
\text{Cost} = \sum_{\lambda_i} \sum_{(\theta,\phi_i)} (1.0 - A_{\text{TE}})^2 + (1.0 - A_{\text{TM}})^2,
\]

where \(A_{\text{TE}}\) and \(A_{\text{TM}}\) are the transverse electric (TE) and transverse magnetic (TM) polarized absorptivities, \(\lambda_i\) are the test wavelengths, and \((\theta,\phi_i)\) are the test incidence angles. By minimizing the Cost function in Eq. (2-1), the GA evolves the nanostructure pattern and layer thicknesses that have near unity absorption over all of the test wavelengths and incidence angles. For this specific mid-IR broadband MMA design, 16 test wavelengths in approximately equally spaced frequency intervals over the range from 2 µm to 5 µm were specified. The test incidence angles were chosen
to be \((\theta, \phi) = \{(0^\circ,0^\circ), (40^\circ,0^\circ), (40^\circ,45^\circ)\}\), covering a FOV up to \(\pm40^\circ\). Throughout the evolutionary process, structures that failed to meet predefined nanofabrication constraints were assigned a high cost and were eliminated from subsequent candidate populations. This ensured that the optimized structure could be fabricated without changes, which is essential for subsequent experimental verification of the broadband MMA response. Using a population of 24 chromosomes, the GA converged to a structure with minimized Cost in 86 generations.

### 2.2.2 Optimized Subwavelength Nanostructure Array for Broadband Absorption

An illustration of the GA-optimized broadband MMA and a detailed diagram of the metallic nanostructure unit cell are displayed in Figure 2-1. The optimized thicknesses are \(t_1 = 398\) nm and \(t_2 = 429\) nm for the top and bottom polyimide layers and 30 nm for the patterned Pd nanostructures. The unit cell period is \(a = 851\) nm with a pixel size of 57 nm. Three well-defined nanostructures, which obey the predefined fabrication constraints, are noted: (1) a cross dipole centered in the unit cell; (2) a large loop centered in the unit cell and interconnected between unit cells; and (3) four smaller, isolated loops centered at the corners of the unit cell. These nanostructure elements and the coupling between them support multiple electromagnetic resonances with narrowly spaced center wavelengths, which produce high absorption across the broad mid-IR band.

The simulated absorptivity of the MMA calculated from the scattering parameters by: 
\[
A = 1 - R,
\]
where \(R\) is the reflectance, is shown in Figure 2-2 for unpolarized, TE polarized, and TM polarized light. The normal incidence spectra in Figure 2-2 (a) demonstrate that the minimum
spectral absorptivity for this structure is greater than 90% over the entire 1.90 \( \mu m \) to 5.47 \( \mu m \) range, achieving a high average value of 98.8% across this band. It is evident from these spectra that the broadband absorption is due to the convolution of multiple strong resonances positioned at optimized wavelengths across the band; the peak values of absorptivity at normal incidence occur at 2.13 \( \mu m \), 3.00 \( \mu m \), 3.70 \( \mu m \), and 5.05 \( \mu m \). Figure 2-2 (b-d) show 2D contour plots of the predicted angular dependence of the absorptivity from normal \( \theta = 0^\circ \) up to \( \theta = 89^\circ \) off-normal incidence for all polarizations. These simulations reveal that more than an octave bandwidth with minimum absorptivity of 89.5% and an average absorptivity of 94.7% is maintained over the 2 \( \mu m \) to 5 \( \mu m \) band at an incidence angle of \( \theta = 55^\circ \), which is wider than the targeted FOV used for optimization.
Comparing the TE and TM responses, the TM absorption at 5 µm drops off more quickly with increasing incidence angle as compared with the TE polarized response, whereas the dip in absorptivity around 4.5 µm becomes more pronounced for TE polarization at large incidence angles. These small changes in MMA response with increasing incidence angle are associated with slight shifts in peak position, strength, and bandwidth of the dominant resonances found at normal incidence.

### 2.2.3 Resonant Electromagnetic Properties

To understand the contribution of the different subwavelength nanostructures to the broadband absorption, the electric volume currents excited within the MMA were simulated for normally incident, TE polarized illumination in the mid-IR range. Figure 2-3 and Figure 2-4 show top-views and cross-sectional views of the current distribution at resonant wavelengths within the band where absorption is highest (2.13 µm, 3.00 µm, 3.70 µm, and 5.05 µm) and at one wavelength outside the band (1.50 µm) where reflection dominates. Outside the absorption band at 1.50 µm, the electric current in the MMA is negligible. In contrast, large electric currents are found on different parts of the Pd nanostructure array at wavelengths corresponding to peak in-band absorption. Specifically, the central crosses support the highest current at the shortest 2.13 µm wavelength. The current distribution spreads from the central cross to include the large and small loops at the intermediate 3.00 µm and 3.70 µm wavelengths. At the longest 5.05 µm wavelength, the electric current is high on all parts of nanostructure except the central cross.
Figure 2-3. Top view of finite-element method simulations of the current distributions in the top Pd screen for normal incidence at 1.50 µm and peak wavelengths of 2.13 µm, 3.00 µm, 3.70 µm, and 5.05 µm.
Compared to other MMAs that excite magnetic resonances,\textsuperscript{21} the strong broadband absorption in this MMA is entirely due to electric resonances. The cross-sectional views of the electric volume currents show that large currents are only present on the Pd nanostructure layer, and not on the ground plane. This occurs because the relatively thick polyimide substrate layer prevents strong coupling between the patterned nanostructure and the ground layer, which inhibits loop currents associated with a magnetic response. These simulation results demonstrate that the

Figure 2-4. Cross-section view of finite-element method simulations of the current distributions for normal incidence at 1.50 µm and peak wavelengths of 2.13 µm, 3.00 µm, 3.70 µm, and 5.05 µm.
broadband performance of this mid-IR EBG-MMA is achieved by exciting multiple closely wavelength-spaced electric resonances on the single-layer nanostructure array.

2.3 Nanofabrication and Optical Measurements

Nanofabrication of the MMA began by depositing 100 nm Pd and 10nm Ti on a Si wafer by electron-beam evaporation (Kurt J Lesker Lab18) at a rate of 1.4 Å/s. The 429 nm thick dielectric substrate layer was prepared by spin coating the polyimide precursor (HD Microsystems PI2556 diluted with T9039 thinner) at 1850 rpm for 40 seconds and soft baking at 120°C for 1 min. The film was cured in a N₂-purged oven for 2 hours at 220°C to fully remove the solvent and imidize the dielectric film. Following this step, the electron-beam resist (Nippon ZEP520A) diluted with anisole 1:1 was spun on the substrate at 3000 rpm for 50 seconds and baked at 180°C for 3 mins. The doubly periodic Pd nanostructure array (3 mm × 3 mm) was defined by electron-beam lithography (Vistec EBPG 5200) at a dose of 270 μC/cm² and developed using n-amyl-acetate for 3 mins and MIBK:IPA=8:1 for 1 min. The top 30 nm Pd was evaporated with a deposition rate of 1 Å/s and lift off by dissolving the resist (Rohm & Haas Microposit Remover 1165). The 398 nm thick dielectric superstrate layer was deposited by spinning the polyimide precursor at 2100 rpm for 40 secs. The structure was soft baked and cured using the same conditions. The thicknesses of the patterned Pd nanostructures were measured by atomic force microscopy (AFM) before depositing the top polyimide layer (AFM, Bruker Icon).

Field emission scanning electron microscope (FESEM) images taken immediately after the Pd nanostructures were defined on the polyimide substrate are shown in Figure 2-5. These images confirm that the complex array features were reproduced in the fabricated MMA sample. The measured unit cell period of $a = 860$ nm closely matched the target $a = 851$ nm. In comparison to the square 57 nm pixels in the design, the fabricated Pd nanostructures had rounded edges, with a
central dimension of ~75 nm. Although this process is compatible with creating a flexible MMA conformable to curved surfaces, the structure was left on the handle wafer for optical characterization.

Figure 2-5. FESEM image of the nanofabricated nanostructures.
The EBG-MMA was characterized using a Fourier transform infrared (FTIR) spectrometer (Brucker Optics IFS-66) equipped with a liquid nitrogen cooled mercury cadmium telluride (MCT) detector. A custom designed set-up was used for normal incidence reflection measurements as shown in Figure 2-6 (a). The measured reflectance was referenced to a Au mirror to determine the absolute reflectivity, \( R \). The absorptivity was determined according to \( A=1-R-T \), where \( T = 0 \). The normal incidence absorptivity plotted in Figure 2-6 (b) shows that minimum absorption remains above 90% independent of polarization at wavelengths between 1.77 \( \mu m \) and 4.81 \( \mu m \) (greater than an octave bandwidth), with an average absorption of 98.1% over this same range. Compared to the full-wave simulation, the bandwidth is reduced from 3.57 \( \mu m \) to 3.04 \( \mu m \) primarily because of the blue shift in the long-wavelength band edge from 5.47 \( \mu m \) to 4.81 \( \mu m \). This is associated with a reduction in the number of absorption maxima from four to three, and a shift in the peaks to 1.97 \( \mu m \), 3.09 \( \mu m \), and 4.23 \( \mu m \). The oblique incidence measurements were made using a specular reflection accessory (Thermo Spectra-tech Model 500) as shown in Figure 2-7 (a). Figure 2-7 (b-d) display 2D contour plots of the measured oblique incidence absorptivity for unpolarized, TE and TM light. These plots show that a full octave of bandwidth with higher than 90% absorptivity is maintained all the way out to incident angles of 55°. In addition, the trends in the angular dependence of the polarized response follow theoretical predictions.
Figure 2-6. (a) Illustration of normal incidence reflection measurement setup. M: mirror, BS: beam splitter, S: sample. (b) Measured absorptivity for unpolarized, TE and TM illumination at normal incidence and compared with simulation result.
These results demonstrate that the nanofabricated broadband MMA achieves the high and wide-FOV in-band absorption expected from simulation of the ideal optimized structure. The primary discrepancy between experiment and theory is the shift in the long-wavelength response, which results in a slight reduction in the bandwidth of the structure. The potential sources of this discrepancy include: (1) an increase in the Pd nanostructure feature size; (2) error in the polyimide superstrate and substrate thickness; and (3) differences in the dispersive properties ($n$ and $k$) of the evaporated Pd film following thermal processing. To determine the most likely causes, the optical response of the MMA was resimulated using the fabricated Pd nanostructure feature dimensions as

Figure 2-7. (a) Illustration of off-normal incidence reflection measurement setup for characterizing the fabricated sample. Contour plot of measured absorptivity as a function of wavelength and angle of incident from normal up to 55° incidence under (b) unpolarize (c) TE and (d) TM illumination.
well as different polyimide thicknesses and plotted in Figure 2-8 (b). These simulations reveal that the feature size modifies the response of the central resonances, but has little impact on the short or long-wavelength response. In contrast, decreasing both the superstrate and substrate polyimide thicknesses by only 10% provides a match to the short-wavelength band edge and better agreement with the long-wavelength band edge. Therefore, the discrepancy is largely due to errors in the polyimide thickness combined with small changes in the thermally processed Pd optical constants.

Figure 2-8. (a) Top: FESEM image of unit cell for the fabricated sample of Pd based MMA, Scale bar is 200 nm. Bottom: Unit cell with 10 nm increase in Pd features and corner rounding to match the nanofabricated structure in the FESEM image. (b) Simulation at normal incidence under unpolarized illumination to study the sensitivity of the MMA to nanostructure feature size changes and polyimide thickness variations.
2.4 Selection of Metal for Broadband Absorption

During this work, the tradeoffs of using Au versus Pd metal for the nanostructure array and ground layers in the four-layer broadband MMA were also evaluated. Figure 2-9 shows a Au-based broadband MMA that was optimized using the Cost function given in Eq. (2-1). Similar to the Pd-based MMA in Figure 2-1, this design provides greater than 90% absorptivity over a band from 1.90 \( \mu \)m to 5.25 \( \mu \)m and a wide-FOV of ±40º. The optimized thicknesses are \( t_1 = 423 \) nm and \( t_2 = 456 \) nm for the polyimide superstrate and substrate layers and 10 nm for the patterned Au nanostructures. The unit cell period is \( a = 998 \) nm with a pixel size of 67 nm. Because Au is more conductive than Pd in the mid-IR, the optimized Au nanostructure film thickness that provides a good impedance match over such a large bandwidth is only 10 nm as compared to 30 nm for the Pd nanostructure films. This makes the optical properties of the Au-based MMA more sensitive to small variations in metal thickness.

To investigate this Au-based structure experimentally, the optimized MMA shown in Figures 2-9 (a) with average Au nanostructure film thicknesses that varied from 10 to 12 nm in increments of ~1 nm were fabricated. The Au-based MMAs were fabricated using the same process flow as Pd-based MMAs, and the top 10 to 12 nm thick Au films were deposited at a rate of 0.5 Å/s. For the thinnest Au films that were not continuous, the effective film thickness was estimated by averaging AFM height measurements from at least five different locations on the sample. The measured absorptivity of the three MMAs is compared in Figure 2-9 (c). The MMA with the 10 nm thick Au features maintains a high absorptivity greater than 90% over a broad band between 1.95 \( \mu \)m and 4.80 \( \mu \)m.
Figure 2-9. (a) Diagram of the optimized Au-based MMA structure. (b) Top left: top view of one unit cell of the design. Bottom left: FESEM image of a unit cell of the fabricated structure, Scale bar is 200 nm. Right: Low-magnification FESEM image of the same structure. Scale bar is 600 nm. (c) Simulation and measurements for Au-based MMA under unpolarized illumination at normal incidence. The average Au thicknesses of the Au nanostructures determined by atomic force microscopy measurements are 10 nm, 11 nm, and 12 nm. (d) Simulation and measurements for Pd-based MMAs with Pd nanostructure thicknesses of 30 nm and 40 nm under unpolarized illumination at normal incidence.
However, the structure has a lower average broadband absorptivity of 94.4% as compared to the optimized Pd-based MMA. Increasing the Au film thickness to 11 nm and 12 nm results in large fluctuations in spectral absorptivity and a reduction in absorption near the long wavelength band edge, giving a lower average absorptivity of 89.2% across the band. To determine the sensitivity of the MMA to small variations in the Au film thickness, optical response for Au nanostructure film thicknesses ranging from 10 to 14 nm in increments of 1 nm were resimulated and plotted in Figure 2-10. These simulations verify the trends observed in the experimental measurements. In contrast, the absorptivity of the optimized Pd-based MMA is relatively insensitive to Pd thickness, as shown in Figure 2-9 (d). In this case, an increase in Pd thickness from 30 nm to 40 nm had a negligible effect on the absorption with only a slight reduction in the long wavelength
band edge from 4.81 \( \mu \text{m} \) to 4.48 \( \mu \text{m} \). This underscores the importance of identifying constituent materials and nanostructure geometries that are robust against process variations for high-performance metamaterial designs.

### 2.5 Summary

In this chapter, a polarization-independent metaldielectric EBG-based MMA with an average absorptivity of 98% over greater than an octave bandwidth from 1.77 to 4.81 \( \mu \text{m} \) and a wide \( \pm 45^\circ \) FOV was studied. A robust GA design approach was employed to identify a doubly periodic metal nanostructure array that excites multiple closely spaced electric resonances for the broad absorption band. The optimized unit cell of the nanostructure array integrates three well-defined electromagnetic elements in a single metal screen layer that meets predefined fabrication constraints. Simulations showed that the incident light induces large electric currents on different parts of the metal nanostructure at wavelengths corresponding to peak in-band absorption, thereby confirming the importance of each of these elements as well as the coupling between them to the high absorption. Comparing designs optimized using Au and Pd nanostructures reveals that Pd improves the broadband optical properties and fabrication reproducibility. This demonstration confirms that a single-layer metal nanostructure array can produce nearly ideal broadband absorption with well-controlled band edge wavelengths, paving the way for a variety of highly customizable optical metamaterial absorbers with super-octave bandwidth.
2.6 References

Chapter 3
Broadband and Wide Field-of-view Plasmonic Metasurface-enabled Waveplates

This chapter demonstrates plasmonic metasurface-based nanostructures for high-efficiency, angle-insensitive polarization transformation over a broad octave-spanning bandwidth. The structures are realized by optimizing the anisotropic response of an array of strongly coupled nanorod resonators to tailor the interference of light at the subwavelength scale. Nanofabricated reflective half-wave and quarter-wave plates designed using this approach have measured polarization conversion ratios and reflection magnitudes greater than 92% over a broad wavelength range from 640 to 1290 nm and a wide field-of-view up to ±40°. This research was enabled through the collaboration with other research groups. Numerical simulation was performed by Dr. Zhihao Jiang under the guidance of Dr. Douglas H. Werner, and optical characterization was conducted by Dr. Ding Ma under the guidance of Dr. Zhiwen Liu.

3.1 Background and Motivation

Optical metasurfaces are a new class of quasi two-dimensional metamaterials composed of a single layer of metallic nanostructures that provide exceptional capabilities for manipulating light in an ultrathin, planar platform\textsuperscript{1-3}. In comparison to their three-dimensional counterparts, metasurfaces also exhibit reduced loss and fabrication complexity, making them attractive for integration in practical optical systems\textsuperscript{4}. By engineering the geometry of the nanostructured metasurfaces, the spectral and spatial dispersion of their optical response can be tailored to generate a specific abrupt interfacial phase change and cross-polarized response on a subwavelength scale.
Such unique optical properties have been exploited to demonstrate a variety of new physical phenomena and associated optical devices over the past few years, including anomalous reflection and refraction, optical wavefront manipulation, frequency-selective near-perfect absorption, spin-hall effect of light, spin-controlled photonics, polarization-dependent unidirectional surface plasmon polariton excitation, and metasurface holograms.

Optical waveplates that achieve broadband polarization conversion with a wide field-of-view (FOV) are highly desirable for systems that perform optical characterization, sensing, and communications functions. However, simultaneously achieving broadband and wide-angle properties is difficult using conventional multilayer stacks of birefringent materials because these structures rely on the dispersive properties of the birefringence. In contrast, metasurfaces provide a promising pathway towards broadband and wide-angle polarization conversion in a submicron-thick layer. A variety of optically thin, metasurface-based polarization-control components have been theoretically proposed and demonstrated, including various polarizers, near-field polarization shapers, and ultrathin waveplates.

Most metasurface-based waveplates have employed homogenous arrays of weakly coupled anisotropic resonant building blocks, including crossed nanodipoles and nanoslits, L-shaped or V-shaped nanoantennas, and elliptical nanoholes. These structures typically suffer from a narrow FOV, limited bandwidth, and/or low efficiency because their anisotropic optical response relies on the resonance of each isolated building block, where strong dispersion and impedance mismatch may exist. More recently, a near-infrared (IR) quarter-wave plate achieved broadband circular-to-linear polarized light conversion by tailoring the dispersive properties of an array of orthogonally coupled nanodipole elements. However, the waveplate had an average power efficiency of less than 50%. In addition, to obtain broadband circularly polarized transmitted light from this design, incident linearly polarized light with a wavelength-dependent electric field polarization direction was required. An alternative inhomogeneous metasurface design strategy has...
also been used to produce an ultrathin broadband quarter-wave plate\textsuperscript{34}. The demonstrated design achieves a power efficiency of only 10% over a narrow FOV, and has a wavelength-dependent directivity of the outgoing polarization.

In this research, wide-angle reflective plasmonic metasurface-based half-wave and quarter-wave plates that have a high polarization conversion efficiency and reflection magnitude over a broad bandwidth in the visible-to-near IR wavelength range are demonstrated. This is achieved by optimizing the anisotropic optical response of the metasurface, which is composed of an array of strongly coupled nanorod resonator building blocks with controlled aspect ratio, to tailor the spectral dispersion of light interference between the metasurface and a metallic reflector. In contrast to previously reported metasurface waveplates, the anisotropic optical response and light interference are controlled across the broad wavelength range and wide-FOV rather than merely near the resonant wavelength of the metasurface building blocks. Both the nanofabricated half-wave and quarter-wave plates achieved measured polarization conversion ratios (PCRs) higher than 92% over more than an octave bandwidth from 640 nm to 1290 nm with a wide field-of-view of $\pm 40^\circ$. This work represents a new state-of-the-art for optical metasurface-based devices, and will enable other types of ultrathin optical components with broad operational bandwidth, wide FOV and high efficiency.

3.2 Principle of Reflective Plasmonic Metasurface-based Waveplate

3.2.1 Structure and Operation of Anisotropic Metasurface

Figure 3-1(a) shows a simplified schematic of the metasurface-based waveplate that is used for simulation. The multilayer structure comprises a top homogeneous anisotropic
metasurface, an intermediate silicon dioxide (SiO$_2$) layer separated from the metasurface by a subwavelength air spacer, and a bottom continuous gold (Au) layer. The metasurface is characterized by uniaxial surface polarizability tensors $\mathbf{\tilde{\alpha}}$ and $\mathbf{\tilde{\alpha}}'$. Because the metasurface has no magnetic component and is modelled as an infinitely thin sheet, the surface polarizability tensors can be simplified to $\mathbf{\tilde{\alpha}} = \text{diag}[\alpha_x,\alpha_y,\alpha_z]$ and $\mathbf{\tilde{\alpha}}' = \text{diag}[0,0,\alpha_z]$. The air layers above and below the metasurface are included in the design to account for the small phase retardation present

Figure 3-1. Schematics of the proposed plasmonic metasurface-based waveplates. (a) 3D view of the multilayer structure model including an anisotropic homogenous metasurface located at a distance of $t_m/2$ above the SiO$_2$ layer. The metasurface is characterized by electric and magnetic surface polarizability tensors $\mathbf{\tilde{X}}_E$ and $\mathbf{\tilde{X}}_M$. Optically thin strongly-coupled nanorod array is used to realize the metasurface. (b) Interference model for evaluating the optical response of the multilayer structure based on the surface polarizability tensor parameters of the metasurface.
in the fabricated nanostructure that has a finite thickness of \( t_m \approx \frac{\lambda_0}{30} \). The value of the phase retardation can thus be approximated by \( e^{i k_0 t_m / 2} \).

The operating principle of the metasurface-based waveplate is illustrated in Figure 3-1(b), and resembles that of a modified Gires-Tournois (G-T) etalon\(^{36}\). Here, a plane wave with prescribed polarization illuminates the multilayer structure from an incident direction \((\theta_i, \varphi_i)\), where \( \theta_i \) denotes the angle between the \( z \)-axis and \( \vec{k}_i \), while \( \varphi_i \) is the angle formed by the \( x \)-axis and the projection of \( \vec{k}_i \) on the \( x-y \) plane. When the incident wave interacts with the multilayer structure, both \( s \)- and \( p \)-polarized reflected and transmitted waves are generated by the metasurface. The transmitted waves undergo multiple reflections between the metasurface and the bottom Au layer, where they interfere with one another to create the final reflected wave. Due to the anisotropy of the metasurface, both co-polarized and cross-polarized reflected waves are collected at the direction \((\theta_i, \varphi_i - 180°)\). The dielectric spacer and the Au ground plane increase the design degrees of freedom for controlling the amplitudes and phases of the waves produced by the multiple reflections, and thus can be optimized to increase the bandwidth of the anisotropic reflection from the three-layer nanostructure. The complex reflection coefficients that describe the polarization-dependent optical response of this structure \((r_{ss}, r_{sp} = r_{ps}, \text{ and } r_{pp})\) can be calculated analytically using an interference model\(^{37}\). By tailoring the dispersive anisotropy of the metasurface as well as the interference of the \( s \)- and \( p \)-polarized wave components, the polarization state of the final reflected wave is controlled over a broad spectral and wide angular range.

### 3.2.2 Anisotropic Optical Response of the Metasurface Nanorod Building Blocks

The desired anisotropic homogenous metasurface can be realized using a single-layer metallic nanoresonator array provided that its periodicity is much smaller than the wavelength of interest\(^3\). As shown in Figure 3-1 (a), a 2D array of Au nanorod building blocks with strong inter-
element coupling was employed because the dimensions and inter-element spacing of this simple geometry can be: (1) optimized to provide a widely varying anisotropic response, and (2) scaled to fabricate devices that operate at visible wavelengths. Before designing nanorod-based metasurfaces with the uniaxial surface polarizability properties required for the quarter-wave and half-wave plate devices, the optical properties of both isolated and strongly coupled pairs of Au nanorods were determined for a normally incident linearly polarized wave with its electric field either parallel or perpendicular to the long axis of the rod as shown in Figure 3-2 (a). The high frequency structure simulator (HFSS) finite element solver was used to perform all the full-wave simulations. For the scattering simulations of the single and coupled nanorod pair elements, the structures were placed in the center of an airbox whose outer edges were assigned a radiation boundary to prevent undesired reflections. The nanorod was oriented such that its long side coincides with the $x$-direction and its short side with the $y$-direction. Two linearly polarized plane waves, one $x$-polarized and the other $y$-polarized, were normally incident from the top surface of the nanorod elements. The scattered fields were evaluated at a distance of 3 $\mu$m below the bottom surface of the nanorod. The phase of the scattered fields was defined relative to that of the excitation when no nanorod elements are present.

Figure 3-2(b) plots the calculated amplitude (normalized) and phase of the scattered light from a single nanorod with a length $a_x = 250$ nm, a width-to-length aspect ratio $a_y/a_x = 0.3$, and a thickness $t_m = 30$ nm as well as a strongly coupled pair of nanorods with an inter-element gap $g = 20$ nm. The electric fields are concentrated at the ends of the isolated nanorod as shown in Figure 3-2(c), resulting in an $x$-directed dipolar resonance at 930 nm with a scattering phase of around 90°. The localized surface plasmon polariton (LSPP) mode causes the nanorod to resonate at a wavelength much larger than twice its length. In contrast, for a pair of strongly coupled nanorods, the enhanced electric fields are effectively confined within the air gap between the two rods as
Figure 3-2. Anisotropic scattering of single and coupled nanorod pair. (a) Schematics of an incident \(x-\) and \(y-\)polarized wave at normal incidence illuminating a single or a coupled nanorod. The dimensions are \(a_x = 250, a_y = 75, t_m = 30, g = 20\) (all in nm). (b) Scattering field magnitudes (normalized) and phases of the single (top) and twin (bottom) nanorod. (c) Electric field distribution on the single nanorod at 930 and 550 nm and electric field distribution on the coupled nanorod at 1125 and 550 nm. (d) Dispersion of \(\Delta \phi\) showing the impact of nanorod length \(a_x\), nanorod aspect ratio \(a_y/a_x\), and gap size \(g\).

Illustrated in Figure 3-2 (c). The capacitance formed in the air gap between the nanorods is much larger than the capacitance at the two ends of each individual nanorod\(^{38}\), resulting in an \(x-\)directed
LSPP dipolar resonance occurring at 1125 nm, which is only 21% larger than the resonating wavelength of the single nanorod. This resonance also has a lower quality factor as demonstrated by the flatter phase response. For the mode corresponding to \( \gamma \)-polarized incident light, the single and paired nanorod elements exhibit a similar response. Specifically, both have a dipolar resonance at 550 nm with a weaker strength and a phase slightly larger than 90° due to the inertia of the electrons at high frequencies and the finite cross section of the nanorods\textsuperscript{39}.

For the nanorod structure in Figure 3-2 (a), the anisotropic scattering behavior causes a large phase difference over the broad wavelength region between the two orthogonally polarized resonating modes even at oblique angles of incidence, which provides a percentage bandwidth of 62% for \( \Delta \phi > 90° \) and 39% for \( \Delta \phi > 135° \). This is central to achieving broadband metasurface-enabled polarization transformations. To understand the effect of the nanorod structure on the phase of the scattered light, the phase difference as the length \( (a_x) \) and aspect ratio \( (a_y/a_x) \) of a single isolated rod varied with all other parameters remaining unchanged was evaluated. As shown in Figure 3-2 (d), increasing the length and decreasing the aspect ratio both broaden the bandwidth of large \( \Delta \phi \) region, which is indicative of the degree of scattering anisotropy. For the strongly coupled nanorod pair, the data in Figure 3-2 (d) also demonstrates that the bandwidth of \( \Delta \phi \) increases exponentially as the air gap size drops due to the large increase in air gap capacitance of the structure. It should be noted that even though the nanorod building block has a wavelength-dependent scattering magnitude, all of the incident waves will be reflected by the bottom solid Au layer. By designing the nanostructure such that it is operating away from its absorption band, the reflection magnitude for both polarizations will possess a flat response.

Most previously demonstrated plasmonic metasurfaces\textsuperscript{1-3} have been designed by optimizing the geometry of the single isolated resonant building block. This investigation reveals that strong inter-element coupling plays a critical role in controlling the scattering phase difference for achieving the tailored anisotropic optical dispersion required for broadband, low-loss
performance. And the bandwidth broadening due to inter-element coupling can be further enhanced when the strongly coupled nanorods are arranged in a 2D array.

3.3 Optimized Broadband and Wide-angle Half-wave Plate

3.3.1 Design and Optimization of a Half-wave Plate

Broadband and wide-angle plasmonic metasurface-based waveplates can be efficiently designed by using the interference model of the multilayer structure and the optical properties of the strongly coupled nanorod array. As a proof-of-concept, here a half-wave plate covering a wide-FOV up to ±40° in the wavelength range from 640 to 1290 nm using the three-layer Au-SiO$_2$-Au nanostructure shown in Figure 3-3 (a) was considered, which transforms an $s$-polarized incident wave from a propagation direction of ($\theta_i$, $\varphi_i = 135^\circ$) into a $p$-polarized reflected wave at a propagation direction of ($\theta_f$, $\varphi_f = -45^\circ$). During the design process, the effective polarizability tensors $\tilde{\chi}_E$ and $\tilde{\chi}_M$ of the anisotropic nanorod array metasurface were retrieved using the complex transmission and reflection coefficients for both polarizations at two angles of incidence. The extracted effective surface polarizabilities were then used in the interference model to calculate the complex reflection coefficients ($r_{ps}$, $r_{ss}$) of the entire nanostructure over a wide angular range. In contrast to most of the previous observations and explanations of multilayer metallodielectric nanostructures in which magnetic resonances are identified$^{40,41}$, here it is found that the near-field interaction between the nanorod array and the continuous Au ground layer is weak enough to be neglected. Consequently, the interference model, which is more efficient in terms of the computation time, can be applied even though the dielectric layer thickness is on a subwavelength scale$^{42}$. 
A powerful covariance matrix adaptation evaluationary strategy (CMA-ES) optimization technique\textsuperscript{43} (see Appendix B) was employed to identify the nanostructure dimensions that meet the challenging multi-objective half-wave plate design criteria, \textit{i.e.} highly-efficient polarization conversion spanning a wide spectral and angular range\textsuperscript{44}. Predefined constraints on the allowable unit cell size, nanorod length and width, as well as the thickness of metal and dielectric layers were incorporated into the optimization algorithm to avoid generating structures that are impractical to fabricate. Additionally, the optical properties of all of the constituent materials were characterized by spectroscopic ellipsometry (RC2® Ellipsometer, J.A. Woollam Co.) over the wavelength range of 240 to 1690 nm and were used in the electromagnetic simulations to minimize discrepancies between the theoretically predicted and experimentally fabricated nanostructure response\textsuperscript{45}.

For each design candidate, the calculated reflection coefficients for both polarizations ($r_{ps}$, $r_{ss}$) in the targeted wavelength and angular range are compared with the user-input-defined target values to determine the \textit{Cost}, which is expressed as:

$$Cost_{hwp} = \sum_{\lambda, \theta} [(|r_{ps}| - 1)^2 + |r_{ss}|^2],$$

where $\lambda$ and $\theta$ denote the wavelength and incident angles included in the optimization. For this work, wavelengths from 640 to 1290 nm in steps of 50 nm and angles from 0° to 40° in steps of 10° were selected. The CMA-ES evolved solutions until it converged on a three-layer metal-dielectric nanostructure with a sufficiently low overall \textit{Cost} value that achieved the desired half-wave plate optical properties. The optimized geometry of the structure that meets these design criteria is shown in Figure 3-3 (a-b). The Au nanorods have a length of 210 nm and a width of 70 nm, which corresponds to an aspect ratio of 0.33. The periodicity in both the $x$- and $y$-directions is 252 nm, giving an inter-element gap of 42 nm in the $x$-direction, which provides strong electromagnetic coupling to the neighboring nanorods. The thickness values for the top Au nanorod
array layer, the SiO$_2$ layer, and the bottom Au layer are 42 nm, 114 nm, and 100 nm, respectively, resulting in a structure that is only 256 nm thick.

The polarization conversion ratio (PCR) of the metasurface-based half-wave plate was calculated using the semi-analytical interference model, defined as:

$$PCR_{hwp} = \frac{|r_{ps}|^2}{(|r_{ps}|^2 + |r_{ss}|^2)}, \quad (3-2)$$

As shown by the theoretical curves plotted in Figure 3-3(d), the calculated $PCR_{hwp}$ is greater than 94% across the targeted broad wavelength and wide angular range, and remains above 90% over a wider band from 640 to 1400 nm. Throughout this spectral and angular range, the reflection magnitude is greater than 95%, thereby achieving a much higher power efficiency than previous plasmonic metamaterial and metasurface-based designs$^{3,4,23,33,34,46}$. Even for angles of incidence as large as 50°, the reflection magnitude of the nanostructure is maintained above 94% with a $PCR_{hwp}$ of greater than 83%. Notably, this example demonstrates that the bandwidth of the 2D array of strongly coupled nanorods is significantly wider than that of the freestanding pairs of nanorods as shown in Figure 3-2 (b).
Figure 3-3. Broadband and wide-angle plasmonic metasurface-based half-wave plate. (a) Tilted 3D view of the metasurface-based half-wave plate. An s-polarized wave incident from an angle of ($\theta_i, \phi_i=135^\circ$) is converted into a p-polarized wave upon reflection. Inset shows the side view of the nanostructure and light path. (b) Unit cell configuration of the optimized half-wave plate. The dimensions are $a_x = 210, a_y = 70, p_x = 252, p_y = 252, t_m = 42, t_d = 114, t_{m2} = 100$ (all in nm). (c) Top-view FESEM image of a portion of the fabricated nanostructure showing the nanorod array. Scale bar: 400 nm. The inset shows the magnified top view of two by three unit cells. Scale bar: 100 nm. (d) Theoretically predicted and experimentally measured polarization conversion ratio (PCR) and reflection magnitude (Refl. Mag.) as a function of wavelength at different angles of incidence ($4^\circ, 20^\circ, 40^\circ$). (e) Theoretically predicted and experimentally measured polarization state in the plane perpendicular to the wave vector at 700, 900, and 1150 nm for different angles of incidence ($4^\circ, 20^\circ, 40^\circ$).
For the optimized 2D nanorod array structure, at large incident angles, the in-band $PCR_{hwp}$ is limited by: (1) the higher quality factors of two resonant peaks that correspond to the $x$- and $y$-directed LSPP dipolar modes of the coupled Au nanorods; and (2) the increase in phase retardation in the dielectric spacer. As a result, in the spectral range bracketed by the two modes, the reflection phase difference between the $x$- and $y$-directed electric field components exceeds 180° for incident angles greater than 40°. This causes the observed decrease in the $PCR_{hwp}$ near the middle of the band between the resonant peaks, which restricts the FOV for > 90% $PCR_{hwp}$ to approximately ±40°.

The state of polarization (SOP) traces of the incident and reflected light are presented in Figure 3-3 (e) at several angles of incidence and wavelengths within the band. Clearly defined cosine-shaped patterns can be identified, indicating that the reflected wave possesses a high degree of linear polarization (DoLP). The SOP patterns of the incident light have their maxima along the −90°/90° directions, while the SOP patterns of the reflected light have their maxima along the 0°/180° directions. The 90° rotation over the broad spectral and wide angular range confirms that the optimized plasmonic metasurface-based half-wave plate indeed transforms a linearly polarized incident wave into a reflected wave with a cross polarization.

### 3.3.2 Experimental Realization of the Half-wave Plate

The three-layer metallodielectric structures were fabricated by first depositing a 10 nm Ti adhesion layer and the bottom 100 nm Au ground plane on a Si handle wafer by electron-beam evaporation (Kurt J Lesker Lab18) at a rate of 1.4Å/s. A second 1 nm Ti adhesion layer was deposited immediately before electron-beam evaporating 114 nm SiO$_2$ dielectric spacer at a rate of 1.4Å/s. The nanorod array pattern was defined by electron-beam lithography (Vistec EBPG
5200A). Specifically, a layer of ZEP520A positive electron-beam resist (ZEP520A, diluted with anisole 1 to 1) was spin coated on the SiO$_2$ dielectric layer at 3000 rpm for 50 sec, baked at 180 °C for 3 min, and exposed with a 100 keV electron beam at a dose of 170 μC/cm$^2$. After exposure, the nanorod features were developed in n-amyl-acetate for 3 min and MIBK:IPA=8:1 for 1 min. A 1 nm Ti adhesion layer and the top 42 nm Au layer were deposited at a rate of 1Å/s. The metal on top of the unexposed resist was lifted-off by dissolving the resist in Microposit 1165 Remover (Rohm & Haas). The thicknesses of the Au and SiO$_2$ layers were verified with Atomic Force Microscopy (AFM, Bruker Icon). FESEM images of the fabricated nanostructure are shown in Figure 3-3 (c). The measured dimensions of Au nanorod length, width, and minimum inter-element gap spacing are 210 ± 5 nm, 70 ± 3 nm, and 42 ± 4 nm. The lithographic process produces nanorods that have rounded corners, which results in effective electromagnetic gap spacing of 50 ± 2 nm.

3.3.3 Optical Characterization of the Half-wave Plate

The $P_{CR}$ and reflection magnitude of the fabricated structure was characterized using a custom-built optical setup as shown in Figure 3-4 that utilizes a supercontinuum source to illuminate the structure at incident angles of 4°, 20°, and 40°. The broadband supercontinuum source was generated by coupling a sub-nanosecond Q-switched pulsed laser into a 20 m long highly-nonlinear photonics crystal fiber (PCF). The strong peak in the power spectrum of the source at 1064 nm (the pump wavelength used to generate the supercontinuum) introduces significant measurement error; thus, data collected in the wavelength range between 1040 and 1080 nm are excluded from the plots. To characterize the plasmonic metasurface-based half-wave plate, an s-polarized beam was produced by a polarizing beam splitter and used as the incident light source. The polarization state of the reflected light was characterized by rotating a broadband analyzer in steps of 10° (Glan-Thompson Calcite Polarizer, Newport 10GT04) in the plane perpendicular to
the wave vector of the reflected light and detecting the transmitted power with an Optical Spectrum
Analyzer (Ando, OSA AQ-6315A).

The optical properties of the fabricated metasurface-based half-wave plate shown in Figure
3-3 (d-e) are in strong agreement with theoretical predictions. The measured $PCR_{hwp}$ and reflection
magnitude both remain above 92% over the targeted broad wavelength range from 640 to 1290 nm
and wide-FOV from 0° to 40°. Even at a larger 50° angle of incidence, the $PCR_{hwp}$ and reflection
magnitude are still 86% and 87%, respectively. In comparison to the design, the long-wavelength
cut-off for the 90% $PCR_{hwp}$ bandwidth at a 4° incident angle is blue-shifted from 1400 to 1330 nm
without a change in the short-wavelength cutoff. This small discrepancy is attributed to the wider
inter-rod gap spacing in the fabricated structure compared to the optimized design dimensions.
This is consistent with the trend found in the freestanding nanorod pair scattering analysis
in Figure 3-2 (c-d), which shows a blue shift in the $x$-directed LSPP dipolar resonance
without affecting the $y$-directed dipolar resonance as the gap increases.

Polarization conversion is experimentally verified by the measured SOP patterns shown in
Figure 3-3 (e). The cosine-shaped patterns confirm the high DoLP of the reflected light. In addition,
the angle between the maxima of the reflected and incident light patterns is 90° throughout the
entire wavelength band and 40° FOV, indicating that the maximum light is reflected from the
sample when the polarizers for the incident and reflected light are oriented orthogonally to each
other. Compared to the current experimentally demonstrated plasmonic metasurface-based
polarization-control devices, the nanorod array half-wave plate presented here achieves a high
power efficiency over a wide spectral and angular range.
3.4 Demonstration of a Metasurface-based Quarter-wave Plate

To show the versatility of the proposed platform and design approach, a metasurface-based quarter-wave plate was also optimized for the same spectral range of 640 to 1290 nm and wide-FOV from 0 to 40°. The three-layer structure shown in Figure 3-5 (a) transforms a circularly polarized incident wave from a direction (θ_i, φ_i = 180°) into a linearly polarized reflected wave at a direction (θ_i, φ_i = 0°), which provides an in-phase superposition of both the s- and p-polarizations. One unit cell of the optimized doubly periodic array is composed of 180 nm long and 70 nm wide Au nanorods that are centered within the 240 nm × 282 nm cell. This array structure
possesses a weaker degree of anisotropy compared to the half-wave plate because of the larger nanorod aspect ratio of 0.5 and wider inter-element gap of 60 nm. As plotted in Figure 3-5 (d), the simulated $PCR_{qwp}$ is greater than 90% over a wider than targeted wavelength range from 620 to 1500 nm for incident angles up to 50°. The reflection magnitude is greater than 93% across the targeted spectral and angular range except for a narrow dip near 680 nm. The SOP traces presented in Figure 3-5 (e) show that the nanostructure transforms the incident wave with a circular SOP pattern into a reflected wave with a cosine pattern that has a maximum along the 45°/225° direction.

Figure 3-5 (c) displays FESEM images of the nanofabricated quarter-wave plate. The measured dimensions of the Au nanorod length, width, and minimum inter-element gap spacing are 180 ± 6 nm, 90 ± 4 nm, and 60 ± 5 nm, which agree well with the design target. Similar to the half-wave plate, the rounded nanorod edges result in an effective gap of 68 ± 3 nm that is wider than the design target. During the characterization process, a commercial quarter-wave plate was used to generate circularly polarized incident light. As shown in Fig. 3-5 (d), the measured $PCR_{qwp}$ is greater than 91% within the targeted wavelength band and angular range, with the reflection magnitude higher than 92%. The reduced $PCR_{qwp}$ value in the long wavelength range can be attributed to the imperfect circular polarization of the incident wave above 1300 nm, which is outside of the operational band of the commercial quarter-wave plate. The measured SOP patterns are in strong agreement with the simulated predictions, indicating that the circularly polarized incident wave is converted to a linearly polarized reflected wave.
Figure 3-5. Broadband and wide-angle plasmonic metasurface-based quarter-wave plate. (a) Tilted 3D view of the metasurface-based quarter-wave plate. A circularly-polarized wave incident from an angle of $(\theta_i, \phi_i=0^\circ)$ is converted into a linearly-polarized wave upon reflection. Inset shows the polarization of the reflected and incident waves in the plane perpendicular to the wave vector. The angle between the electric field and the plane of incidence of the reflected light is $45^\circ$. (b) Unit cell configuration of the optimized quarter-wave plate. The dimensions are $a_x = 180$, $a_y = 90$, $p_x = 240$, $p_y = 282$, $t_m = 40$, $t_d = 150$, $t_m2 = 100$ (all in nm). (c) Top-view FESEM image of a portion of the fabricated nanostructure showing the nanorod array. Scale bar: 400 nm. The inset shows the magnified top view of two by three unit cells. Scale bar: 100 nm. (d) Theoretically predicted and experimentally measured polarization conversion ratio (PCR) and reflection magnitude (Refl. Mag.) as a function of wavelength at different angles of incidence $(4^\circ, 20^\circ, 40^\circ)$. (e) Theoretically predicted and experimentally measured polarization state in the plane perpendicular to the wave vector at 700, 900, and 1150 nm for different angles of incidence $(4^\circ, 20^\circ, 40^\circ)$. 
3.5 Summary

In conclusion, a versatile approach to create highly efficient, wide-FOV plasmonic metasurface-based waveplates that control the polarization response of light over more than an octave bandwidth from the visible to the near-IR was demonstrated. The ultrathin reflective waveplates are realized by tailoring the spectral phase properties of a strongly coupled anisotropic nanorod array in conjunction with the interference of light between the array and the ground plane. A half-wave plate and a quarter-wave plate were designed and fabricated, achieving a PCR and reflection magnitude higher than 92% over more than an octave bandwidth from 640 to 1290 nm within a wide field-of-view of ±40° for both devices. Beyond the two proof-of-concept waveplates, the general design approach presented in this work can be extended to nanostructured optical components that exhibit arbitrary polarization conversion properties, as well as other optical functionalities endowed by metasurfaces with engineered dispersion, over a broad wavelength and wide angular range.
3.6 References

Chapter 4

Dielectric Nanoresonator-Based Lossless Optical Perfect Magnetic Mirror with Near-Zero Reflection Phase

In this chapter, an all-dielectric lossless optical mirror for the realization of controllable reflection phase based on an array of isolated dielectric nanoresonators is presented. This dielectric mirror is comprised of a cross-shaped amorphous silicon nanoresonator array that has been designed to achieve a 99.8% reflectivity and zero reflection phase at the wavelength of 0.99 μm. The measured results from the fabricated sample match the theoretical predictions with 99.5% reflectivity and near-zero degree reflection phase at 1 μm, which is very close to the targeted wavelength. This concept and approach pave the way for synthesizing lossless artificial reflecting electromagnetic boundaries with arbitrary phase response and hold great promise in applications ranging from nanocavities to nanowaveguides and nanoantennas. This research was enabled through the collaboration with other research groups. Numerical simulations were conducted by Zhihao Jiang under the guidance of Dr. Douglas H. Werner, and spectral holography measurements were performed by Dr. Ding Ma under the guidance of Dr. Zhiwen Liu.

4.1 Background and Motivation

Optical devices that exhibit electromagnetic boundaries with near-unity reflectivity are often used as mirrors, which are critical components in almost any optical system. Conventional mirrors, which are made of noble metals, create a reflected electric field that is out-of-phase with respect to the incident field due to their near-perfect electric conducting characteristics. By virtue of duality, a boundary with a near-perfect magnetic conducting property gives rise to an in-phase
reflection, \textit{i.e.}, a near-zero reflection phase, resulting in an enhanced electric, rather than magnetic, field at/near the interface. Such in-phase reflecting mirrors, also referred to as perfect magnetic conducting (PMC) or high-impedance surfaces, have been shown to enable a wide range of useful devices, including miniaturized cavities, zero cutoff waveguides, low-profile electromagnetic radiators, electromagnetic band gap metasurfaces, and so on\textsuperscript{1-4}.

PMC surfaces have been extensively explored in the radio frequency range since they were first proposed in 1999 by Sievenpiper \textit{et al.}\textsuperscript{1} for various applications such as in microwave circuits and antennas\textsuperscript{2-4}. It was not until very recently that the PMC surface concept has garnered interest in the optical community for its potential applications in molecular spectroscopy, photodetectors, nanocavities, nanoantennas, and so on\textsuperscript{5, 6}. The previously reported designs of optical PMC surfaces have utilized metallic fish scale\textsuperscript{6-8} structures. However, due to their inherent asymmetrical geometry, such devices only operate for a linearly polarized incident wave with a particular azimuthal angle. At the resonant wavelength, the reported reflectivity is around 70\% due to relatively high absorption loss in the metal, while the minimum reflection phase reaches only about 30 degrees, far from the condition required for a true PMC surface. Other methods such as carbon nanotube-based structures synthesized under high-temperature conditions have also been reported\textsuperscript{9}, where the achievable reflection phase is only about 20 degrees.

In the metamaterials community, Mie resonance-based dielectric particles, which are alternative candidates to metallodielectric structures, have been investigated over the past several years to achieve resonant electric and/or magnetic responses from the microwave regime up to the optical wavelengths\textsuperscript{9-20}. Arrays of nonmagnetic high-index dielectric resonators distributed in low-index surroundings have been demonstrated to provide electric and/or magnetic resonances with low loss at infrared wavelengths\textsuperscript{21, 22}.

In this letter, a methodology is introduced to synthesize a lossless optical mirror composed of dielectric nanoresonators, which exhibits near-unity reflectivity and a controllable reflection
phase at optical wavelengths. In particular, a PMC mirror with doubly periodic cross-shaped amorphous silicon (a-Si) nanoresonators was employed to achieve a polarization-insensitive response at normal incidence. This dielectric mirror was designed, fabricated, and fully characterized, experimentally demonstrating a reflectivity higher than 99% and a near-zero reflection phase at 1 μm. Such a structure can be tailored to provide near-unity reflectivity with an arbitrarily pre-defined reflection phase, showing great promise in lossless control of the phase of light.

4.2 Electromagnetic Design and Simulation

The unit cell of the dielectric nanoresonator that comprises the artificial mirror with controllable reflection phase is shown in Figure 4-1 (a). The nanoresonator, supported by a fused silica substrate, has a cross shape and is composed of a-Si. Since the dielectric constant of fused silica is much smaller than that of the a-Si, field leakage into the substrate is insignificant and therefore does not affect the performance of the resonator. This configuration ensures an independent azimuthal angle response for normally incident waves. The scattering property of the unit cell was predicted by full-wave simulation using the high frequency structure simulator (HFSS) finite element solver. In the numerical simulation, periodic boundary conditions were assigned to the lateral walls in both the x- and y-directions to obtain an infinite periodic array. A Floquet port was defined on top of the unit cell to produce an incident wave illuminating the structure at normal incidence. Due to the fact that the substrate has a thickness of more than 500 wavelengths in practice, it can be treated as an infinitely half-space. As a result, in the simulation model, a fused-silica substrate with a finite thickness was included, which is in direct contact with
the Floquet port beneath it in order to mimic an infinite half-space. Measured permittivity of both $\alpha$-Si and fused silica were incorporated into the model, which allows the dispersive properties of the constitutive materials to be accurately accounted for.

The structural dimensions of the nanoresonator were tuned in order to achieve a mirror condition with pre-defined phase response—out-of-phase and in-phase perfect reflections at 1.15 and 1.00 $\mu$m, respectively. The resulting design has a periodicity of 597 nm and a thickness of 366 nm. The cross-shaped $\alpha$-Si resonator has a width $W=116$ nm and a length $L=431$ nm. The simulated reflectivity and reflection phase are shown in Figure 4-2 (a). Two strong peaks are present at 1.14 and 0.99 $\mu$m, with reflectivity values larger than 99.8% and reflection phases of close to 180 and 0 degrees, respectively. These two reflection peaks correspond to the first ($TE_{01\delta}$) and second ($TM_{01\delta}$ mixed with a weak $TE_{01\delta}$) resonant modes of the dielectric nanoresonator, which give rise to artificial perfect electric and magnetic conducting conditions, respectively. As Figure 4-3 shows,
at 1.14 μm, when the incident electric field is polarized in the x-direction, a dipole-like pattern is observed in the magnetic field, while a curl-shaped pattern can be identified in the electric field. At 0.99 μm, the electric field exhibits a mixed curl-shaped and dipole-like pattern, with a stronger strength in the dipole mode response. Similarly, the magnetic field also exhibits a mixed dipole-like and curl-shaped pattern. This pattern indicates that at this wavelength, a $TM_{01δ}$ mode mixed with a weak $TE_{01δ}$ mode is excited inside the structure, primarily due to the low quality factor attributed to the limited permittivity value provide by the $a$-Si. Such mode mixing further gives rise to the high reflection in the broadband wavelength range between the two peaks, where the reflection phase varies gradually from 180 to 0 degrees.

Figure 4-2. (a) Simulated (top red) and measured (bottom blue) reflection amplitude of the cross-shaped $a$-Si nanostructure. (b) Simulated (top red) and measured (bottom blue) reflection phase of the cross-shaped $a$-Si nanostructure. (c) Simulated (top red) and measured (bottom blue) transmission amplitude spectrum. (d) Simulated (top red) and measured (bottom blue) transmission phase spectrum.
The cross-shaped geometry was chosen instead of a cubic or cylindrical shape, which have been previously studied in the mid-infrared and microwave regimes\textsuperscript{19,20}, because a smaller filling factor and geometry size are required in order to facilitate operation in the near infrared at the two resonant wavelengths.

Figure 4-3. (a) Magnetic field (left) and electric field (right) distributions in the $\alpha$-Si nanoresonator shown in Figure 4-1 (a) at the electric resonant wavelength of 1.14 $\mu$m. (b) Magnetic field (left) and electric field (right) distributions in the $\alpha$-Si nanoresonator at the magnetic resonant wavelength of 0.99 $\mu$m.
fundamental modes. Removing the corner area of a cubic shape, where the field confinement is weak, allows the $TM_{01\delta}$ and $TE_{01\delta}$ modes to be well confined with a relatively smaller filling factor. Also, the higher-order modes can be pushed to higher frequencies where they are sufficiently far from the PMC operating condition to allow for a continuous phase variation within the band. Moreover, the parameters of the cross-shape geometry provide more flexibility to manipulate these resonances. A parametric study was conducted in order to show that the reflection phase, as well as the particular wavelength where the PMC mirror condition occurs, can be effectively controlled by the dielectric nanoresonators. In this study, the effects of the geometrical parameters, including the thickness of the resonator as well as the length and width of the cross bar, were considered. When the thickness of the $a$-Si film increases from 300 to 390 nm with the length fixed at 116 nm and width fixed at 431 nm, as shown in Figure 4-4 (a), the zero-reflection-phase condition and the high-reflectivity band undergo a red shift from 0.92 to 1.02 μm due to the increased volume of the nanoresonator. Similar phenomena can also be observed when the length or the width of the cross bar increases with the other parameters fixed, as shown in Figure 4-4 (b) and (c). However, these two parameters have a weaker effect on the PMC band wavelength as compared to the thickness of the nanoresonators. When the resonator length is increased from 380 to 460 nm, the PMC and PEC bands are shifted from 0.96 to 1.01 μm and from 1.15 to 1.19 μm, respectively, whereas when the resonator width is increased from 100 to 125 nm, the PMC and PEC bands are shifted from 0.97 to 1.01 μm and from 1.13 to 1.21 μm, respectively. Hence, by changing the geometrical parameters, different degrees of shift can be obtained in the wavelengths of the two modes and the corresponding PMC and PEC mirror bands.
Figure 4-4. (a) Simulation study of reflection amplitude (top) and phase (bottom) shift with different $a$-Si nanoresonator thickness ($t_d$). (b) Reflection amplitude (top) and phase (bottom) shift with different $a$-Si resonator length ($L$). (c) Reflection amplitude (top) and phase (bottom) shift with different $a$-Si resonator width ($w$).
4.3 Fabrication and Characterization

The designed dielectric optical magnetic mirror was fabricated by depositing a 366 nm-thick $a$-Si film onto a 500 μm-thick fused silica substrate at 220°C using a plasma enhanced chemical vapor deposition (PECVD) system (Applied Materials P-5000 cluster). Then a 3 mm by 3 mm dielectric mirror pattern was defined by electron-beam lithography (Leica EBPG-5HR) with positive electron-beam resist (diluted Zep520A:Anisole=1:1, Zeon) and a 10 nm thermally evaporated Au layer on top to avoid the charging effect during E-beam exposure. The top Au film was removed by wet etching, using TFA Au etchant, before the developing process. The resist was developed in n-amyl acetate for 3 mins, then rinsed in methyl isobutyl ketone and isopropyl alcohol (8:1) for 1 min. After the pattern was defined, a 50 nm-thick Cr layer was deposited by thermal deposition on top of the $a$-Si film as the etch mask at a rate of 1.8 Å/s and was subsequently lifted-off by dissolving the undeveloped resist in Microposit Remover 1165. A 3 mm by 3 mm adjacent Au reference mirror pattern was defined by contact lithography. A 5 nm Ti adhesion layer, 60 nm of Au, and a 50 nm Cr etch mask layer were deposited by electron-beam evaporation (Kurt J Lesker Lab-18), and lifted-off by dissolving the undeveloped resist. Then, an inductive coupled plasma reactive ion etch (ICP-RIE, versalock) was used to transfer the pattern from the Cr etch mask to the $a$-Si film layer with chlorine (Cl$_2$, 40 scem) chemistry at a pressure of 2 mTorr, a substrate power of 200 W, and an ICP power of 300 W. Finally, the intermediary Cr mask was removed by Cr wet etchant (Transcene Cr etchant 1020) before optical measurement. The well-defined isolated $a$-Si resonator array with nearly vertical sidewalls is shown in the field enhanced scanning electron microscopy (FESEM) image in Figure 4-1 (b).

The optical performance of the fabricated sample was verified by measuring the reflection and transmission coefficients using a spectral holography technique$^{23-26}$, which has the capability to determine both the amplitude and the relative phase difference. Let us denote the complex
amplitude of the signal beam used to probe the sample as \( S(\omega) \), that of the reference beam as \( R(\omega) \), and the reflection and transmission coefficients of a sample as \( r(\omega) \) and \( t(\omega) \), respectively, where \( \omega \) is the angular frequency. The reflection coefficient characterization was performed using a Michelson interferometer, as illustrated in Figure 4-5 (a). A supercontinuum source was generated by propagating a sub-ns pulse at 1064 nm in a 20 m long photonic crystal fiber. The high spatial coherence of the supercontinuum, and hence its capability to focus, is ideally suited to this application due to the small size of the sample. The reflected beam was combined with the reference beam by a cubic beam splitter (BS), and a spectral hologram was recorded using an optical spectrum analyzer (OSA). A reference hologram was also captured from the adjacent reference Au mirror. The complex reflection coefficient of the device was then determined from the ratio of the reconstructed signals as described below.

The reflection interferograms, or hologram of the device in the frequency domain, can be expressed by

\[
l_S(\omega) = |S(\omega)r(\omega) + R(\omega)|^2
= |S(\omega)r(\omega)|^2 + |R(\omega)|^2 + S^*(\omega)r^*(\omega)R(\omega) + S(\omega)r(\omega)R^*(\omega)
\]  

(4-1)

The measurement was then repeated on the adjacent Au mirror to obtain a reference hologram:

\[
l_r(\omega) = |S(\omega)m e^{i(\omega/c)\Delta L} + R(\omega)|^2
= |S(\omega)m|^2 + |R(\omega)|^2 + S^*(\omega)m^* e^{-i(\omega/c)\Delta L} R(\omega) + S(\omega)m e^{i(\omega/c)\Delta L} R^*(\omega)
\]  

(4-2)

where \( m \) is the Au mirror reflectivity (approximately 1) and \( \Delta L \) represents the path length change between the two measurements. The complex reflection coefficient \( r(\omega) \) can thus be obtained by taking the ratio between \( S(\omega)r(\omega)R^*(\omega) \) and \( S(\omega)m e^{i(\omega/c)\Delta L} R^*(\omega) \), which can be digitally filtered from the respective holograms. An uncertainty on \( \Delta L \) arises when using the reference mirror in place of the dielectric mirror sample to perform the second measurement. Just a few microns of difference will result in a significant change in phase. Here the fact that for a lossless medium there
is a π/2 phase difference between the reflection and transmission coefficients is applied. Hence, the value of ΔL can be calculated by comparing with the transmission coefficient measurement conducted with a Mach-Zehnder interferometer, as shown in Figure 4-5 (b). The spectral holograms for the transmission measurement in the frequency domain can be expressed by

\[ I'_s(\omega) = |S(\omega)t(\omega) + R(\omega)|^2 = |S(\omega)t(\omega)|^2 + |R(\omega)|^2 + S^*(\omega)t^*(\omega)R(\omega) + S(\omega)t(\omega)R^*(\omega) \]  

(4-3)

Similarly, a reference hologram is obtained by removing the sample and repeating the measurement.

\[ I'_r(\omega) = |S(\omega) + R(\omega)|^2 = |S(\omega)|^2 + |R(\omega)|^2 + S^*(\omega)R(\omega) + S(\omega)R^*(\omega) \]  

(4-4)

The complex transmission coefficient \( t(\omega) \) is retrieved by taking the ratio of the last terms of equations (4-3) and (4-4).

The measured complex reflection and transmission coefficients are plotted in Figure 4-2 and compared with the simulation results. The reflectivity is maintained above 90% from 950 nm to 1130 nm with the phase gradually changing from -50° to 170°. The PMC condition is achieved at the wavelength of 1 μm with a reflectivity of 99.5% and near-zero reflection phase. The primarily \( TM_{01\delta} \) resonant mode was shifted by 10 nm compared to the simulation (0.99 μm with 99.8% reflectivity and zero reflection phase). The measured result shows strong agreement in performance between the designed and fabricated dielectric magnetic mirror with only a 1% difference in the \( TM_{01\delta} \) resonate mode. Similarly, the \( TE_{01\delta} \) resonate mode shifted from 1.14 μm, as predicted by simulations, to 1.11 μm, which was measured. This discrepancy is mainly due to the imperfect sidewall angle (about 87 degrees) of the a-Si resonator created during the etching
Figure 4-5. Schematic of the spectral holography setup for (a) reflection measurement, and (b) transmission measurement configurations. BS: beam splitter, PCF: photonic crystal fiber, OSA: optical spectrum analyzer.
process, which induces a smaller effective filling factor that shifts the resonance peak toward the shorter wavelength, matching the results of the parametric study in Figure 4-4.

### 4.4 Summary

In conclusion, a dielectric magnetic mirror composed of a doubly periodic array of cross-shaped $\alpha$-Si nanoresonators operating at 1 $\mu$m with near unity reflectivity and zero phase change in the electric field upon reflection was demonstrated. Simulations show that the zero reflection phase is achieved primarily by a $TM_{01\delta}$ resonant mode at 0.99 $\mu$m, with a corresponding reflectivity of 99.8%. The performance of the fabricated device was verified using spectral holography measurements, which agree well with simulation results, confirming a reflectivity of 99.5% and near-zero reflection phase at the magnetic resonance wavelength of 1$\mu$m. The results demonstrate that in-phase reflection at optical wavelengths can be obtained for an all-dielectric optical mirror with minimal loss, providing a practical route to engineering arbitrary boundary conditions using simple dielectric nanostructures.
4.5 Reference


Chapter 5

Formation and Frequency Response of Two-Dimensional Nanowire Lattices

In this chapter, ordered two-dimensional (2D) lattices were formed by assembling silica coated solid Au nanowires between coplanar electrodes using alternating current (AC) electric fields. The observed lattice structures were tunable with AC frequency, which varied lattice periodicity in real-time. Electric field simulations were performed to understand the formation of 2D lattice and how the observed lattice periodicity depends on the assembly conditions. Directed self-assembly of well-ordered 2D metallic nanowire lattices and reconfigured by changes in field conditions could enable new types of switchable optical or electronic devices. This research was enabled through the collaboration of nanowire synthesis and assembly experiments conducted by Sarah J. Boehm under the guidance of Dr. Christine D. Keating.

5.1 Motivation and Background

Nanowire assemblies are of interest for a wide variety of optical and electronic applications. However, achieving specific assembly organizations for desired device functions remains a largely unmet challenge. Approaches that take advantage of surface patterning and/or applied fields are perhaps the most promising. Such strategies offer new opportunities to incorporate functional components onto patterned substrates such as integrated circuit chips. For example, patterns of topography or chemical functionality have been used to control the location of particle assemblies on surfaces. Micropatterned electrodes allow localized application of electric fields, which can be used to control particle location and organization by inducing fluid flow, particle migration (e.g., dielectrophoresis), and field-dependent interparticle interactions.
The resulting forces can be tuned in real-time by varying the magnitude and frequency of the applied field, allowing assembly reorganization and optimization.

Alternating current (AC) electric-field gradients generated by micropatterned electrodes have been used to form ordered assemblies of colloidal particles on-demand at predetermined locations on a substrate. For example, dielectrophoretic (DEP) forces have been used to position individual microns-long metallic or semiconducting nanowires and nanotubes across narrow electrode gaps (less than wire length) to make electrical contacts for subsequent device characterization.\textsuperscript{20-23} When gaps larger than the particle size are used, chains of nano- or microparticles form to bridge the electrodes. Chaining has been observed in dielectric,\textsuperscript{24-27} metallic,\textsuperscript{28-34} and mixed-composition (e.g., Janus) particles.\textsuperscript{35-38} These assemblies rely on mutual DEP and field-induced dipolar interactions between adjacent particles to form chains parallel to the field lines.\textsuperscript{28,39} For dielectric particles, such as polystyrene, these chains have been shown to combine to create close-packed 2D colloidal crystals at high particle density.\textsuperscript{24,25,27,40,41} Open lattice-like structures were recently reported for polystyrene ellipsoid particles.\textsuperscript{24} Although chaining has been seen for metal nanospheres and nanowires,\textsuperscript{28-33} field-induced 2D crystallization for metallic particles has not been reported. The optical and electronic properties of metallic particles, and the responsiveness of these properties to particle organization, make 2D assemblies of metallic nanowires particularly interesting as potential device components.

Here, assembly of silica-coated solid Au nanowires in AC electric fields applied between coplanar electrodes was studied. The presence of a \textit{ca.} 30 nm silica shell prevented direct electrical contact between the Au nanowire cores. Induced interparticle interactions led to formation of well-ordered 2D nanowire lattices that filled the gap between electrodes over a 50× nearly 100 \(\mu\text{m}\) area. Electric field simulations of charge density for adjacent particles and electric force calculations as a function of wire position were performed to understand how field-induced dipoles led to the
observed lattice periodicity. Electric-field force is used to describe forces due to DEP, electrostatics and dipole-dipole interactions. Frequency-dependence of lattice periodicity were observed and lattice structures could be controlled by frequency-dependent dipolar coupling, which varied lattice periodicity in real-time. The ability to form—and rapidly reconfigure—assemblies of colloidal nanowires at predetermined locations is of interest for optical and electronic applications.

5.2 Assembly Experiment Setup

In the assembly experiment, an aqueous suspension of particles was placed within a silicone spacer on top of the patterned microelectrodes, and the voltage/frequency of the applied bias was gradually increased to form nanowire assemblies that spanned the gap between electrodes. The Au nanowires were 2.5 \( \mu \text{m} \) long with diameter of 290 nm and were coated with a 30 nm thick silica shell as shown in Figure 5-1 (b). The nanowires were suspended in deionized (DI) water, and assembly concentrations were adjusted to cover the electrode gap area. Coplanar Au electrodes were lithographically patterned (see Appendix B) onto glass cover slips such that nanowires could

![Figure 5-1. (a) Side and top view representation of coplanar electrode setup, showing stem/bulb (S/B) geometry and optical micrograph of electrode gap (transmitted light; intensities in this image were inverted such that electrodes appear bright for clarity). (b) Transmission electron micrograph image of Au nanowires.](image-url)
be viewed from below during the assembly process by reflectance optical microscopy. An alternating parallel (stem) and circular (bulb) geometry (S/B) for the coplanar electrode gaps as illustrated in Figure 5-1 (a) were used. The AC voltage was applied across the electrodes through contact to probe pads using a function generator. The conditions ranged from 200 to 2000 V/cm for peak field strength and 100 kHz to 10 MHz for frequency. Electrode gap sizes (i.e., stem region) is about 50 µm (>10× nanowire length).

5.3 Nanowire Lattice Formation and Frequency Response

5.3.1 Lattice Formation

Field-driven assembly of metallic particles across bare electrodes can result in electrical shorting, which can damage particles, removing the driving force for further assembly and can lead to poor quality assembled structures. This can be avoided by immediately shutting off the voltage or introducing a dielectric layer on the electrodes. Here, this limitation was overcome by coating the individual nanowires with a thin dielectric layer (30 nm silica).

The assembly process for silica-coated, solid Au nanowires can be seen at high magnification in Figure 5-2 and over a wider area in Figure 5-3 (a-c). Particles were allowed to sediment from suspension onto the surface for two minutes before the external AC voltage was applied. Each image shown in Figure 5-2 was collected after the respective field conditions had been applied for one minute. Voltage and frequency were stepped up during the course of the assembly process because immediate application of high field conditions would result in higher defect lattices. In the initial stages of assembly, dielectrophoresis led to migration of wires to the
regions of highest field gradient, resulting in their collection along the electrode edges with their long axes oriented parallel to the field lines as shown in Figures 5-2 and Figure 5-3 (a-c). Additional particles were then attracted to the ends of the assembled wires by mutual DEP, which ultimately resulted in the formation of particle chains that spanned across both stem and bulb areas as field strength was increased as illustrated in Figure 5-3. The formation of 2D lattices that spanned the entire gap region between electrodes occurred as peak field strength and frequency increased (417 V/cm, 150-300 kHz), as shown in Figure 5-2. All wires in the lattice structures were
oriented with their long axes parallel to the field lines, which refers to as the $x$-direction. Wires did not closely approach each other end-to-end along the $x$-direction, but rather had nearest-neighbors in the $y$-direction offset such that only a small portion of the particle tips were overlapping. This led to a relatively open lattice structure similar to the common “running bond” brickwork pattern. The S/B electrode design facilitated formation of these 2D lattices by pre-organization of chains within the bulb regions, which were then funneled into higher field stem regions.

Similar to 2D crystallization of dielectric particles in applied AC fields, field-induced dipole-dipole interactions between adjacent particles provide the driving force for lattice creation.\textsuperscript{24-26,40} This can be understood by considering the charge distribution on the nanowire array in a nonuniform electric field. Figure 5-4 shows the simulated charge density on a small portion of the Au nanowire array. In the initial phase of the assembly process, the polarized nanowires tend to produce chains with overlapping metal segments due to the attractive force between the oppositely charged regions of the wires. After a high enough wire density is reached, the chains crystallize into a “running bond” brickwork structure in which the field-induced charge alternates from positive to negative in both the $x$- and $y$-directions of the 2D lattice. Because the induced dipole forces are strong compared to other forces in this system (due to, e.g., electrothermal fluid motion, Brownian motion, electrostatic repulsion, van der Waals attraction between particles, \textit{etc.})\textsuperscript{19,43}, the observed structures depend on the factors that influence field-induced charge distribution, which include the wire geometry as well as the externally applied bias.
Figure 5-3. (a-c) Solid Au nanowires assembled within a stem/bulb electrode at constant field conditions (800 V/cm, 400 kHz, for the stem region). Arrow points to a chain of nanowires as it funnels from the bulb to the stem region of the electrode gap in less than two seconds. (d) Simulation showing field gradient at the top surface of the patterned stem/bulb electrodes using experimental bias conditions. Red indicates highest field gradient and blue indicates lowest. The field gradient is highest in the stem region.
5.3.2 Frequency Dependent Lattice Restructuring

The $x$-direction packing density of the solid Au wire 2D lattice depended on the frequency of the applied AC voltage, with decreased end-to-end distances observed at lower frequency. Figure 5-5 (a) and (b) show the same assembly at 100 kHz and 900 kHz, respectively, under otherwise identical conditions. The lattice structure is more open at the higher frequency. The experimental data of this difference was quantified by measuring the length along the $x$-direction over which the tips of adjacent wires overlapped. This lattice overlap decreased from $1.00 \pm 0.29 \mu m$ to $0.75 \pm 0.25 \mu m$ as the frequency was increased from 100 kHz to 900 kHz, at

Figure 5-4. (Top) Cartoons of solid Au nanowires illustrating induced dipole interactions between neighboring particles. (Bottom) Three-dimensional simulation showing charge density within solid Au nanowires in a non-uniform electric field. Electric field: (340 V/cm at 900 kHz, nanowire overlap length set at 0.75 μm.)
constant field strength. This frequency-dependent particle spacing provides a way to dynamically tune the structure of an assembly, specifically its $x$-axis lattice periodicity, in real-time.

The frequency-dependence of the lattice packing density was studied theoretically by conducting 3D electric field simulations on a simplified four-wire portion of the structure. The Au wire core was considered to be a perfect conductor, while the silica shell and surrounding water were modeled as lossy dielectrics having a permittivity $\varepsilon$ and a conductivity $\sigma$. The positions of the top and bottom wires were fixed within the electrode gap, while the left and right wires between them were moved along the $x$-axis as shown in Figure 5-6. The $x$-component of the electric force ($F_x$) on the moving wires was calculated as a function of Au overlap length to determine the energetically stable nanowire array configuration where $F_x = 0$.

Experimentally measured dielectric properties were not available for the suspending DI water or the sol-gel fabricated silica shell. Therefore, the simulation was repeated for a range of...
physically realistic dielectric properties to identify values that provided the best agreement with experiment. The DI water used in these experiments has a pH ~3 and contains ions from the wire fabrication process, making it more conductive than pH 7 DI water.\textsuperscript{44} In addition, the sol-gel silica films are porous, resulting in a water filled silica matrix with higher conductivity and permittivity than high-quality thermal or plasma deposited oxide.\textsuperscript{45} To account for these factors, the conductivity of DI water was varied from $5 \times 10^{-6}$ S/m to $1 \times 10^{-2}$ S/m and silica from $5 \times 10^{-7}$ S/m to $1 \times 10^{-3}$ S/m. For all simulations, the real part of the permittivity for the DI water was fixed at 80. The silica shell permittivity was estimated using Maxwell’s mixing rule for a 90% silica and 10% water matrix, giving a value of 4.88.\textsuperscript{46} For a given simulation, the conductivity of the silica shell was always maintained at least one order of magnitude smaller than that of DI water.

The electric-field simulations show that nanowire overlap length decreases with increasing frequency when the DI water conductivity is higher than $7 \times 10^{-4}$ S/m and the silica conductivity is in the range of $1 \times 10^{-5}$ to $1 \times 10^{-4}$ S/m. In contrast, when both the DI water and silica conductivities are either high or low, little to no frequency dependence is observed in the lattice packing density. This can be understood by considering a simplified electrical equivalent circuit model of the nanowire lattice, which is composed of an interconnected network of parallel resistor (R) and capacitor (C) circuit elements, as shown in Figure 5-7. The two dominant types of RC pairs are associated with the physical overlap between adjacent wires in the $y$-direction and the end-to-end spacing between adjacent wires in the $x$-direction of the lattice. The network approaches being purely resistive when the conductivity of both the DI water and porous silica shell are high and purely reactive (capacitive) when their conductivities are low. In these limits, the voltage drop (field distribution) across the network (lattice) is essentially independent of frequency, and hence the lattice remains unchanged as the frequency is varied. Between these limits, however, the ratio of the admittances for the two types of RC pairs changes significantly.
Figure 5-6. Simulation of the electric force (x-component) on the Au nanowires in an AC electric field (340 V/cm, 100 or 900 kHz) as a function of Au overlap with vertical neighbor, d (µm). The top and bottom wires in each simulation are fixed while the left and right wires are simultaneously moved in the x-direction. The stable nanowire array configuration occurs when the electric force = 0.
with frequency. Consequently, the voltage division (field distribution) between the RC pairs changes resulting in changes in the electrostatic forces and nanowire overlap. Using conductivity values of $4 \times 10^{-3}$ S/m for DI water and $4 \times 10^{-5}$ S/m for the silica shell, the simulated overlap length shifted from 0.75 μm at 100 kHz to 0.50 μm at 900 kHz. This agrees well with the 0.25 μm shift measured experimentally. The discrepancy in the absolute overlap lengths at the upper and lower frequency is due to the simplified lattice structure used in the simulation compared to that of the experiment.

Figure 5-7. Equivalent RC circuit diagram shows coupling between neighboring nanowires in both x-and y-directions.
5.4 Summary

A versatile and on-demand particle assembly at predetermined sites on the substrate, dictated by the position of the coplanar electrodes was demonstrated. The overall lattice periodicity of the Au nanowires could be varied in real-time by altering the applied frequency. The silica-coated, solid Au-core nanowires used here go beyond previous work on AC-field driven assembly of 2D colloidal crystals, which has largely focused on dielectric particles. It should ultimately be possible to take advantage of the optical properties of metal-containing particles in reconfigurable devices where changes in particle organization alter assembly properties, e.g., optical reflectance, absorbance, or transmittance.
5.5 References


Chapter 6

Summary and Future Work

6.1 Summary

The goal of this research was to realize metamaterials with tailored optical properties for IR and optical wavelengths and to develop new techniques to explore metamaterials with novel functionalities. First, metamaterials composite of periodic nanostructures precisely defined by top-down technique were investigated. The proper nanostructures were optimized to achieve the desired optical response, and the constituent materials were chosen based on their measured optical constants to maximize the design performance. Metamaterials with metallo-dielectric multilayer and all-dielectric nanoresonators were exploited in this work. Second, a bottom-up technique using electric-field-assisted directed self-assembly was developed to construct well-ordered metallic nanowire lattices with tunable and reconfigurable capabilities.

In Chapter 2, a multilayer Pd/polyimide structure containing a single-layer nanostructure metal screen was demonstrated to achieve near-ideal absorptivity from 2 μm to 5 μm with a wide field-of-view (FOV) over ±45°. The designed absorber consisted of three strongly coupled nanostructure elements inside of a unit cell, and the multiple resonances induced by these elements resulted in high absorption across the broad mid-IR band. To confirm its performance, the structure was fabricated and characterized using Fourier transform infrared spectroscopy (FTIR). The measured absorptivity of the sample had an average value of 98% over a ±45° incidence-angle range from 1.77 μm to 4.81μm with a minor blue shift from the designed values. This demonstration advances the state-of-the-art in the field of high-performance broadband
metamaterial absorbers that can be reliably fabricated using a single patterned layer of metal nanostructures.

The research in Chapter 3 developed a new approach to design optical wave plates with high polarization conversion efficiency and desired phase modulation over a broad bandwidth and wide FOV. The anisotropic optical response was achieved with a thin layer of optimized nanostructures and was enhanced by constructive interference through the Au/SiO$_2$ multilayer. Both half-wave and quarter-wave plates were studied with strongly coupled nanorod arrays and were fabricated for characterization. A custom-built optical setup was used to verify the performance of the design, and the measured results matched the simulation well, exhibiting greater than 92% polarization conversion ratio and reflection magnitudes in the wavelength regime between 640 nm and 1290 nm from normal incidence up to ±40°. This work outlines a versatile strategy to create metasurface-based photonics with diverse optical functionalities.

In addition to the metallo-dielectric multilayer-based metamaterials, in Chapter 4, a high-efficiency optical magnetic mirror based on Mie resonance was synthesized at wavelength of 1 μm using a doubly periodic array of $a$-Si nanostructures on a fused silica substrate. The high reflectivity and zero phase modulation were achieved by exciting the primarily magnetic $TM_{015}$ resonant mode within the cross-shaped dielectric nanoresonators. This concept and approach pave the way for synthesizing lossless artificial reflecting electromagnetic boundaries with arbitrary phase response and hold great promise for applications ranging from nanocavities to nanowaveguides and nanoantennas.

To explore the variety of the nano building blocks for metamaterials beyond top-down techniques, assembly of pre-synthesized complex nanoparticles in a controllable way is desired. Chapter 5 described the patterning of well-ordered and tunable 2D lattices with 2.5 μm long, 290 nm diameter Au nanowires coated with a 30 nm-thick SiO$_2$ shell using electric-field-assisted directed self-assembly. By switching the frequency of the applied AC electric field from 100 kHz
to 900 kHz, the overlap between the adjacent nanowires changed from 1.00 ± 0.29 μm to 0.75 ± 0.25 μm. Electric field simulations and electric force calculation were conducted to understand the formation and frequency response of the 2D lattices. An equivalent RC model was proposed to understand and predict the frequency range where the overlap between the adjacent nanowires depends on the applied AC field conditions. This work provides an alternative way to obtain reconfigurable metamaterials along with a potential approach to construct complex nano building blocks that are not achievable by conventional top-down technique.

6.2 Recommendations for Future Work

6.2.1 Light-Induced Tunable Assembly of a-Si:H Nanodisks

Fundamental studies of interparticle organization within co-assemblies of two or more particle types are an important step towards increasing the functional diversity of assembled structures. Optical fields are of particular interest because they provide a way to control a material’s fundamental property, particle polarizability, in real-time. For example, particle subpopulations within mixtures of different particle types can be selectively addressed to alter their interaction potentials. Cooperative responses to changing conditions are of fundamental interest and are exciting in the longer-term as reconfigurable materials.

Metallic nanoparticles have strong polarizability and can be collected by positive dielectrophoresis (DEP) across the working frequency range. In contrast with metallic nanoparticles, semiconductor nanoparticles have weaker but tunable conductivities. Hence, their collective behavior could be controlled and switched between negative DEP and positive DEP by changing the particle conductivity and the value of the Clausius-Mossotti (CM) factor, as described in equation (1-3), with light illumination. The effect of an external light source on the
semiconductor nanowire assembly has been utilized to improve the yield of CdSe nanowire assembly due to the optical enhancement of conductivity \(^1\). Manipulation of nanoparticles using optoelectronic tweezers (OET), where projected light patterns on an \(a\)-Si:H thin film function as virtual electrodes, have also been demonstrated \(^2\)-\(^5\). However, assembly of particle mixtures into active reconfigurable arrays by real-time modification of particle polarizability has not yet been investigated.

\(a\)-Si:H nanoparticles with a diameter of 1\(\mu\)m and thickness of 150 nm, synthesized by top-down fabrication with plasma enhanced chemical vapor deposition (PECVD) deposited \(a\)-Si:H film, are shown in Figure 6-1 (a). The polarizability of these particles can be modulated by a light-induced conductivity change, as illustrated in Figure 6-1 (b). The fabrication process and effective conductivity characterization approach for the \(a\)-Si:H particles are explained in Appendix C. The real parts of the CM factor, which determines the sign of the DEP force as described in equation (1-3) for \(a\)-Si:H nanoparticles with different conductivity, are plotted as the dashed curves in Figure 6-2. These different conductivity values correspond to different light illumination intensities, and the change of the CM factor of the \(a\)-Si:H particles has been experimentally observed. The
permittivity of the particles and the solution, which is ethylene glycol (EG), are 11.7 and 37.7 respectively, and the conductivity of EG is approximately $1.04 \times 10^{-4}$ S/m. EG was chosen as the dispersion medium instead of water because it is less polarizable and induces fewer surface charges on the $a$-Si:H nanoparticles. Also, the relatively high vapor pressure makes EG less volatile, allowing for a longer observation time during assembly experiments. At frequencies below 1MHz, the CM factor of the particles changes drastically from negative to positive values when the effective conductivity of the particles increases upon light illumination. The photogenerated carriers inside of the particles increase the susceptibility of the nanoparticles to applied electric fields and field-induced dipolar interparticle forces and, hence, modify the collective behavior and the organization of the particles.

Figure 6-3 shows preliminary experimental results of controllable co-assembly and selective assembly of nanoparticles with two different types. The experiment was conducted with a Nikon inverted optical microscope (TE2000U) with a halogen lamp and observed with
transmitted light under a 100x oil immersion lens. The electric field was applied on a coplanar interdigitated electrode (IDT) with a similar electric field distribution as in Chapter 5. The
Figure 6-3. (a)-(c) Sequentially increased, reduced, and increased illumination intensity to selectively choose the assembly particle type at 500 kHz, $10^4$ V/cm. (d) Selectively assembled single-particle-type 2D crystal within the mixed particle solution under low illumination intensity. (e) 2D crystal with mixed particle type is formed by increasing the $\alpha$-Si:H nanoparticle conductivity, and both types of particles exhibit positive DEP.
assembly was conducted under a fixed applied electric field of $10^4$ V/cm at 500 kHz. The red and yellow particles were made with similar processes, except that the red particles were fabricated with PECVD-deposited $a$-Si:H film, and the yellow particles with electron-beam-evaporated $a$-Si film. The effective conductivities of the yellow particles dispersed in EG were measured with the same approach described in Appendix C, and their frequency response is plotted in Figure 6-2 as the solid curves. The effective particle conductivity is higher; therefore, their crossover frequency shifted to a higher frequency compared to the PECVD fabricated particles. Accordingly, they exhibited positive DEP at frequencies below 5 MHz regardless of the illumination intensity. The higher effective particle conductivity for the yellow particles may be attributed to: (1) Larger surface area due to higher porosity of the material, which may induce larger surface charges when dispersed into solution; (2) Trapped charges inside and at the surface of the particles during the electron-beam evaporation process$^6$; (3) Higher defect density inside of the material because of the higher density of dangling bonds$^7$. The yellow particles maintain positive DEP under all illumination conditions once the electric field is applied, as seen in Figure 6-3 (a) to (e). In contrast, the red particles were only assembled with positive DEP at high illumination intensity, as seen in Figure 6-3 (a), (c) and (e); otherwise, negative DEP pushed them away from the gap region due to the low effective particle conductivity.

This photoexcitation effect offers a powerful new way to selectively address and modify the organization of particles in a mixed population in real time. Future studies of this capability can be conducted for index-modulated optics and disordered photonics.

### 6.2.2 Frequency-Dependent Reconfigurable Assembly of $a$-Si Nanodisks

Anisotropic nanoparticles, which contain unequal axis lengths, are common and have interesting phenomena and applications beyond isotropic particles. Upon applying an electric field,
the polarizable particles align in the electric field so that their longest axis is parallel to the electric field. More interestingly, if the anisotropic nanoparticles are made with lossy dielectric materials, their orientation can be changed with the frequency of the applied electric field due to the torques, as detailed in Appendix D. This effect can be used to characterize the conductivity of semiconductors without contacting electrodes and to determine the dielectric properties of cells. Recently, researchers have demonstrated that frequency-dependent reconfigurable orientation modulation can be achieved in the nanoscale with TiO$_2$ anisotropic nanoparticles. However, semiconductor nanoparticles are more interesting for optical devices because of their higher refractive index and low loss in the optical regime.

$a$-Si nanodisks with a diameter of about 1 $\mu$m and a thickness of 350 nm, synthesized by electron-beam-evaporated a-Si film, are shown in Figure 6-4. When these particles were dispersed in EG, their orientation and organization could be controlled by adjusting the applied electric field. The experiments were conducted under a Nikon inverted optical microscope (TE 2000U) with a reflection module and were observed with a 100x oil immersion lens. The electric
Figure 6-5. Electron-beam evaporated $a$-Si particle orientation and frequency dependence with the same applied electric field of $10^4$ V/cm. (a) At 400 kHz (b) at 3 MHz.
field was applied on a coplanar interdigitated electrode (IDT) with a similar field distribution as in Chapter 5.

As shown in Figure 6-5 (a), the $a$-Si nanodisks were collected across the gap with their longest axis parallel to the applied electric field at frequencies of 400 kHz and formed close-packed 2D crystals with a similar lattice structure to the gold nanowires mentioned in Chapter 5. Compared to Figure 6-3, $a$-Si nanodisks were arranged in a condenser array. Instead of laying with the circular facet parallel to the substrate, the particles rotated 90° to align their circular facet perpendicular to the substrate, as shown in the inset of Figure 6-5 (a). This is due to a higher particle density than that in 6.2.1, and in this format, more particles were collected in the same area. However when the frequency increased to 3MHz, the nanodisks rotated 90°, with the shortest axis parallel to the electric field, and all the particles were rearranged into parallel stacked nanodisk chains, as shown in Figure 6-5 (b).

Further studies will be carried out to scale down the particle size for use in optical devices with tunable responses in the visible and near-infrared range.

6.2.3 Nanoparticle Assembly with Predefined Structures

In Chapter 5, section 6.2.1 and 6.2.2, particle-particle/particle-substrate interactions using coplanar electrodes have been investigated and can be harnessed to create complex structures. More interestingly, in addition to enabling the construction of dynamically reconfigurable patterns, this approach also provides a new way to modulate the performance of pre-patterned optical systems/devices by manipulating the interaction between the nanoparticles and optical systems/devices. The preliminary research presented in this section explores the assembly of nanoparticles on a semiconductor micro-ring structure, as shown in Figure 6-6. A heavily doped Si substrate (resistivity of 0.01~.02 $\Omega$.cm) with a prefabricated micro-ring pattern served as the
bottom electrode as illustrated in Figure 6-6 (a) and (b), and a transparent ITO cover glass was placed on top of a 120 μm-thick spacer to serve as the top electrode. A voltage was applied vertically between the ITO glass and patterned substrate, which contained a 20 μm-diameter micro-ring, as shown in Figure 6-6 (b). The distribution of the electric field gradient was simulated with COMSOL Multiphysics, and the results are plotted in Figure 6-6 (c) and (d) for the side view and top view. The largest electric field gradient (red regions) occurred around the ring and was the location of the strongest positive dielectrophores force. These regions more preferentially attract the nanoparticles.

Experimental demonstrations were conducted with metal-coated dielectric particles and solid dielectric particles in water solutions and were observed under a customized Zeiss microscope.

![Figure 6-6. (a) Schematic figure of the assembly setup. (b) FESEM image of Si micro-ring with a radius of 10 μm, width of 500 nm and thickness of 230 nm. (c) Simulated electric field gradient distribution in the cross-sectional view. (d) Simulated electric field gradient distribution from the top view.](image-url)
with a reflected dark-field module and a 20x objective lens, and the results are presented in Figure 6-7. Two arrangements of the metal-coated particles were observed, which can be switched back and forth by changing the frequency of the applied electric field: (1) at 10 kHz, particles were collected around the side of the micro-ring, as shown in Figure 6-7 (a), which was due to the combined effect of positive DEP and the electrohydrodynamic effect; (2) at 100 kHz, particles were collected on top of the micro-ring, as shown in Figure 6-7 (b), when the positive DEP was dominant and the electrohydrodynamic effect was negligible. For the solid dielectric particles, the frequency range at which the particles could be assembled around the micro-ring was smaller due to their relatively lower conductivity and polarizability. At 10 kHz, solid polystyrene particles were collected around the micro-ring, as shown in Figure 6-7 (c), with a result similar to metal-coated particles at low frequency. Similar phenomena were observed for TiO$_2$ and SiO$_2$ particles as well.

![Figure 6-7](image.png)

Figure 6-7. (a-b) 1μm-diameter Ag-coated SiO$_2$ particle with applied electric field of 830 V/cm at two different frequencies. (c) 1μm-diameter polystyrene particle with applied electric field of 500 V/cm.
Similar to what was described in Figure 6-6, two closely placed Si micro-rings with 200 nm spacing were also fabricated to serve as the bottom assembly electrode, as shown in Figure 6-8 (a). Both metal-coated dielectric particles and solid dielectric particles were also assembled on
the double ring electrode. Similar to their assembly on the single micro-ring electrode, Ag-coated SiO₂ particles also exhibited two assembly arrangements when assembled with an AC field with different frequencies, as shown in Figure 6-8 (b) and (c). At 20 kHz, positive DEP dominated the assembly process, and the Ag coated SiO₂ particles were attracted evenly onto the top of the two micro-rings. At a lower frequency of 3 kHz, the electrohydrodynamic effect arose and pushed all the particles to the center area. For the solid polystyrene particles, the assembly happened at lower frequencies compared to the metal-coated dielectric particles and were collected between the two micro-rings, as shown in Figure 6-8 (d), due to the combined effect of positive DEP and the electrohydrodynamic effect. The variety of the particle types and the tunability of the assembly provide a versatile way to modulate the properties of the micro-ring resonator structure.

The studied assembly approach on micro-rings has potential applications as tunable optical resonators. Micro-ring resonators are an important component for integrated photonic circuits due to their compact volume and high quality factor $Q^{15,16}$. Recently, single-mode lasers based on the Parity-Time-Symmetric micro-ring lasers and dark-state lasers$^{17-19}$ have been demonstrated. By varying the gain/loss contrast between coupled micro-rings, the exceptional point where the two split resonance lines fuse together can be obtained. A slight perturbation of the system will cause modes within it to split, which is proportional to the square root of the perturbation. Hence, such schemes are excellent candidates for detection and sensing with ultra-high sensitivity. By integrating the assembly strategies to vary the gain/loss in a semiconductor micro-ring resonators system, the modes within the system can be modulated. Future studies will be conducted with InGaAsP micro-ring lasers to investigate the exceptional points in the system by introducing loss with nanoparticle assembly for mode modulation and study of ultra-sensitive detection.
6.3 References


Appendix A

Electromagnetic Design Optimization

A.1 Genetic Algorithm (GA)

GA is an design approach inspired by evolution in nature to optimize multiple parameters simultaneously\(^1\). The initial population is randomly generated and encoded in binary strings containing the structural information. Figure A-1 illustrates a unit cell; the different colors represent different materials that are encoded in the initial population with a ‘0’ or a ‘1’. The performance of the candidates is evaluated by Full-wave Electromagnetic Analysis and compared to the cost function. The multiple parameters are optimized by mimicking the evolution process in nature through inheritance, mutation, selection and crossover as shown in the flow chart in Figure A-2. This loop iterates till the minimum cost is obtained.

Figure A-1. Example of a unit cell.
Figure A-2. Flow chart of GA.
A.2 Covariance Matrix Adaptation Evolution Strategies (CMA-ES)

CMA-ES is a self-adaptive evolutionary algorithm with an optimization process\(^2\) as illustrated in Figure A-3. At the initialization stage, generation with a distribution mean of \(<X>\) is chosen and marked as \(<X>_{g=0}\). In addition, the number of population \(\lambda\), number of selected children \(\mu\), and step-size \(\sigma\) are selected at the beginning of the process. \(w_i\) is the recombination weight of the \(i^{th}\) best child, \(\mu_{\text{eff}}\) is the effective number of children with weighted averages, \(c_\sigma\) is learning rate for step size control, \(c_c\) is learning rate for the rank-one update of the covariance matrix, \(d_\sigma\) is damping factor for step size control, and \(c_{\text{cov}}\) is learning rate for the covariance matrix update. Also initialized at this step are covariance matrix \(C\), eigenvectors of the covariance matrix \(B\) and eigenvalues of the covariance matrix \(D\). The sample distribution is described by the multivariate Normal distribution of \(N(<X>, \sigma^2C)\) and the population is evaluated with the cost function. If the optimized parameters are not found, the best \(\mu\) of \(X_{i,1:\lambda}\) will be selected and the new mean for the next generation will be obtained from \(X_{i,1:\mu}\). The step-size \(\sigma\), conjugate evolution path \(p_\sigma\), covariance matrix \(C\) and evolution path \(p_c\) will be updated. Sample distribution of the new generation will be obtained with updated mean and covariance matrix. The process continues until the population concentrates over the global optimum.
Figure A-3. Flow chart of CMA-ES².
## Appendix B

### Assembly Electrode Fabrication

<table>
<thead>
<tr>
<th>Process</th>
<th>Recipe of the Steps</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sample Clean</td>
<td>Rinse the substrate with Acetone, IPA</td>
</tr>
<tr>
<td></td>
<td>Blow dry with N$_2$ and bake at 100 °C for 1 min</td>
</tr>
<tr>
<td>Pattern the Electrodes</td>
<td>Spin PMGI SF6 (MicroChem) at 4000 rpm for 45 sec</td>
</tr>
<tr>
<td></td>
<td>Bake at 250 °C for 5 mins</td>
</tr>
<tr>
<td></td>
<td>Spin 3012 (MicroChem) at 4000 rpm for 45 sec</td>
</tr>
<tr>
<td></td>
<td>Bake at 97 °C for 1 min</td>
</tr>
<tr>
<td></td>
<td>GCA 8000 stepper exposure 0.45 sec</td>
</tr>
<tr>
<td></td>
<td>Develop: CD 26 (MicroChem) 1 min, rinse with DI water and blow dry with N$_2$</td>
</tr>
<tr>
<td></td>
<td>Deep UV flood exposure 700 s @5.3 mW</td>
</tr>
<tr>
<td>Metal evaporation and Lift-off</td>
<td>Develop: 101A (MicroChem) 1 min, rinse with DI water and blow dry with N$_2$</td>
</tr>
<tr>
<td></td>
<td>Electron-beam evaporation (Kurt Lesker Lab-18) Ti/ Au=10:50 nm</td>
</tr>
<tr>
<td></td>
<td>Dissolve photoresist and liftoff metal in Microposit Remover 1165</td>
</tr>
</tbody>
</table>
Appendix C

Nanoparticle Fabrication and Characterization

C.1 PECVD aSi:H Particle Fabrication:

<table>
<thead>
<tr>
<th>Process</th>
<th>Recipe of the Steps</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sample Clean</td>
<td>Dip Si wafer in BOE 10:1 for ~5 mins</td>
</tr>
<tr>
<td></td>
<td>Rinse with DI water and blow dry with N₂</td>
</tr>
<tr>
<td>Film Deposition (PECVD)</td>
<td>Deposit SiGe (1.2 nm/s)</td>
</tr>
<tr>
<td></td>
<td>GeH₄:SiH₄:H₂ 25:5:300 sccm</td>
</tr>
<tr>
<td></td>
<td>Pressure 2 Torr</td>
</tr>
<tr>
<td></td>
<td>Power 100 W</td>
</tr>
<tr>
<td></td>
<td>Temperature 220 °C</td>
</tr>
<tr>
<td></td>
<td>Susceptor spacing 500 mils</td>
</tr>
<tr>
<td></td>
<td>Deposit aSi (1.35 nm/s)</td>
</tr>
<tr>
<td></td>
<td>SiH₄:Ar 30:700 sccm</td>
</tr>
<tr>
<td></td>
<td>Pressure 1.5 Torr</td>
</tr>
<tr>
<td></td>
<td>Power 200 W</td>
</tr>
<tr>
<td></td>
<td>Temperature 220 °C</td>
</tr>
<tr>
<td></td>
<td>Susceptor spacing 500 mils</td>
</tr>
<tr>
<td></td>
<td>Deposit SiO₂ (1.74 nm/s)</td>
</tr>
<tr>
<td></td>
<td>N₂O:SiH₄:N₂ 840:20:1400 sccm</td>
</tr>
<tr>
<td></td>
<td>Pressure 3.5 Torr</td>
</tr>
<tr>
<td></td>
<td>Power 300 W</td>
</tr>
<tr>
<td></td>
<td>Temperature 220 °C</td>
</tr>
<tr>
<td>Susceptor spacing</td>
<td>340 mils</td>
</tr>
<tr>
<td>-------------------</td>
<td>----------</td>
</tr>
<tr>
<td><strong>Pattern</strong></td>
<td></td>
</tr>
<tr>
<td>Spin HMDS at 4000 rpm for 45 sec</td>
<td></td>
</tr>
<tr>
<td>Bake at 97 °C for 1 min</td>
<td></td>
</tr>
<tr>
<td>Spin 3012 (MicroChem) at 4000 rpm for 45 sec</td>
<td></td>
</tr>
<tr>
<td>Bake at 97 °C for 1 min</td>
<td></td>
</tr>
<tr>
<td>GCA 8000 stepper exposure 0.5 sec</td>
<td></td>
</tr>
<tr>
<td>Develop: CD 26 (MicroChem) 1 min, rinse with DI water and blow dry with N₂</td>
<td></td>
</tr>
<tr>
<td><strong>Film Etch</strong></td>
<td></td>
</tr>
<tr>
<td>SiO₂ etch at 25 °C (3 nm/s)</td>
<td></td>
</tr>
<tr>
<td>Pressure</td>
<td>5 mTorr</td>
</tr>
<tr>
<td>RF₁</td>
<td>150 W</td>
</tr>
<tr>
<td>RF₂</td>
<td>700 W</td>
</tr>
<tr>
<td>CF₄</td>
<td>40 sccm</td>
</tr>
<tr>
<td>Si/SiGe stack etch at 25 °C (5 nm/s)</td>
<td></td>
</tr>
<tr>
<td>Pressure</td>
<td>5 mTorr</td>
</tr>
<tr>
<td>RF₁</td>
<td>75 W</td>
</tr>
<tr>
<td>RF₂</td>
<td>500 W</td>
</tr>
<tr>
<td>Cl₂</td>
<td>40 sccm</td>
</tr>
<tr>
<td><strong>Particle Release</strong></td>
<td></td>
</tr>
<tr>
<td>BOE 10:1 3 mins</td>
<td></td>
</tr>
<tr>
<td>Rinse with DI water and blow dry with N₂</td>
<td></td>
</tr>
<tr>
<td>Put the sample in H₂O₂ at 65 °C for 3 mins</td>
<td></td>
</tr>
<tr>
<td>Replace the solution with ethylene glycol</td>
<td></td>
</tr>
</tbody>
</table>
C.2 Effective Conductivity Characterization Approach

The effective conductivity of the particles could be retrieved based on equation (1-3). The sign of DEP force which is related to $K_f$ in equation (1-3a) can be determined experimentally with a stem/bulb, interdigitated, or quadruple electrode as shown in Figure C-1. Below the transition frequency where CM factor is 0, particles are assembled to the high electric field gradient region, and above the transition frequency, particles are pushed away from the high electric field gradient region or collected in the low electric field gradient region. Based on the recorded frequency response, effective conductivity of particles can be calculated with the following matlab code.
Matlab code for retrieving the effective conductivity:

clear
c1c
e1=37.7*8.85e-12; % medium permittivity
e2=11.7*8.85e-12; %particle permittivity
r1=1.07e-4; %medium conductivity
r2=1e-4; %particle conductivity
con=[1.4e-2 1.5e-2 1.8e-2 2.5e-2 2.7e-2 2e-4 1.2e-3 2e-3 4e-3 7.5e-3];
set(0,'DefaultAxesColorOrder',[1 0 0 1 0 0 0 1 1 0 0 0 0],...
    'DefaultAxesLineStyleOrder','-|--|:',) % set curve format

for m=1:10
    r2=con(m);
end

for i=1:199701

Figure C-1. Experiment observation of positive DEP (PDEP, left column) and negative DEP (NDEP, right column). (a) Stem/Bulb electrode with aSi:H particles. (b) Interdigitated electrode with aSi:H particles. (c) Quadruple electrode with polystyrene particles.
\( w(i) = 2\pi \times 1000 \times (i + 99); \) \hspace{1cm} \text{% set frequency range}
\( ep = e_2 - j \times r_2 / w(i); \)
\( em = e_1 - j \times r_1 / w(i); \)
\( K(i) = \text{real}((ep - em) / (ep + 2 \times em)); \) \hspace{1cm} \text{% CM factor}
end

semilogx((w/2/pi),K,'linewidth',3); \hspace{1cm} \text{% plot CM factor}
hold all end

legend(num2str(con', '=%d'))
xlabel('Frequency', 'FontSize', 30);
ylabel('Clausius-Mossotti factor', 'FontSize', 30);
axis([3e5 2e7 -0.5 1]);
set(gca,'FontSize',30)
grid on
Appendix D
Anisotropic Lossy Dielectric Particle Rotation

Anisotropic dielectric lossy particles have frequency responded electro-rotation effect due to the unbalanced torque exert on the particles. By employing the effective moment method, the net torque on an ellipsoid particle can be expressed as:

\[
\langle T^e \rangle_\alpha = \frac{1}{2} \text{Re} \left[ (P_{\text{eff}})_\beta^* E_{0,y} - (P_{\text{eff}})_y E_{0,\beta}^* \right] \quad (D-1)
\]

The subscripts \( \alpha, \beta \) and \( \gamma \) follow the convention of a right-handed coordinate system, that is, \( x \to y \to z \to x \). The effective moment components can be expressed as:

\[
(P_{\text{eff}})_\alpha = 4\pi abc \varepsilon_1 K_\alpha E_{0,\alpha}
\]

where \( \alpha \) represents \( x, y \) and \( z \), \( \varepsilon_1 \) is the medium permittivity, \( E_0 \) is the electric field vector with magnitude and frequency \( \omega \). \( a, b \) and \( c \) are the length of three axis of the particle.

\[
K_\alpha = \frac{\varepsilon_\alpha - \varepsilon_1}{3[\varepsilon_1 + (\varepsilon_\alpha - \varepsilon_1)L_\alpha]}
\]

\[
L_\alpha = \frac{abc}{2} \int_0^\infty \frac{ds}{(s+a^2)R_s} \quad (D-4)
\]

\( L_x \) is the depolarization factor in \( x \) axis, and expressions for \( L_y \) and \( L_z \) can be obtained by substituting \( y \) or \( z \) for \( x \) and \( b \) or \( c \) for \( a \). Hence, expression of torque in equation (D-1) can be derived as:

\[
\langle T^e \rangle_\alpha = \frac{2}{3} \pi abc \varepsilon_1 (L_\gamma - L_\beta) E_{0,\beta} E_{0,\gamma} \text{Re}[K_\beta K_\gamma] \quad (D-5)
\]

where the signs of the torque depend on the relative magnitudes of the depolarization factors and the frequency \( \omega \).
Reference


VITA

Lan Lin

Education

Ph.D. 2010–2016
Electrical Engineering
The Pennsylvania State University, University Park, PA, USA

M.S. 2006–2009
Optical Engineering
Chinese Academy of Sciences, Chengdu, China

B.S. 2002-2006
Optical Engineering
Zhejiang University, Hangzhou, China

Selected Publications


