

NANOPHOTONICS AND PLASMONICS BASED BIOSENSORS

AKANKSHA NINAWE



**DEPARTMENT OF ELECTRICAL ENGINEERING
INDIAN INSTITUTE OF TECHNOLOGY DELHI**

APRIL 2022

©Indian Institute of Technology Delhi (IITD), New Delhi, 2022

NANOPHOTONICS AND PLASMONICS BASED BIOSENSORS

by

AKANKSHA NINAWE

Department of Electrical Engineering

Submitted

in fulfillment of the requirements of the degree of Doctor of Philosophy

to the



INDIAN INSTITUTE OF TECHNOLOGY DELHI

April 2022

DEDICATED

TO

MY FAMILY

CERTIFICATE

This is to certify that the thesis entitled “Nanophotonics and Plasmonics Based Biosensors” being submitted by **Ms. Akanksha Ninawe** to the **Indian Institute of Technology Delhi**, for the award of the degree of **Doctor of Philosophy** in the Department of Electrical Engineering, is a record of bonafide research work carried out by Ms. Akanksha Ninawe. Ms. Akanksha Ninawe has worked under my guidance and supervision and has fulfilled the requirements for the submission of this thesis, which to my knowledge has reached the requisite standard. The results contained in this thesis have not been submitted in part or in full to any other University or Institute for the award of any degree or diploma.

Dr. Anuj Dhawan

(Supervisor)

Department of Electrical Engineering

Indian Institute of Technology Delhi

New Delhi, 110016, India.

ACKNOWLEDGEMENTS

Firstly, I want to thank and express my sincere gratitude to my supervisor Prof. Anuj Dhawan for providing me the opportunity to pursue my research interests in the fascinating field of plasmonics. I am grateful to him for his immense knowledge, guidance and continuous support during my Ph.D. His constant motivation and generous support have been the driving force to complete my Ph.D. work.

I am thankful to the faculty members of my research committee, Prof. Swades De, Prof. Manan Suri, and Prof. P. K. Muduli, for their support during my research. I would also like to thank the faculty members and staff members of the department of electrical engineering at IIT Delhi.

I am eternally grateful to Prof. Xianfan Xu, my supervisor at Birck Nanotechnology Center, Purdue University during my overseas visiting doctoral fellowship program. I am privileged to conduct parts of my doctoral research work under his esteemed and constructive guidance and for providing me a comfortable learning space in an unfamiliar land while helping me explore my research capabilities. I also want to thank all group members of Prof. Xu's research group for an enriching learning experience at Purdue.

I wish to sincerely thank Prof. Ambarish Ghosh at IISc Bangalore for the invaluable research guidance on the collaborative work at Purdue. I also want to thank his students for the timely fabrication of the chiral samples and the intense research discussions which led to fruitful outcomes.

I wish to thank all the group members of NPPL for sharing their research insights with me while working on simultaneous projects.

I wish to acknowledge the financial support through MHRD institute fellowship for my research work at IIT Delhi. I would also like to sincerely acknowledge the financial support provided through the prestigious Overseas Visiting Doctoral Fellowship (OVDF), awarded by SERB for conducting my research activities at Purdue University.

I am indebted to my friends for being with me through thick and thin.

I would like to express my deepest gratitude to the most important people in my life, my Mother and Didi, who have unconditionally supported me in all my life's endeavors. I heartily thank my little nephew for his delightful and loving presence in my life, surely expressed beyond words.

I thank GOD for the divine protection and the blessings showered on me always.

Akanksha
March, 2022
New Delhi

ABSTRACT

Nanophotonics is a widely researched area that investigates light-matter interactions at a nanometer scale and plasmonics is one major subset in the field of nanophotonics that essentially deals with optical manipulation of light on engineered nanostructures and metamaterials. The incorporation of plasmonically engineered subwavelength nanostructures or metamaterials which can lead to optical manipulation and thus enhanced light-matter interactions resulting in various applications requires easy fabrication implementation. With the latest advances in nanofabrication technology, fabrication of subwavelength dimensions of nanostructures has become feasible using sophisticated top-down and bottom-up fabrication approaches. Further, the cost effectiveness is an important factor which should be taken into consideration in the implementation of plasmonic nanostructures and wafer-scalable metamaterials. The optical manipulation in designed subwavelength structures becomes realizable by controlling several dimensional parameters. The primary goal of this thesis is to study plasmonic sensors based on nanostructures and metamaterials majorly for three plasmonic applications, namely, surface-enhanced infrared absorption spectroscopy, narrowband absorber metamaterial and chiro-plasmonic sensing.

The first part of the thesis numerically investigates plasmonic nanostructures for efficient surface enhanced infrared absorption (SEIRA) substrates based on several configurations of bowtie nanoring and crossed-bowtie nanoring nanoantennas with embedded bowtie nanoantennas and crossed-bowtie nanoantennas. The numerical modelling is carried out using Finite-difference Time Domain (FDTD) simulations for studying the spectral properties of the nanostructures, including the spectral response in the desired spectral regime and electric field

distribution at the plasmon resonances noted in the proposed nanostructures. The SEIRA enhancement factor is found to be $\sim 1.7 \times 10^5$ which is substantially large compared to the previously reported enhancement factor values for bowtie nanoantennas or nanoring antennas.

The second part of this thesis presents a numerical modelling of simple structure for narrowband absorption in mid-infrared regime. The structure is based on simple design of a diabolo antenna which can be easily fabricated using a two-step fabrication process, essentially, a lithography and metallization process, as opposed to typical absorber designs employed tri-layered metal-insulator-metal based configurations. The two metasurfaces employed are ‘Babinet’ counterparts. The lithography step can be easily carried out using a photon polymerization laser direct write technique followed by a metallization process. The numerical modelling is carried out using Finite Element Method (FEM).

Finally, the third application studied is based on investigations of chiro-plasmonic metamaterials exhibiting giant chiro-optical responses. The chiral films, upon experimental investigations have shown a giant chiral response, the largest ever reported for a wafer-scalable chiro-plasmonic films. The samples in the study have shown large morphological variations which are inherent due to the bottom-up fabrication process employed for fabrication of the films. The experimental findings are correlated with computational models which are numerically investigated using Finite Element Method (FEM). The information from the computational predictions and its concurrence with the experimental data is highly relevant for enantiomer selective identification and sensing applications since the wafer scalable nanostructured films considered in this study are inherently porous easily allowing the diffusion of chiral molecules in regions of enhanced chiral electromagnetic hotspots.

This thesis work thus investigates key research areas of great interest to the scientific community in the field of nanophotonics and plasmonics and light-matter interactions at nanoscale focusing on applications pertaining to surface enhanced infrared absorption spectroscopy, narrowband absorption metamaterials and chirality detection using wafer-scalable metamaterials.

सारांश

नैनोफोटोनिक्स एक व्यापक रूप से शोध किया गया क्षेत्र है जो नैनोमीटर पैमाने पर प्रकाश-पदार्थ अंतःक्रियाओं की जांच करता है और नैनोफोटोनिक्स के क्षेत्र में प्लास्मोनिक्स एक प्रमुख उपसमुच्चय है जो अनिवार्य रूप से इंजीनियर नैनोस्ट्रक्चर और मेटामटेरियल्स पर प्रकाश के ऑप्टिकल हेरफेर से संबंधित है। प्लास्मोनिक रूप से इंजीनियर सबवेवलेंथ नैनोस्ट्रक्चर या मेटामटेरियल्स का समावेश जो ऑप्टिकल हेरफेर को जन्म दे सकता है और इस प्रकार विभिन्न अनुप्रयोगों के परिणामस्वरूप बढ़े हुए प्रकाश-पदार्थ की बातचीत को आसान निर्माण कार्यान्वयन की आवश्यकता होती है। नैनोफाइब्रिकेशन तकनीक में नवीनतम प्रगति के साथ, परिष्कृत टॉप-डाउन और बॉटम-अप फैब्रिकेशन दृष्टिकोणों का उपयोग करके नैनोस्ट्रक्चर के सबवेवलेंथ आयामों का निर्माण संभव हो गया है। इसके अलावा, लागत प्रभावशीलता एक महत्वपूर्ण कारक है जिसे प्लास्मोनिक नैनोस्ट्रक्चर और वेफर-स्केलेबल मेटामटेरियल्स के कार्यान्वयन में ध्यान में रखा जाना चाहिए। डिज़ाइन किए गए सबवेवलेंथ संरचनाओं में ऑप्टिकल हेरफेर कई आयामी मापदंडों को नियंत्रित करके साकार हो जाता है। इस थीसिस का प्राथमिक लक्ष्य तीन प्लास्मोनिक अनुप्रयोगों, अर्थात् सतह-वर्धित अवरक्त अवशोषण स्पेक्ट्रोस्कोपी, नैरोबैंड अवशोषक मेटामटेरियल और कायरो-प्लास्मोनिक सेंसिंग के लिए प्रमुख रूप से नैनोस्ट्रक्चर और मेटामटेरियल पर आधारित प्लास्मोनिक सेंसर का अध्ययन करना है।

थीसिस का पहला भाग कुशल सतह वर्धित अवरक्त अवशोषण (SEIRA) सबस्ट्रेट्स के लिए संख्यात्मक रूप से प्लास्मोनिक नैनोस्ट्रक्चर की जांच करता है, जो कि बोटाई नैनोरिंग और क्रॉस-बोटाई नैनोरिंग नैनोएन्टेनस के कई कॉन्फिगरेशन के आधार पर एम्बेडेड बोटाई नैनोएंटेना और क्रॉस-बोटाई नैनोएंटेनस के साथ होता है। संख्यात्मक मॉडलिंग को नैनोस्ट्रक्चर के वर्णक्रमीय गुणों का अध्ययन करने के लिए परिमित-

अंतर समय डोमेन (FDTD) सिमुलेशन का उपयोग करके किया जाता है, जिसमें वांछित वर्णक्रमीय शासन में वर्णक्रमीय प्रतिक्रिया और प्रस्तावित नैनोस्ट्रक्चर में नोट किए गए प्लास्मोन प्रतिध्वनि पर विद्युत क्षेत्र वितरण शामिल है। SEIRA एन्हांसमेंट फैक्टर $\sim 1.7 \times 10^5$ पाया गया है जो कि बोटाई नैनोएंटेना या नैनोरिंग एंटेना के लिए पहले बताए गए एन्हांसमेंट फैक्टर वैल्यू की तुलना में काफी बड़ा है।

इस थीसिस का दूसरा भाग मध्य-अवरक्त शासन में नैरोबैंड अवशोषण के लिए सरल संरचना का एक संख्यात्मक मॉडलिंग प्रस्तुत करता है। संरचना एक डायबोलो एंटीना के एक साधारण डिजाइन पर आधारित है जिसे आसानी से दो-चरण निर्माण प्रक्रिया का उपयोग करके बनाया जा सकता है, अनिवार्य रूप से, एक लिथोग्राफी और धातुकरण प्रक्रिया, त्रि-स्तरित धातु-इन्सुलेटर-धातु आधारित कॉन्फ़िगरेशन नियोजित विशिष्ट अवशोषक डिज़ाइनों के विपरीत। नियोजित दो मेटासर्फेस 'बेबिनेट' समकक्ष हैं। धातुकरण प्रक्रिया के बाद फोटॉन पोलीमराइजेशन लेजर डायरेक्ट राइट तकनीक का उपयोग करके लिथोग्राफी चरण को आसानी से किया जा सकता है। परिमित तत्व विधि (एफईएम) का उपयोग करके संख्यात्मक मॉडलिंग की जाती है।

अंत में, अध्ययन किया गया तीसरा आवेदन काइरो-प्लास्मोनिक मेटामटेरियल्स की जांच पर आधारित है जो विशाल कायरो-ऑप्टिकल प्रतिक्रियाओं को प्रदर्शित करता है। प्रायोगिक जांच पर कायरल फिल्मों ने एक विशाल कायरल प्रतिक्रिया दिखाई है, जो वेफर-स्केलेबल काइरो-प्लास्मोनिक फिल्मों के लिए अब तक की सबसे बड़ी रिपोर्ट है। अध्ययन में नमूनों ने बड़ी रूपात्मक विविधताएं दिखाई हैं जो फिल्मों के निर्माण के लिए नियोजित बॉटम-अप फैब्रिकेशन प्रक्रिया के कारण अंतर्निहित हैं। प्रयोगात्मक निष्कर्षों को कम्प्यूटेशनल मॉडल के साथ सहसंबद्ध किया जाता है, जिन्हें परिमित तत्व विधि (एफईएम) का उपयोग करके संख्यात्मक रूप से जांचा जाता है। कम्प्यूटेशनल भविष्यवाणियों की जानकारी और प्रायोगिक डेटा के साथ इसकी सहमति एनैन्टीओमर चयनात्मक पहचान और संवेदन अनुप्रयोगों के लिए अत्यधिक प्रासंगिक है क्योंकि इस अध्ययन में मानी जाने वाली वेफर स्केलेबल नैनोस्ट्रक्टेड फिल्में स्वाभाविक रूप से झरझरा

होती हैं, जो आसानी से बढ़े हुए कायरल इलेक्ट्रोमैग्नेटिक हॉटस्पॉट के क्षेत्रों में कायरल अणुओं के प्रसार की अनुमति देती हैं।

इस प्रकार यह थीसिस कार्य नैनोफोटोनिक्स और प्लास्मोनिक्स के क्षेत्र में वैज्ञानिक समुदाय के लिए बहुत रुचि के प्रमुख अनुसंधान क्षेत्रों की जांच करता है और वेफर-स्केलेबल मेटामटेरियल्स का उपयोग करके सतह वर्धित अवरक्त अवशोषण स्पेक्ट्रोस्कोपी, नैरोबैंड अवशोषण मेटामटेरियल्स और कायरलिटी डिटेक्शन से संबंधित अनुप्रयोगों पर ध्यान केंद्रित करते हुए नैनोस्केल पर प्रकाश-पदार्थ बातचीत करता है।

Contents

CERTIFICATE	i
ACKNOWLEDGEMENTS	ii
ABSTRACT	iv
Contents	vii
List of Figures	ix
List of Tables	xiv
Chapter 1: Introduction	1
1.1. Overview	1
1.2. Surface plasmons	2
1.3. Historical background and generic applications	2
1.4. This thesis	5
Chapter 2: Theory and literature review	8
2.1. Fundamentals of plasmonics	8
2.1.1. Optical properties of metals	8
2.1.2. Surface plasmon polaritons	9
2.1.3. Excitation of surface plasmons	10
2.1.4. Localized surface plasmon resonance	12
2.2. Computational methods in Electromagnetics	14
2.2.1. Finite Difference Time Domain (FDTD) method	14
2.2.1. Finite Element Method (FEM)	18
2.3. Surface enhanced infrared absorption (SEIRA) spectroscopy	19
2.3.1. Theoretical aspects of SEIRA	19
2.3.2. SEIRA substrates: Review	21
2.4. Plasmonic absorbers	23
2.4.1. Historical background	23
2.4.2. Narrowband absorbers	24
2.4.3. Multi-band or broadband absorbers	26
2.4.4. Applications of plasmonic absorbers	27
2.5. Chiral plasmonics	27
2.5.1. Introduction and background	27
2.5.2. Fabrication approaches	31

Chapter 3: Multiple-hotspot SEIRA substrates based on plasmonic bowtie nanoring antennas containing embedded nanoantennas	34
3.1 Chapter summary	34
3.2 Introduction.....	35
3.3 Numerical Finite Difference Time Domain Simulations	40
3.4 Results and Discussion	41
3.4 Conclusion	62
Supplementary data.....	63
Chapter 4: Numerical investigation of a narrowband absorber with a simple structure	66
4.1 Chapter summary	66
4.2 Introduction.....	66
4.3 Results and Discussion	69
4.4 Conclusion	80
Appendix A: FEM simulation results for TM wave illumination.....	80
Appendix B: FEM simulation results showing the effect of L variation on reflection and transmission and trapping of electromagnetic fields in dielectric spacer with H variation	81
Chapter 5: Chiro-optical response of a wafer scale metamaterial with ellipsoidal metal nanoparticles	83
5.1 Chapter summary	83
5.2 Introduction.....	84
5.3 Methods.....	86
5.3.1. Numerical simulations: FEM-COMSOL	86
5.3.2. Fabrication	87
5.3.3. Optical Characterization	89
5.4 Results and Discussion	90
5.5 Conclusions.....	105
Supplementary data.....	107
Chapter 6: Conclusions, Perspective and Prospects.....	108
6.1. Summary	108
6.2. Perspective and Prospects	109
References.....	112
Publications.....	155
BIODATA.....	156

List of Figures

Chapter 1

Figure 1.1. Demonstration of the Lycurgus cup when light is reflected through the cup (left) and transmitted through the cup (right). Reprinted from [18]. 3

Chapter 2

Figure 2. 1. (a) Schematic representation of 2-dimensional propagating transverse magnetic SPP waves at the interface of metal-dielectric in the x-direction. (b) The characteristic exponential decay of the z-component of the electric field (Ez) away from the interface. δ_1 and δ_m are the dielectric and metal penetration depths respectively. Reprinted from [58]. 9

Figure 2.2. Dispersion relation of SPPs at metal-dielectric interface established between a metal with negligible collision frequency and a dielectric (air and silica). Reprinted from [59]. 11

Figure 2.3. Excitation of SPPs using prism coupling techniques; Kretschmann and Otto configurations, respectively. Reprinted from [59]. 11

Figure 2.4. Phase matching using dielectric grating for the coupling of SPPs. Reprinted from [59]. 12

Figure 2.5. A three-dimensional Yee unit cell showing electric and magnetic field components. Reprinted from [62]. 15

Figure 2.6. Ellipticity and ORD in the band region corresponding to the absorption wavelength λ_j . The scheme represents a positive Cotton effect. Reprinted from [141]. 29

Chapter 3

Figure 3.1. (A) Schematic representation of bowtie nanostructures embedded bowtie nanoring antenna (B-B NA) in periodic array. (B) Single unit cell showing key dimensions of the structure: Hollow bowtie nanoring x-span, L ; bowtie nanoring thickness, t ; bowtie nanoring gap, G ; embedded bowtie nanoantenna gap, g ; embedded bowtie triangles side length, l ; structure height, H ; bowtie nanoring central flare angle is kept constant at 60° . O and M are at the middle of the gaps between the tips of bowtie nanoring and embedded bowtie nanostructures (C) Gap electric field enhancement spectra at points O and M correlated with far-field reflection and transmission spectral profiles. Spatial distribution of the electric field enhancement at the two resonance wavelengths $M1$ and $M2$ noted from gap electric field enhancement spectra (D) $\lambda = 3640$ nm and (E) $\lambda = 6810$ nm. The dimensions of the structure are $L = 1000$ nm, $t = 20$ nm, $l = 300$ nm, $G = 10$ nm, $g = 10$ nm, and $H = 40$ nm. 43

Figure 3.2. Demonstration of spectral tunability for applications related to SEIRA sensing with variation of L and t in B-B NA. (A) Gap electric field enhancement spectra at point O with variation of L . (B) Gap electric field enhancement at point M with variation of L . Other constant parameters are $t = 20$ nm, $G = 10$ nm, $g = 10$ nm, and $H = 40$ nm and ratio L/l is kept constant at 3.33. (C) Gap electric field enhancement at point O with variation of t . (D) Gap electric field enhancement at point M with variation of t . Other constant parameters are $L = 700$ nm, $l = 210$ nm, $G = 10$ nm, $g = 10$ nm, and $H = 40$ nm. 45

Figure 3.3. (A) Gap electric field enhancement spectra at points O and M . (B) Far-field reflection and transmission spectral profiles correlated with near-field gap electric field enhancement spectrum at point O . Spatial distribution of the electric field enhancement at the noted resonances from gap electric field enhancement spectra C) $\lambda = 940$ nm (D) $\lambda = 1640$ nm (E) $\lambda = 2880$ nm. The dimensions of the structure are $L = 350$ nm, $t = 20$ nm, $l = 100$ nm, $G = 10$ nm, $g = 10$ nm, and $H = 40$ nm..... 49

Figure 3.4. (A) Schematic representation of crossed-bowtie nanostructures embedded bowtie nanoring antenna (CB-B NA) in periodic array. (B) Single unit cell showing key dimensions of the structure: Hollow bowtie nanoring x-span, L ; bowtie nanoring thickness, t ; bowtie nanoring gap, G ; embedded crossed-bowtie nanoantenna gap, g ; embedded crossed-bowtie triangles side length, l ; structure height, H ; bowtie nanoring central flare angle is kept constant at 60° . O and M are at the middle of the gaps between the tips of bowtie nanoring and embedded crossed-bowtie nanostructures (C) Gap electric field enhancement spectra at points O and M . (D) Spatial distribution of the electric field enhancement at the main resonance $\lambda = 2870$ nm noted from gap electric field enhancement spectra. The dimensions of the structure are $L = 350$ nm, $t = 20$ nm, $l = 100$ nm, $G = 10$ nm, $g = 20$ nm, and $H = 40$ nm..... 50

Figure 3.5. (A) Schematic representation of bowtie nanostructures embedded crossed-bowtie nanoring antenna (B-CB NA) in periodic array. (B) Single unit cell showing key dimensions of the structure: Hollow crossed-bowtie nanoring x-span, L ; crossed-bowtie nanoring thickness, t ; crossed-bowtie nanoring gap, G ; embedded bowtie nanoantenna gap, g ; embedded bowtie triangles side length, l ; structure height, H ; bowtie nanoring central flare angle is kept constant at 60° . O , M and M^* are at the middle of the gaps between the tips of crossed-bowtie nanoring, embedded bowtie nanostructures along the longitudinal and crosswise cavities of crossed-bowtie nanoring antenna respectively. (C) Gap electric field enhancement spectra at points O , M , and M^* . (D) Spatial distribution of the electric field enhancement at resonance $\lambda = 2870$ nm for the hollow nanoring crossed-bowtie. (E) Spatial distribution of the electric field enhancement at resonance $\lambda = 2820$ nm for B-CB NA. The dimensions of the structure are $L = 350$ nm, $t = 20$ nm, $l = 100$ nm, $G = 20$ nm, $g = 10$ nm, and $H = 40$ nm..... 52

Figure 3.6. (A) Schematic representation of crossed-bowtie nanostructures embedded crossed-bowtie nanoring antenna (CB-CB NA) in periodic array. (B) Single unit cell showing key dimensions of the structure: Hollow crossed-bowtie nanoring x-span, L ; crossed-bowtie nanoring thickness, t ; crossed-bowtie nanoring gap, G ; embedded crossed-bowtie nanoantenna gap, g ; embedded crossed-bowtie triangles side length, l ; structure height, H ; bowtie nanoring central flare angle is kept constant at 60° . O , M and M^* are at the middle of the gaps between the tips of crossed-bowtie nanoring, embedded crossed-bowtie nanostructures along the longitudinal and crosswise cavities of crossed-bowtie nanoring antenna respectively. (C) Gap electric field enhancement spectra in case of horizontally polarized incident radiation at points O , M and M^* . (D) Spatial distribution of the electric field enhancement at resonance $\lambda = 2870$ nm in horizontal polarization incident radiation. (E) Spatial distribution of the electric field enhancement at resonance $\lambda = 2870$ nm in vertical polarization incident radiation. The dimensions of the structure are $L = 350$ nm, $t = 20$ nm, $l = 100$ nm, $G = 20$ nm, $g = 20$ nm, and $H = 40$ nm..... 53

Figure 3.7. Effect of variation of dimensional parameters of bowtie nanostructures embedded bowtie nanoring antenna (B-B NA) on the electric field enhancement spectra: (A) Gap electric field enhancement spectra at O for variation of L . (B) Gap electric field enhancement spectra at M for variation of L . (C) Gap electric field enhancement spectra at O for variation of cavity fill factor L/l . The typical dimensions of the structure are $L = 350$ nm, $t = 20$ nm, $l = 100$ nm, $G = 10$ nm, $g = 10$ nm, and $H = 40$ nm..... 57

Figure 3.8. Spatial distribution of the electric field enhancement for variation of cavity fill factor L/l for bowtie nanostructures embedded bowtie nanoring antenna (B-B NA). The typical dimensions of the structure are $L = 350$ nm, $t = 20$ nm, $l = 100$ nm, $G = 10$ nm, $g = 10$ nm, and $H = 40$ nm. 58

Figure 3.9. Effect of variation of dimensional parameters of bowtie nanostructures embedded bowtie nanoring antenna (B-B NA) on the electric field enhancement spectra: (A) Gap electric field enhancement spectra at O for variation of G . (B) Gap electric field enhancement spectra at M for variation of g . (C) Gap electric field enhancement spectra at O for variation of H . The typical dimensions of the structure are $L = 350$ nm, $t = 20$ nm, $l = 100$ nm, $G = 10$ nm, $g = 10$ nm, and $H = 40$ nm. 60

Figure 3.10. Effect of variation of dimensional parameters of bowtie nanostructures embedded bowtie nanoring antenna (B-B NA) on the electric field enhancement spectra: (A) Gap electric field enhancement spectra at O for variation of t . (B) Gap electric field enhancement spectra at M for variation of t . (C) Gap electric field enhancement spectra at O for variation of the underlying substrate. The dimensions of the structure are $L = 350$ nm, $t = 20$ nm, $l = 100$ nm, $G = 10$ nm, $g = 10$ nm, and $H = 40$ nm. 61

Figure 3.11. Fabrication process flow for the proposed bowtie and crossed bowtie nanorings with embedded nanoantennas. 64

Figure 3.12. Far-field reflection and transmission spectra correlated with near-field gap electric field enhancement spectrum at point O for (A) CB-B NA, (B) B-CB NA and (C) CB-CB NA. Electric field distributions plotted at the wavelengths corresponding to dips and peak of the transmission spectrum around the Fano spectral window noted in CB-CB NA. 64

Figure 3.13. Electric field enhancement spectra for variation of L for solid bowtie structure. Other constant parameters are $H = 40$ nm and periodicity = 2500 nm. 65

Chapter 4

Figure 4.1. (a) Schematic of gold coated diabolo array antenna based under x-polarized electric field illumination. (b) Lattice single unit of the array with structural parameters indicated. (c) Vertical cross-section view of the lattice single unit cell showing the meta-surface layer by layer design. Optimized dimensions are lattice periodicity, $\Lambda = 4$ μm ; thickness of dielectric deposited on glass, $H = 600$ nm; antenna length, $L = 2.5$ μm ; antenna width, $W = 1.5$ μm ; neck length of antenna, $l = 600$ nm; neck width of antenna, $w = 250$ nm, thickness of gold coating, $t = 55$ nm. (d) Schematic of the two-step fabrication process, a lithography or laser-direct-write process followed by a metallization process. 68

Figure 4.2. FEM calculations (a) R, T, A of our MM for x-polarized incident light with resonances at 4.065 μm and 4.277 μm . (b) Perspective views of E-field and H-field spatial distributions at resonances (c) Locations of E- and H-field hotspots (d) Spectral near-field electric field intensities at E-field hotspots (e) Spectral near-field magnetic field intensities at H-field hotspots. 70

Figure 4.3. E-field [V/m] and H-field [A/m] distribution plots and field directions at resonance wavelengths (a) 4.065 μm and (b) 4.277 μm along the transverse XY plane on surfaces S1 (row 1) and S2 (row 2). 72

Figure 4.4. Far-field A-spectra for variation of (a) L (b) W (c) H (d) Λ (e) θ in XZ plane and (f) θ in YZ plane. 74

Figure 4.5. Fano resonance investigation as a comparison of two geometrical cases of strong (optimized) and weak far-field absorptions. (a) Far-field A-spectra for cases A and B. (b) Near field spatial E-field and H-field at resonances on surface S1 (upper exposed diabolo antenna) and S2 (bottom bowtie aperture antenna) for Case B. (c) Spectral E- and H-field comparison of both cases at EM hotspots. 79

Figure 4.6. FEM simulation results (a) R, T, A of our MM for TM polarized incident light with resonances at 4.005 μm and 4.262 μm . (b) Field distribution plots and field directions at resonances along the transverse XY plane on surface S1 [upper exposed diablo antenna] (row 1) and surface S2 [bottom bowtie aperture antenna] (row 2); E-field [V/m]: H-field [A/m]..... 81

Figure 4.7. FEM simulation results (a) R and (b) T spectra for variation of L (c) Surface E-field distribution at S1 for plasmonic state in first band M1 and plasmonic state in second band M2 at the LSP wavelengths corresponding to each L varied. 82

Figure 4.8. Effect of varying H of the absorber FEM simulation results showing longitudinal ZX E-field (column 1) and H-field (column 2) distributions as a function of increasing H 82

Chapter 5

Figure 5.1. Fabrication technology. (A) Fabrication of SiO_2 helices using GLAD at 84° tilt of substrate with incoming vapor flux. (B) Coating of standing helices with silver at 10° tilt with incoming flux. 89

Figure 5.2. Scanning electron micrograph of (A) left-handed and (B) right-handed chiral samples. Spectral response of dissymmetry factor g for (C) left-handed and (D) right-handed chiral samples measured at four different locations..... 93

Figure 5.3. (A) FEM unit cell. For simulating the response of inter-helical nanostructures, periodic boundary conditions (PBC) are employed on the faces of the unit cell as shown. In the case when the response of a standalone single strand of helix is simulated, PBCs in x-y plane are replaced with perfectly matched layers (PMLs) as shown. (B) Geometrical parameters of the helical template of the nanostructure showing pitch (p), width (w) and thickness (d) of the silica helical template. (C) Three computational models of the nanoparticle distribution on the silica helical template for Plus 1 (Left-Handed sample) studied with helix simulation design parameters; turns = 2, $w = 120 \text{ nm}$, $p = 270 \text{ nm}$, and $d = 100 \text{ nm}$. Model I: All spherical particles. Model II: Modified from Model I, consists of 4 ellipsoidal particles replaced by spherical particles. Model III: Largely consists of ellipsoidal particles of varying orientations and axial ratios. (D) Existence of multipoles in the structure is shown by maps of electric field and surface current density (shown by arrows) on (i) assembly of spherical and ellipsoidal NPs in Model II at $\lambda = 610 \text{ nm}$ and (ii) assembly of ellipsoidal NPs in one iteration case of Model III at $\lambda = 612 \text{ nm}$. The length of the current arrow is proportional the current. 98

Figure 5.4. (A) Scatter plot of the center-to-center distances (r) and radii (a) of spherical particles of Model 1. (B) Histogram representing the weighted contribution of r/a values obtained from Model 1, showing most particles are at close separations (small r/a)..... 99

Figure 5.5. Comparison of simulated results with experimentally measured CD transmittance (A) FEM calculations for Model 1 for studying the effect of variation of periodicity of the lattice unit on ΔT . Each spectrum is vertically translated by 1.4. (B) FEM computed CD transmittance for the standalone single strand of helix of Model 1. (C) Comparison of ΔT for the three computational models for a standalone single strand of helix in PML boundary condition showing red-shifted spectra due to the presence of ellipsoidal particles as the model evolves from Model 1 to Model 3 (D) Measured CD transmittance for Plus 1 sample at four different locations on the sample..... 101

Figure 5.6. (A) Measured and calculated absolute transmittance. The calculated transmittance is obtained from one iteration case of Model III. (B) Measured and calculated circular differential transmittance (ΔT) response. 102

Figure 5.7. Histogram representing the weighted contribution of r/a values obtained from an average helix on the sample for an iteration case of Model 3 for randomly generated Θ_i and AR_i of the ellipsoids on the helix..... 104

Figure 5.8. (A) Calculated transmission spectra of helix of Model 3 under LCP and RCP excitation and dissymmetry factor g . (B) Maps of local optical chirality enhancements on the helix under LCP and RCP excitation at $\lambda = 691$ nm and $\lambda = 758$ nm..... 106

Figure 5.9. Experimentally measured effect of linear dichroism on (A) Left-handed sample and (B) Right-handed sample by substrate rotation..... 107

Figure 5.10. Effect of varying (A) pitch (with constant width of 120 nm) and (B) width (with constant pitch of 270 nm) of the helical template of Model 2 in PML boundary condition on dissymmetry factor g .
..... 107

List of Tables

Chapter 3

Table 3.1. Comparison of the SEIRA EFs of the proposed structures in this work with published literature 62

Chapter 5

Table 5. 1. Comparison of the experimentally realized dissymmetry factors for different small area and large-area chiro-plasmonic structures reported in literature. 93