#### **REPORT DOCUMENTATION PAGE** Form Approved OMB NO. 0704-0188 The public reporting burden for this collection of information is estimated to average 1 hour per response, including the time for reviewing instructions, searching existing data sources, gathering and maintaining the data needed, and completing and reviewing the collection of information. Send comments regarding this burden estimate or any other aspect of this collection of information, including suggesstions for reducing this burden, to Washington Headquarters Services, Directorate for Information Operations and Reports, 1215 Jefferson Davis Highway, Suite 1204, Arlington VA, 22202-4302. Respondents should be aware that notwithstanding any other provision of law, no person shall be subject to any oenalty for failing to comply with a collection of information if it does not display a currently valid OMB control number. PLEASE DO NOT RETURN YOUR FORM TO THE ABOVE ADDRESS. 2. REPORT TYPE 3. DATES COVERED (From - To) 1. REPORT DATE (DD-MM-YYYY) 15-Aug-2011 - 14-Aug-2015 21-05-2016 Final Report 4. TITLE AND SUBTITLE 5a. CONTRACT NUMBER Optical Physics and Imaging Science: Spin-optics in W911NF-11-1-0333 Metamaterials 5b. GRANT NUMBER 5c. PROGRAM ELEMENT NUMBER 611102 6. AUTHORS 5d. PROJECT NUMBER Natalia Litchinitser, Alexander Cartwright, Vladimir Drachev 5e. TASK NUMBER 5f. WORK UNIT NUMBER 7. PERFORMING ORGANIZATION NAMES AND ADDRESSES 8. PERFORMING ORGANIZATION REPORT NUMBER State University of New York (SUNY) at Bu Sponsored Projects Services 402 Crofts Hall Buffalo, NY 14260 -7016 9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS 10. SPONSOR/MONITOR'S ACRONYM(S) (ES) ARO 11. SPONSOR/MONITOR'S REPORT U.S. Army Research Office NUMBER(S) P.O. Box 12211 Research Triangle Park, NC 27709-2211 60577-PH.57 12. DISTRIBUTION AVAILIBILITY STATEMENT Approved for Public Release; Distribution Unlimited 13. SUPPLEMENTARY NOTES The views, opinions and/or findings contained in this report are those of the author(s) and should not contrued as an official Department of the Army position, policy or decision, unless so designated by other documentation. 14. ABSTRACT The objective of this project is to investigate fundamental optical phenomena at the interface of two emerging fields of modern optical physics - singular optics and optical metamaterials. Singular optics is a fascinating emerging area of modern optics that considers spin and orbital angular momentum properties of light and brings a new dimension to the science of light and physics in general. Recent developments in the field of metamaterials and transformation optics enable unprecedented control over light propagation and a possibility of "engineering" space for light now needing in structured light related abanaments in artical aburging We domanst 15. SUBJECT TERMS metamaterials, structured light, orbital angular momentum 19a. NAME OF RESPONSIBLE PERSON 17. LIMITATION OF 15. NUMBER 16. SECURITY CLASSIFICATION OF: ABSTRACT OF PAGES Natalia Litchinitser a. REPORT b. ABSTRACT c. THIS PAGE 19b. TELEPHONE NUMBER UU UU UU UU 716-645-1032

#### **Report Title**

Optical Physics and Imaging Science: Spin-optics in Metamaterials

#### ABSTRACT

The objective of this project is to investigate fundamental optical phenomena at the interface of two emerging fields of modern optical physics - singular optics and optical metamaterials. Singular optics is a fascinating emerging area of modern optics that considers spin and orbital angular momentum properties of light and brings a new dimension to the science of light and physics in general. Recent developments in the field of metamaterials and transformation optics enable unprecedented control over light propagation and a possibility of "engineering" space for light propagation, opening a new paradigm in structured light related phenomena in optical physics. We demonstrated that unique optical properties of metamaterials open unlimited prospects to "engineer" light itself. Thanks to their ability to manipulate both electric and magnetic field components, metamaterials open new degrees of freedom for tailoring complex polarization states and orbital angular momentum of light. We proposed and demonstrated several approaches to structured light manipulation on the nanoscale using metal-dielectric, all-dielectric and hyperbolic metamaterials. These new functionalities, including polarization and orbital angular momentum conversion, beam magnification and de-magnification, and sub-wavelength imaging using novel non-resonant hyperlens are likely to enable a new generation of on-chip or all-fiber structured light applications.

Enter List of papers submitted or published that acknowledge ARO support from the start of the project to the date of this printing. List the papers, including journal references, in the following categories:

(a) Papers published in peer-reviewed journals (N/A for none)

Received Paper

- 05/21/2016 33.00 Vashista C. de Silva, Piotr Nyga, Vladimir P. Drachev. Scattering suppression in epsilon-near-zero plasmonic fractal shells, Optical Materials Express, (10 2015): 2491. doi: 10.1364/OME.5.002491
- 05/21/2016 41.00 Jingbo Sun, Mikhail I. Shalaev, Natalia M. Litchinitser. Experimental demonstration of a non-resonant hyperlens in the visible spectral range, Nature Communications, (05 2015): 1. doi: 10.1038/ncomms8201
- 05/21/2016 40.00 Jinwei Zeng, Jie Gao, Ting S. Luk, Natalia M. Litchinitser, Xiaodong Yang. Structuring Light by Concentric-Ring Patterned Magnetic Metamaterial Cavities, Nano Letters, (08 2015): 5363. doi: 10.1021/acs.nanolett.5b01738
- 05/21/2016 39.00 Natalia M. Litchinitser, Mikhail I. Shalaev, Jingbo Sun, Alexander Tsukernik, Apra Pandey, Kirill Nikolskiy. High-Efficiency All-Dielectric Metasurfaces for Ultracompact Beam Manipulation in Transmission Mode, Nano Letters, (09 2015): 0. doi: 10.1021/acs.nanolett.5b02926
- 05/21/2016 38.00 Jingbo Sun, Xiaoming Liu, Ji Zhou, Zhaxylyk Kudyshev, Natalia M. Litchinitser. Experimental Demonstration of Anomalous Field Enhancement in All-Dielectric Transition Magnetic Metamaterials, Scientific Reports, (11 2015): 1. doi: 10.1038/srep16154
- 05/21/2016 37.00 Zhaxylyk A Kudyshev, Ildar R Gabitov, Andrei I Maimistov, Roald Z Sagdeev, Natalia M Litchinitser. Second harmonic generation in transition metamaterials, Journal of Optics, (11 2014): 114011. doi: 10.1088/2040-8978/16/11/114011
- 05/21/2016 36.00 Jingbo Sun, Xi Wang, Tianboyu Xu, Zhaxylyk A. Kudyshev, Alexander N. Cartwright, Natalia M. Litchinitser. Spinning Light on the Nanoscale, Nano Letters, (05 2014): 2726. doi: 10.1021/nl500658n
- 05/21/2016 35.00 Jingbo Sun, Jinwei Zeng, Xi Wang, Alexander N. Cartwright, Natalia M. Litchinitser. Concealing with Structured Light, Scientific Reports, (02 2014): 1. doi: 10.1038/srep04093
- 05/21/2016 34.00 A. A. Krokhin, J. Arriaga, L. N. Gumen, V. P. Drachev. High-frequency homogenization for layered hyperbolic metamaterials, Physical Review B, (02 2016): 75418. doi: 10.1103/PhysRevB.93.075418
- 08/31/2012 1.00 N. M. Litchinitser. Structured Light Meets Structured Matter, Science, (08 2012): 0. doi: 10.1126/science.1226204
- 09/01/2013 13.00 Jingbo Sun, Jinwei Zeng, Natalia M. Litchinitser. Twisting light with hyperbolic metamaterials, Optics Express, (06 2013): 14975. doi:
- 09/01/2013 14.00 Mikhail I. Shalaev, Zhaxylyk A. Kudyshev, Natalia M. Litchinitser. Twisted Light in a Nonlinear Mirror, Optics Letters (accepted), (10 2013): 0. doi:
- 09/01/2013 15.00 Zhaxylyk A. Kudyshev, Martin C. Richardson, Natalia M. Litchinitser. Microwave routing with virtual hyperbolic metamaterials, Nature Communications, (10 2013): 0. doi:

09/01/2013 16.00	Optics Communications, (03 2013): 179. doi:
09/01/2013 18.00	Xi Wang, Jingbo Sun, Apra Pandey, Alexander N. Cartwright, Jinwei Zeng, Natalia M. Litchinitser. Manipulating Complex Light with Metamaterials, Scientific Reports (in review), (01 2014): 0. doi:
09/01/2013 19.00	Natalia M. Litchinitser, Ji Zhou, Jingbo Sun. Indefinite by nature: from ultraviolet to terahertz, Asia Materials (Nature Publishing Group) (repeat review), (01 2014): 0. doi:
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Number of Papers published in peer-reviewed journals:

(b) Papers published in non-peer-reviewed journals (N/A for none)

Received Paper

TOTAL:

Number of Papers published in non peer-reviewed journals:

#### (c) Presentations

 N. M. Litchinitser, Z. A. Kudyshev, M. I. Shalaev, J. Sun, S. Will Nonlinear and Singular Optics in Metamaterials, 8th International Congress on Advanced Electromagnetic Materials in Microwaves and Optics – Metamaterials 2014 Copenhagen, Denmark, 25-30 August 2014

2. J. Sun, M. Shalaev, Z. Kudyshev, J. Zeng, X. Wang, A. N. Cartwright, and N. M. Litchinitser, Twisting Light with Metamaterials, 2014 Summer Topicals Meeting Series, 14 Jul 2014 - 16 Jul 2014, Montreal, Canada

3. N. M. Litchinitser, J. Sun, M. Shalaev, Beam Manipulation in the Meta-World, Metamaterials Science & Technology Workshop, San Diego, California, 20 July - 22 July 2015.

4. A. Bozhko, V.P. Drachev, A. Krokhin, "Surface plasmon propagation along smeared metal-dielectric interfaces," Bulletin of the American Physical Society 60 (2015).

5. N. M. Litchinitser, J. Sun, J. Zeng, X. Wang, M. Shalaev, and A. N. Cartwright, Structured light in linear and nonlinear nanostructures, Optical Manipulation Conference'14, Pacifico Yokohama, Japan, April 2014.

6. N. M. Litchinitser, J. Sun, M. I. Shalaev, Non-resonant hyperlens in the visible range, Metamaterials, Metadevices, and Metasystems, Conference 9544, SPIE Optics + Photonics 2015, San Diego, California, United States, 9 - 13 August 2015.

7. N. M. Litchinitser, Linear and Nonlinear Optics in Hyperbolic Metamaterials, Second International Workshop on Hyperbolic Metamaterials, Nonlinear Physics Centre, Australian National University, Canberra, December 2014.

8. N. M. Litchinitser, Structured Light in Photonic Metamaterials, Photonics North 2014, Montreal, Canada, May 2014.

## Non Peer-Reviewed Conference Proceeding publications (other than abstracts):

Received	Paper
09/01/2013 29.00	G. Ruane, P. Kanburapa , G. Swartzlander, C.F. Carlson. A Vortex-phase Filtering Scheme for Obtaining Spatial Information from an ArbitraryUnresolved Source, CEIS University Technology Showcase 03/26/13. 26-MAR-13, . : ,
TOTAL:	1

#### **Peer-Reviewed Conference Proceeding publications (other than abstracts):**

Received	Paper
05/21/2016 45.00	N. M. Litchinitser, Z. A. Kudyshev, M. I. Shalaev , J. Sun , S. Will . Nonlinear and Singular Optics in Metamaterials , 8th International Congress on Advanced Electromagnetic Materials in Microwaves and Optics – Metamaterials 2014Copenhagen, Denmark, 25-30 August 2014 . 25-AUG-14, . : ,
05/21/2016 51.00	Natalia M. Litchinitser, Jingbo Sun, Mikhail I. Shalaev, Tianboyu Xu, Yun Xu, Apra Pandey. Structured light-matter interactions in optical nanostructures, SPIE Optics & Photonics. 31-AUG-15, . : ,
05/21/2016 50.00	Jingbo Sun, Natalia M. Litchinitser, Mikhail I. Shalaev. Non-resonant hyperlens in the visible range , SPIE Optics&Photonics, Proceedings Volume 9544: Metamaterials, Metadevices, and Metasystems 2015. 01-SEP-15, . : ,
05/21/2016 48.00	Xi Wang, Tianboyu Xu, Alexander N. Cartwright, Natalia M. Litchinitser , Jingbo Sun. Twisting light using nano-waveguide arrays , CLEO/QELS. 05-JUN-14, . : ,
05/21/2016 46.00	Jinwei Zeng, Xi Wang, Alexander N. Cartwright, Natalia M. Litchinitser, Jingbo Sun, Mikhail Shalaev, Zhaxylyk Kudyshev. Twisting Light with Metamaterials, 2014 IEEE Photonics Society Summer Topical Meeting Series. 14-JUL-14, Montreal, QC, Canada. : ,
08/31/2012 4.00	Jinwei Zeng, Apra Pandey, Xi Wang, Steven Shipsey, Alexander N. Cartwright, and Natalia M. Litchinitser. Optical Vortices in Metamaterials, SPIE Optics+Photonics, Metamaterials. 15-AUG-12, . : ,
08/31/2012 5.00	Natalia M. Litchinitser, Apra Pandey, Jinwei Zeng, Xi Wang, Steven Shipsey, and Alexander N. Cartwright. Unconventional Light in Unconventional Materials, SPIE Optics+Photonics, Metamaterials. 15-AUG-12, . : ,
09/01/2013 21.00	Mikhail I. Shalaev, Zhaxylyk A. Kudyshev, Alexander Cartwright, Natalia M. Litchinitser. Second Harmonic Generation with Optical Vortices in Negative-Index Metamaterials, CLEO: QELS_Fundamental ScienceSan Jose, California United StatesJune 9-14, 2013. 09-JUN-13, . : ,
09/01/2013 22.00	Jingbo Sun, Jinwei Zeng, Alexander N. Cartwright, Natalia M. Litchinitser . Beam Transformations with Indefinite Metamaterials, CLEO: QELS_Fundamental Science (CLEO_QELS) 2013 paper: QTu3A.3. 09-JUN-13, . : ,
09/01/2013 26.00	Fatema Alali, Natalia M. Litchinitser. Gaussian Beams in Near-Zero Transition Metamaterials, Frontiers in Optics. 14-OCT-12, . : ,
09/01/2013 27.00	Apra Pandey, Jinwei Zeng, Xi Wang, Alexander N. Cartwright, Natalia M. Litchinitser. Interaction of Structured Light with Metamaterials, Frontiers in Optics. 14-OCT-12, . : ,
09/01/2013 30.00	Garreth J. Ruane, Grover A. Swartzlander. Elliptical vortex coronagraph, Frontiers in Optics. 14-OCT-12, . : ,
09/01/2013 32.00	Ke Liu, Huina Xu, Haifeng Hu, Qiaoqiang Gan, Cartwright, A.N. Rainbow-colored photonic bandgap structure fabricated by holographic lithography,

SPIE Optics. 15-AUG-12, . : ,

#### Number of Peer-Reviewed Conference Proceeding publications (other than abstracts):

## (d) Manuscripts

Received	Paper
05/21/2016 53.00	Vashista C. de Silva, Piotr Nyga, Vladimir P. Drachev. Scattering Suppression of Silica Microspheres with Semicontinuous Plasmonic Shell, arXiv:1501.00233 (01 2015)
08/31/2012 9.00	Garreth J. Ruane and Grover A. Swartzlander, Jr Optical vortex coronagraphy with an elliptical aperture, Optics Express (07 2012)
08/31/2012 10.00	Fatema Alali, Natalia M. Litchinitser. Gaussian Beams in Near-Zero Transition Metamaterials, Optics Express (08 2012)
09/01/2013 28.00	Jingbo Sun,, Jinwei Zeng, Natalia M. Litchinitser. Cloaking with Structured Light: Hiding in the Darkness, Science (to be submitted) (09 2013)
09/01/2013 31.00	G. Ruane, G. Swartzlander. Structured Darkness, Nature Photonics (to be submitted) (01 2014)
TOTAL:	5

#### Number of Manuscripts:

Books

Received Book

TOTAL:

05/21/2016 42.00 Jingbo Sun, Natalia Litchinitser. Metamaterials, United States: Woodhead Publishing;, (01 2016)

- 05/21/2016 44.00 Jinwei Zeng , Xi Wang, Mikhail I. Shalaev, Alexander N. Cartwright, Natalia M. Litchinitser. Tailoring Nonlinear Interactions in Metamaterials, Switzerland: Springer, (01 2015)
- 05/21/2016 43.00 Natalia M. Litchinitser, Vladimir M. Shalaev. Metamaterials: State-of-the Art and Future Directions, Hoboken, NJ, USA: John Wiley & Sons, Inc., (01 2015)
- 09/01/2013 12.00 Natalia M. Litchinitser, Vladimir M. Shalaev. Metamaterials: state-of-the art and future directions, Wiley: Photonics series edited by David Andrews, Chapter 22 of Volume 2/Nanophotonic Structures and Materials, (01 2014)

TOTAL:

4

#### Patents Submitted

#### **Patents Awarded**

#### Awards

During the time of this award, the PI, Natalia Litchinitser, was proposed to Associate Professor with tenure in 2011, and to Full Professor in 2014. She was appointed as a Visiting Professor, Chiba University, Japan, 2014?present time.

Other awards (Litchinitser):

Fellow of the American Physical Society (APS), 2014

Fellow of the Optical Society of America (OSA), 2011

UB Exceptional Scholar Sustained Achievements Award (2014)

School of Engineering and Applied Sciences Senior Researcher of the Year Award (2015)

Best Abstract Award in the 43rd Winter Colloquium on the Physics of Quantum Electronics,

Snowbird, Utah, January 6?10, 2013. The Winter Colloquium on the Physics of Quantum Electronics, known as PQE is an annual physics conference organized by Prof. Marlan Scully, attracts the world's experts in laser physics, quantum physics, and many other areas.

Co-PI Vladimir Drachev was tenured in 2015.

Co-PI Alexander Cartwright was appointed as Provost and Executive Vice Chancellor by the SUNY Trustees as of September 15, 2014 and named Interim President of the Research Foundation for SUNY on January 23, 2014 Dr. Cartwright was named to the Carnegie Math Pathways Advisory Board by the Carnegie Foundation for the Advancement of Teaching. In September of 2015, Dr. Cartwright was appointed by NYS Governor Andrew Cuomo to the Photonics Institute Board of Officers. In December of 2014, Dr. Cartwright earned appointment as a Fellow of the National Academy of Inventors. He is a Fellow of SPIE - The International Society for Optical Engineering since 2014. His technology for fabricating a rainbow-colored polymer using a one-step, low-cost holographic lithography method was one of just five inventions worldwide to be named to the Society of Manufacturing Engineers (SME)'s 2013 list of Innovations that Could Change the Way You Manufacture.

Graduate Students			
NAME	PERCENT_SUPPORTED	Discipline	
Ethan Gibson	0.09		
Chicheng Ma	0.06		
Mikhail Shalaev	0.35		
Salih Silahli	0.01		
Xi Wang	0.19		
Tianboyu Xu	0.06		
Yun Xu	0.10		
Jinwei Zeng	0.27		
Apra Pandey	0.00		
Tania Moein	0.00		
Garreth Ruane	0.25		
Kevin Roccapriore	0.10		
FTE Equivalent:	1.48		
Total Number:	12		

## **Names of Post Doctorates**

NAME	PERCENT_SUPPORTED	
Jingbo Sun	1.00	
Wiktor Walasik	0.04	
Zhaxylyk Kudyshev	0.00	
FTE Equivalent:	1.04	
Total Number:	3	

Names of <b>F</b>	Faculty	Supported
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NAME	PERCENT_SUPPORTED	National Academy Member
Natalia Litchinitser	0.10	
Alexander Cartwright	0.02	
Grover Swartzlander	0.08	
Vladimir Drachev	0.10	
FTE Equivalent:	0.30	
Total Number:	4	

## Names of Under Graduate students supported

NAME	PERCENT_SUPPORTED	Discipline
Yining Zang	0.00	Electrical Engineering
Ricardo Lopez	0.00	Texas Academy of Math and Science at UNT
Evan Hathaway	0.00	Physics
James Bader	0.00	Physics
Trevor Bontke	0.00	Physics
Scott Will	0.00	Electrical Engineering
FTE Equivalent:	0.00	
Total Number:	6	

Student Metrics
This section only applies to graduating undergraduates supported by this agreement in this reporting period
The number of undergraduates funded by this agreement who graduated during this period: 2.00 The number of undergraduates funded by this agreement who graduated during this period with a degree in science, mathematics, engineering, or technology fields: 2.00
The number of undergraduates funded by your agreement who graduated during this period and will continue to pursue a graduate or Ph.D. degree in science, mathematics, engineering, or technology fields: 2.00
Number of graduating undergraduates who achieved a 3.5 GPA to 4.0 (4.0 max scale): 2.00 Number of graduating undergraduates funded by a DoD funded Center of Excellence grant for Education, Research and Engineering: 0.00
The number of undergraduates funded by your agreement who graduated during this period and intend to work for the Department of Defense 0.00
The number of undergraduates funded by your agreement who graduated during this period and will receive scholarships or fellowships for further studies in science, mathematics, engineering or technology fields: 0.00

## Names of Personnel receiving masters degrees

<u>NAME</u>

**Total Number:** 

## Names of personnel receiving PHDs

NAME		
Ethan Gibson		
Jinwei Zeng		
Xi Wang		
Apra Pandey		
Tania Moein		
Total Number:	5	
	Names of other research staff	
NAME	PERCENT_SUPPORTED	
FTE Equivalent:		

**Total Number:** 

Sub Contr	ractors (DD882)			
1 a. University of North Texas1 b. 1155 Union			n Circle #305250	
	Denton	ТХ	762035017	
Sub Contractor Numbers (c): R889645				
Patent Clause Number (d-1):				
Patent Date (d-2):				
Work Description (e): -the subcontractor s	hell conduct fabrication and charac	cterization of	the optical metamater	
Sub Contract Award Date (f-1): 2/14/14 12:00AM				
Sub Contract Est Completion Date(f-2): 2/14/16 12:00AM				
1 a. University of North Texas	a. University of North Texas 1 b. Office of Sponsored Projects			
	PO Box 305	5250		
	Denton	TX	762035017	
Sub Contractor Numbers (c): R889645				
Patent Clause Number (d-1):				
Patent Date (d-2):				
Work Description (e): -the subcontractor s	hell conduct fabrication and charac	cterization of	the optical metamater	
Sub Contract Award Date (f-1): 2/14/14 12:00AM				
Sub Contract Est Completion Date(f-2): 2/14/16 12:00AM				
Inventi	ons (DD882)			

**Scientific Progress** 

See attachment

## **Technology Transfer**

Established collaboration with the NRL group on structured light in metamaterials (Dr. Jas Sanghera, Dr. Jesse Frantz).

## FINAL REPORT

### Natalia Litchinitser, Alexander Cartwright, and Vladimir Drachev

#### **RESEARCH AREA 12: PHYSICS 12.5** Optical Physics and Imaging Science: Spin-optics in Metamaterials

## Report Period Begin Date: 08/15/2011, Report Period End Date: 08/14/2015

### 1. Foreword

The synergy of complex materials and complex light is expected to add a new dimension to the science of light and its applications. Please see our Perspective article: Structured Light meets Structured Matter, Science, Vol. 337 no. 6098 pp. 1054-1055 (2012).

#### 2. Table of Contents

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3. List of Illustrations	

**Figure 1.** M. I. Shalaev, J. Sun, A. Tsukernik, A. Pandey, K. Nikolskiy, and N. M. Litchinitser, High-Efficiency All-Dielectric Metasurfaces for Ultra-Compact Beam Manipulation in Transmission Mode, Nano Lett. 15, pp 6261–6266 (2015).

**Figure 2.** J. Sun, J. Zeng, and N. M. Litchinitser, Twisting light with hyperbolic metamaterials, Opt. Express 21, 14975-14981 (2013).

**Figure 3.** M. I. Shalaev, Z. A. Kudyshev, and N. M. Litchinitser, Second Harmonic Generation with Optical Vortices in Negative-Index Metamaterials, Opt. Lett. 38, 4288-4291 (2013).

**Figure 4.** Left: J. Zeng, X. Wang, A. Pandey, J. Sun, A. N. Cartwright, and N. M. Litchinitser, Manipulating Complex Light with Metamaterials, Nature Publishing Group Scientific Reports, 1-6 DOI: 10.1038/srep02826 (2013). Right: J. Zeng, J. Gao, T. S. Luk, N. M. Litchinitser, and X. Yang, Structuring Light by Concentric-Ring Patterned Magnetic Metamaterial Cavities, Nano Lett. 15, 5363–5368 (2015).

**Figure 5.** J. Sun, J. Zeng, X. Wang, A. N. Cartwright, and N. M. Litchinitser, Concealing with Structured Light, Scientific Reports 4, 4093, 2014 [Highlighted in IEEE Spectrum]

**Figure 6.** Schematic of a nano-waveguide array that induces wave front shaping; SEM picture of the nano-waveguide array prepared by FIB. J. Sun, X. Wang, T. Xu, Z. A. Kudyshev, A. N. Cartwright, and N. M. Litchinitser, Spinning Light on the Nanoscale, Nano Lett. 14 2726, 2014 [Highlighted in Nature Nanotechnology].

**Figure 7.** J. Sun, N. M. Litchinitser, J. Zhou, Indefinite by nature: from ultraviolet to terahertz, ACS Photonics, 2014 (on the cover).

**Figure 8.** J. Sun, M. I. Shalaev, and N. M. Litchinitser, Experimental Demonstration of Non-Resonant Hyperlens in the Visible Range, Nature Communications 6, Article number: 7201 (2015). doi:10.1038/ncomms8201.

**Figure 9.** J. Sun, X. Liu, J. Zhou, Z. Kudyshev, and N. M. Litchinitser, Experimental Demonstration of Anomalous Field Enhancement in All-Dielectric Transition Magnetic Metamaterials, NPG Scientific Reports 5, 16154 (2015).

Figure 10. Homemade cost effective small-sample spectrometer.

**Figure 11.** (a) Scanning electron microscopy image of the fishnet film; The sample had the following layers deposited onto it: 10 nm Al<sub>2</sub>O<sub>3</sub>/45 nm Ag/42 nm Al<sub>2</sub>O<sub>3</sub>/45 nm Ag/10 nm Al<sub>2</sub>O<sub>3</sub>. (b) Scanning electron microscopy image of the fishnet film; The vertical substructure of the sample contains 10 nm Al<sub>2</sub>O<sub>3</sub>/45 nm Ag/42 nm Al<sub>2</sub>O<sub>3</sub>/45 nm Ag/10 nm Al<sub>2</sub>O<sub>3</sub>.

**Figure 12.** Absorption spectra of DNA and aromatic proteins. The CoNP plasmon resonance in absorption is shown in red, optical density in arbitrary units. The red bar shows the Raman shift range at the laser excitation wavelength 266 nm. The enhancement of the CoNPs will cover this range.

### 4. Statement of the Problem Studied

The objective of this project was to investigate fundamental optical phenomena at the interface of two emerging fields of modern optical physics - singular optics and optical metamaterials. Singular optics (or Structured Light) is a fascinating emerging area of modern optics that considers spin and orbital angular momentum properties of light and brings a new dimension to the science of light and physics in general. Optics facilitates the realization of many spin- and orbital angular momentum related effects that were predicted in a myriad of other physical systems where direct experimental observations are challenging or impossible. Moreover, recent developments in the field of metamaterials and transformation optics enable unprecedented control over light propagation and a possibility of "engineering" space for light propagation, opening a new paradigm in structured light related phenomena in optical physics.

#### 5. Summary of the Most Important Results

#### 5.1 High-Efficiency All-Dielectric Metasurfaces for Ultra-Compact Beam Manipulation

Metasurfaces are two-dimensional structures enabling complete control on light amplitude, phase, and polarization. Unlike plasmonic metasurfaces, silicon structures facilitate high transmission, low losses and compatibility with existing semiconductor technologies. We have experimentally demonstrated an all-dielectric resonant metasurface with full 0-to- $2\pi$  phase control at near infra-red (NIR) wavelength. We designed and fabricated high-efficiency beam deflector and light converter for generating optical vortex beam, carrying an orbital angular momentum (OAM). In addition to 0 to pi phase control enabled by a single magnetic or electric resonance, overlapping of magnetic and electric resonances in the frequency domain enable an additional phase control with optical impedance matching and, as a result, high efficiency in transmission mode with full  $2\pi$  phase manipulation.

Fabricated devices made with silicon and, in sharp contrast to plasmonic metasurfaces, are compatible with complementary metal–oxide–semiconductor technology. Demonstrated metasurfaces have relatively high transmission coefficients of 36% for beam deflector and 45% for vortex beam converter. These devices can be fabricated in one lithographical step, alleviating the need for cascading or working in reflection mode to achieve full phase control. Silicon-based metasurfaces can be used for fabrication of miniaturized high-efficiency optical components for NIR photonics, such as flat lenses, beam deflectors, anti-reflection coatings and phase modulators. Elimination of metals and, as a result, no Ohmic losses, compared to plasmonic counterparts, and not requiring cross-polarized field interaction to cover full phase, makes them well-suited for integration on optical chip and for large-scale production.



**Figure 1.** M. I. Shalaev, J. Sun, A. Tsukernik, A. Pandey, K. Nikolskiy, and N. M. Litchinitser, High-Efficiency All-Dielectric Metasurfaces for Ultra-Compact Beam Manipulation in Transmission Mode, Nano Lett. 15, pp 6261–6266 (2015).

## 5.2 Twisting Light with Hyperbolic Metamaterials

In this project we proposed a novel, miniaturized astigmatic optical element based on a single biaxial hyperbolic metamaterial that enables the conversion of Hermite-Gaussian beams into vortex beams carrying an orbital angular momentum and vice versa. As an example, we designed a biaxial anisotropic metamaterial that introduces a  $\pi/2$  phase shift between two orthogonal components of a Hermite-Gaussian beam due to the optical path difference and at the same time astigmatically focuses these orthogonal components such that they recombine in a symmetric Laguerre-Gaussian beam. The proposed device will be realized using an array of silver



**Figure 2.** J. Sun, J. Zeng, and N. M. Litchinitser, Twisting light with hyperbolic metamaterials, Opt. Express 21, 14975-14981 (2013).

nanowires in dielectric matrix.

The advantages of the proposed approach over the existing bulk optics based techniques include compactness and therefore, compatibility with ultra-compact opto-electronic circuits, potential reconfigurability and an increased tolerance to misalignment.

## 5.3 Structured Light in a Nonlinear Mirror

Opposite directionality of the Poynting vector and the wave vector, an inherent property of negative index metamaterials, was predicted to enable backward phase-matching condition for a second harmonic generation process. As a result, such a nonlinear negative index slab acts as a nonlinear mirror. In our initial studies, we predicted that second harmonic generation with structured light carrying orbital angular momentum and propagating in negative index metamaterials results in a possibility of generating a backward propagating beam with simultaneously doubled frequency, orbital angular momentum and reversed rotation direction of the wavefront. These results may find applications for high-dimensional communication systems, quantum information processing, and optical manipulation on nanoscale. Currently we are developing an experimental setup for the realization of second harmonic generation in negative index materials.



**Figure 3.** M. I. Shalaev, Z. A. Kudyshev, and N. M. Litchinitser, Second Harmonic Generation with Optical Vortices in Negative-Index Metamaterials, Opt. Lett. 38, 4288-4291 (2013).

#### 5.4 Manipulating Structured Light with Metamaterials

Recent developments in the field of metamaterials have revealed unparalleled opportunities for "engineering" space for light propagation; opening a new paradigm in spin- and quantum-related phenomena in optical physics. In this project we investigate how unique optical properties of metamaterials could be used to "engineer" light itself. We proposed and demonstrated for the first time a novel way of complex light manipulation in few-mode optical fibers using optical metamaterials. Most importantly, these studies highlight how unique properties of metamaterials, namely the ability to manipulate both electric and magnetic field components of electromagnetic



**Figure 4.** Left: J. Zeng, X. Wang, A. Pandey, J. Sun, A. N. Cartwright, and N. M. Litchinitser, Manipulating Complex Light with Metamaterials, Nature Publishing Group Scientific Reports, 1-6 DOI: 10.1038/srep02826 (2013). Right: J. Zeng, J. Gao, T. S. Luk, N. M. Litchinitser, and X. Yang, Structuring Light by Concentric-Ring Patterned Magnetic Metamaterial Cavities, Nano Lett. 15, 5363–5368 (2015).

waves, open new degrees of freedom in engineering complex polarization states of light at will, while preserving its orbital angular momentum state. These results lay the first steps in manipulating complex light in optical fibers, likely providing new opportunities for high capacity communication systems, quantum information, and on-chip signal processing.

### 5.5 Cloaking with Structured Light: Hiding in the Darkness

While cloaking made its way from the domain of science fiction to a real world demonstration, a majority of the proposed cloaking structures are capable of cloaking microscopic objects and require complex materials fabrication. Typically, cloaking devices rely on structuring optical space for light with metamaterials to mold light around the object. However, design complexity, polarization effects, bandwidth, losses and the physical size of the cloaks are some of the remaining challenges that preclude the development of practical applications especially at optical frequencies. A possible alternative to structuring the "space" to mold light propagation is to structure the light itself. Structured light already enabled such fascinating functionalities as the vortex coronagraph, or optical traps and tweezers.



**Figure 5.** J. Sun, J. Zeng, X. Wang, A. N. Cartwright, and N. M. Litchinitser, Concealing with Structured Light, Scientific Reports 4, 4093, 2014 [Highlighted in IEEE Spectrum]

We developed a macroscopic cloak based on structured light, an optical vortex. An optical vortex is light twisted in a corkscrew fashion around its axis of propagation and having a singularity (darkness) in the center. The dark core of a vortex beam can be used to conceal a macroscopic object such as, for example, a metal rod. The proposed design consists of two spiral phase plates enclosing a microscopic object. The first spiral phase plate transforms the incident beam into a vortex beam with a helical wave front that passes around the object without touching it.

The second phase plate subsequently transforms the vortex beam back to its original form. The proposed cloak is polarization independent, easy to fabricate, operates at wavelengths ranging from 560 to 700 nm, and can be used to cloak macroscopic objects as long as they are smaller than vortex core. The design can be extended to realize a multi-directional cloaking using a series of spiral plates pairs and opens a route for the realization of macroscopic, lossless, and polarization independent cloaking devices.

#### **5.6 Spinning Light on the Nanoscale**

The synergy between structured light and nanostructured materials opens entirely new opportunities in fundamental and applied science. Potential applications include imaging, increasing capacity through space division multiplexing, micromanipulation, probing of atomic forbidden states and building higher dimensional quantum encryption systems<sup>1</sup>. Many of these new applications call for ultra-compact sources of structured light, or beams carrying orbital angular momentum that can be integrated on a chip or directly on an optical fiber. However, until now, a majority of approaches to generating orbital angular momentum beams have been based

on macroscopic bulky optical components such as spiral phase plates, cylindrical lens converters, q-plates, spatial light modulators, or specialty fibers.

Recent progress in nanostructured optical materials has enabled new ways of manipulating intensity, polarization and phase distribution of light beams. Nanostructures and metamaterials



**Figure 6.** Schematic of a nano-waveguide array that induces wave front shaping; SEM picture of the nano-waveguide array prepared by FIB. J. Sun, X. Wang, T. Xu, Z. A. Kudyshev, A. N. Cartwright, and N. M. Litchinitser, Spinning Light on the Nanoscale, Nano Lett. 14 2726, 2014 [Highlighted in Nature Nanotechnology].

facilitate a new class of planar optical elements where their optical properties originate from spatial distribution of their refractive index rather than their shape. Moreover, arrays of nano-slits or nano-holes milled in a metallic film have been demonstrated to produce converging lenses or diverging lenses by realizing different propagation constants in the slits or holes of different sizes. These structures open new paths towards the realization of ultra-compact optical components for beam manipulation on the nanoscale.

In this work, we design and fabricate an array of nano-waveguides with a circular graded size distribution, which changes the propagation constant in a prescribed way. Each nano-waveguide introduces a specific phase change determined by its radius, and therefore, by carefully choosing the spatial distribution of the nano-waveguide radii, a total phase change of  $2\square$  can be imposed on the wave front of the beam upon its propagation through such an array. As a result, a conventional (Gaussian) laser beam transmitted through such a structure acquires an OAM and is

transformed into a vortex beam. Such structures are compact, versatile and can be readily integrated with optical fibers or on

## 5.7 Indefinite by Nature: from Ultraviolet to Terahertz

A class of strongly anisotropic materials having their principle elements of dielectric permittivity or magnetic permeability tensors of opposite signs, so-called indefinite or hyperbolic materials, has recently attracted a significant attention. These materials enabled such novel properties and potential applications as all-angle negative refraction, high density of states, and imaging beyond diffraction limit using a so-called hyperlens. While several studies identified a few examples of negative refractions in birefringence crystals existing in



**Figure 7.** J. Sun, N. M. Litchinitser, J. Zhou, Indefinite by nature: from ultraviolet to terahertz, ACS Photonics, 2014 (on the cover).

nature, a majority of known to date optical materials with hyperbolic dispersion relations are engineered composite materials, "metamaterials," such as metal-dielectric subwavelength multilayered structures or metal nanowires in a dielectric matrix. We investigate naturally existing indefinite materials for a range of frequencies from terahertz to ultraviolet. These include graphite, MgB<sub>2</sub>, cuprate and ruthenate. Spectroscopic ellipsometry was used to characterize the dielectric properties of graphite and MgB<sub>2</sub>, and a fitting method based on reflectance spectra is used to determine the indefinite permittivity of the cuprate and ruthenate. We investigate the mechanisms behind indefinite properties of these materials.

#### 5.8 Near-Field Optical Fiber Endoscopy for High Resolution Imaging

State-of-the-art optical endoscopy is non- or minimally invasive technique, but its resolution is limited. For the reference, naked eye resolution is  $\sim 125$ mm, typical endoscope resolution is  $\sim 10$ mm, and the best demonstrated resolution with advanced image processing is  $\sim 0.3$ mm. The resolution is limited by diffraction. Hyperlens can overcome the diffraction limit. A graded-Index waveguide can be used to transmit the image. Our solution was to combine these two capabilities to demonstrate sub-wavelength resolution endoscope.



**Figure 8.** J. Sun, M. I. Shalaev, and N. M. Litchinitser, Experimental Demonstration of Non-Resonant Hyperlens in the Visible Range, Nature Communications 6, Article number: 7201 (2015). doi:10.1038/ncomms8201.

This project was focused on the following three tasks: i) Development of a fiber endoscope with subwavelength resolution; ii) Demonstration of a fiber-coupled metamaterials hyperlens; iii) Demonstration of submicron resolution imaging device for in-vivo diagnostics. This device is based on strongly anisotropic metamaterials that feature opposite signs of the two permittivity tensor components. Such metamaterials have been shown to support propagating waves with very large wave numbers (that would evanescently decay in ordinary dielectrics). The hyperlens converts the evanescent waves into propagating waves that are consequently imaged by conventional optical systems, like microscopes, in the far field that could only be combined with conventional imaging systems through free space bulk optics.

A metamaterial hyperlens offers a unique solution to overcome the diffraction limit by transforming evanescent waves responsible for imaging subwavelength features of an object into propagating waves. However, the first realizations of optical hyperlenses were limited by a narrow working bandwidth and significant resonance-induced loss. Recently, we experimentally demonstrated a non-resonant waveguide-coupled hyperlens operating in the visible wavelength range. A detailed investigation of various materials systems proves that a radial fan-shaped configuration is superior to the concentric layer-based configuration in that it relies on non-

resonant negative dielectric response, and, as a result, enables broadband and low-loss performance in the visible range.

#### **5.9** Nonlinear Optics in Transition Metamaterials

Transition metamaterials, a class of artificial graded-index materials with dielectric permittivity and magnetic permeability gradually changing from positive to negative value have attracted significant attention in the past several years. Light propagation in such structures is of great interest from the perspective of both fundamental science and application, owing in particular to

the prediction of strong field enhancement near the zero-refractive-index point when under oblique incidence of electromagnetic radiation. This prediction opens unparalleled opportunities for efficient nonlinear optical wave interactions, such as second harmonic generation, sum and difference frequency generation, and other parametric processes at significantly reduced input intensities.

The physics of the phenomena of resonant field enhancement in such graded-index metamaterials can be summarized as follows: For incident, transverse-magnetic polarized waves, the thin layer near the zero-index point (transition point) can be considered as a very thin capacitor that accumulates infinitely large electric field energy if we neglect the effects of dissipation and spatial dispersion. Note that such energy accumulation occurs only for obliquely incident waves, since the electric field at oblique incidence has a nonzero component in the direction of propagation. Because electric displacement is necessarily continuous, the electric field anomalously increases in magnitude as dielectric permittivity tends to zero. To date, most studies of electromagnetic wave propagation in transition metamaterials have been limited to linear wave propagation. However, strong and localized field enhancement near the transition point would result in entirely new regimes of electromagnetic wave mixing with reduced input intensities. In our recent theoretical studies we predicted that resonant field enhancement of obliquely incident light in a quadratically nonlinear transition metamaterial enables an ultra-compact platform for the second harmonic generation at significantly reduced input powers. The remaining question is how to realize transition metamaterials in order to experimentally demonstrate the effects predicted using analytical and numerical approaches.



**Figure 9.** J. Sun, X. Liu, J. Zhou, Z. Kudyshev, and N. M. Litchinitser, Experimental Demonstration of Anomalous Field Enhancement in All-Dielectric Transition Magnetic Metamaterials, **NPG Scientific Reports** 5, 16154 (2015).

Since fabrication of bulk, graded-index double-negative (i.e. both  $\varepsilon$  and  $\mu$  being negative) metamaterials is still challenging, and material losses are still problematically high, our current work is focused on the design and fabrication of hyperbolic metamaterials with graded refractive index that possess significantly lower losses.



**5.10 Instrumentation (UNT contribution)** 

**Figure 20.** Homemade cost effective small-sample spectrometer.

The robust spectroscopy technique for the characterization of metamaterial samples with small area of about 30  $\mu$ m has been developed and installed at UNT and SUNY. The collection part of the system is shown in Fig.10.

The main advantage is about 7 times lower cost relative to the commercial systems, which would allow doing spectroscopy of small samples in transmission and reflection modes for two linear orthogonal polarizations.

# **5.11 Optimization of the Fishnet Nanofabrication with e-Beam Lithography (UNT contribution)**

The goal of this nano-fabrication effort was to optimize parameters of the ebeam lithography method to make possible fabrication of the fishnet with the period close to 200 nm. The fishnet structure of 400 nm period shown in Fig. 11(a) was fabricated with e-beam writing (SUNY) and following by metal deposition and lift-off (UNT). It shows good quality of the metal structure.

The visually good fishnet can be made down to 200 nm period. The 280 nm period grating with



**Figure 11.** (a) Scanning electron microscopy image of the fishnet film; The sample had the following layers deposited onto it: 10 nm  $Al_2O_3/45$  nm Ag/42 nm  $Al_2O_3/45$  nm Ag/10 nm  $Al_2O_3$ . (b) Scanning electron microscopy image of the fishnet film; The vertical substructure of the sample contains 10 nm  $Al_2O_3/45$  nm Ag/42 nm  $Al_2O_3/45$  nm Ag/10 nm  $Al_2O_3/45$  nm Ag/10 nm  $Al_2O_3/45$  nm Ag/10 nm  $Al_2O_3$ .

an appropriate for optical applications quality is shown in Fig. 11(b).

# **5.12** High Quality UV Plasmonics in Cobalt Nanoparticles: A Potential Building Block for UV Metamaterials (UNT contribution)

The goal of this effort is to develop fundamentals and applications of high quality UV plasmonics in magnetic nanoparticles.

Electron spin dependent optical and photonic phenomena in metals at nanoscale merge two fields, spintronics and plasmonics. Various technological applications of magnetic nanoparticles makes this activity truly interdisciplinary and include high-density recording, magnetic processing, catalysis and biomedical applications such as magnetic resonance imaging, cells separation, drug targeting and delivery, and magnetic fluid hyperthermia and cancer therapy. Spin dependent optics is demonstrated for hybrid, magnetic-plasmonic systems like Co/Au particles where an enhanced magnetic field controlled attenuation of the propagated light have been demonstrated due to dynamic, electromagnetically induced electron spin accumulation in the nonmagnet. The multilayer magnetic/plasmonic nanostructructures can enable ultrafast control in hybrid nanophotonic devices for future telecommunications and data recording technologies. These hybrids involve noble metals, Ag or Au, since the quality of their plasmon resonance is highest. The plasmon resonance of magnetic nanoparticles such as Co is in the ultraviolet spectral range, which is the range for bio-molecules resonances and attractive for the two-photon absorption in the visible. However, it is common belief that the quality of the plasmon resonance of Co is quite low, which follows, in particular, from the experimental data for permittivity of bulk cobalt by Johnson and Christy. A long lasting search for plasmonic materials in the ultraviolet spectral range does not consider Co as a promising candidate. One of



**Figure 12.** Absorption spectra of DNA and aromatic proteins. The CoNP plasmon resonance in absorption is shown in red, optical density in arbitrary units. The red bar shows the Raman shift range at the laser excitation wavelength 266 nm. The enhancement of the CoNPs will cover this range.

resonance in magnetic nanoparticles.

the criteria for a high quality plasmonic material is that the number of electrons involved in interband transitions must be low, and at the highest possible frequency. This simple and quite obvious criteria significantly reduces the number of materials that are likely to have favorable optical properties, by the simple fact that all materials with partially occupied d or f states are going to perform poorly across the visible due to interband transitions.

We found that Co nanoparticles with high quality crystal structure support an excellent plasmon resonance at about 275 nm, which is comparable with the noble metals. Exchange interaction of electrons splits the energy bands between spin-up (majority) electrons and spin-down (minority) electrons so that mostly minority electrons with a partially populated d-band affect the plasmon In these studies we involve magnetic properties of Co and find that magnet selected nanoparticles show high-quality plasmon resonance. The suggested mechanism involves an effect of the spin polarization on the density of states of d-band and, as a consequence, on the Co nanoparticle permittivity. Our experiments show that Co nanoparticles have the quality factor of the resonance in the deep UV as good as for Au in the visible range as it is illustrated in Fig. 12.

Our results show that chemically synthesized Co nanoparticles is the only DUV plasmonic nanoparticles with magnetic properties. Since this result open new properties of quite studied material, we have performed experimental studies to provide strong evidence of this statement. We propose that exchange interaction of electrons splits the energy bands between spin-up (majority) electrons and spin-down (minority) electrons so that mostly minority electrons with a partially populated d-band affect the plasmon resonance in magnetic nanoparticles.

Note that the spectral position of the plasmon resonance in CoNPs matches the oligonucleotide resonances (Fig. 12). The 260/280 ratio in absorption is well known method to characterize oligonucleotide to aromatic proteins ratio. Resonance Raman at 266 excitation wavelength shows almost no protein features. The red bar in Fig. 12 illustrates Raman shift up to 2000 cm<sup>-1</sup>. The CoNP plasmon resonance in absorption is shown in red line and suggests that whole spectral range of DNA Raman will be completely covered.

# 5.13 Scattering Suppression in Epsilon-Near-Zero Plasmonic Fractal Shells (UNT contribution)

Published in De Silva, Vashista C.; Nyga, Piotr; Drachev, Vladimir P., Optical Materials Express 5(11), 2491-2500 (2015).

Light scattering by core-shell particles made of dielectric and metal, manifests in a variety of phenomena predicted many decades ago [1-3]. The scattering suppression for coated confocal ellipsoids was introduced by Aden and Kerker [1-3] in the approximation of long wavelength planar electromagnetic waves. This scattering suppression is called "invisibility" there. In the core-shell spheres, total sizes could be up to one fifth of the wavelength to achieve sensible invisibility for planar electromagnetic waves [4,5]. In this case, the wave can penetrate through the shell and the effect is associated with the out-of-phase scattering properties of plasmonic materials and the dielectric core. By varying the relative dimensions of the dielectric core and high quality continuous metal shell, the sharp optical resonances of these nanoparticles can be varied over hundreds of nanometers in wavelength, across the visible and into the infrared region of the spectrum [6,7]. In contrast, semi-continuous shells provide broadband response similar to the planar semi-continuous films, as it was shown for the visible spectral range [8-10]. The optical properties of the metal-dielectric semi-continuous films are influenced by multiple surface plasmon resonances (SPRs) in metal nanostructures, accumulating and building up electromagnetic energy in a broad spectral range at the nanometer scale [11-22]. A universal phenomenon in the localization of optical energy in inhomogeneous plasmonic media is the formation of hot spots, spatially fluctuating field with spikes in nanometer-size regions determined by the minimum scale of the nanoplasmonic system [14,15]. The picture of nanolocalization of the optical energy in disordered clusters is called inhomogeneous localization [16,17] assuming that there are different plasmonic eigenmodes, which coexist at close frequencies and have completely different localization sizes, ranging from the minimum scale to the scale of the entire systems. Each eigenmode may consist of a different number of sharp hot spots. Note that such type of disordered geometry has typically fractal dimensions [12,13]. As it

is known for the planar fractal films, the critical value of the metal coverage, called percolation threshold [11], results in a variety of plasmon resonances covering a spectral range from the visible to infrared. Thus the fractal films being synthesized on the microspheres can be promising aerosolized obscurants in the extremely broad visible-infrared spectral range.

This work studies silica-gold core-shell microspheres with plasmonic fractal shells. The similarities and differences with the planar noble metal fractal films have been experimentally established, which were not addressed in the earlier publications [8-10]. We show that the forward scattering of the silica microspheres is strongly suppressed. Also the reflection of the fractal shells does not grow with coverage approaching the percolation threshold. This is in contrast to the planar fractal films, where the forward scattering reduces along with the backscattering increasing as the metal coverage increases. Even more counterintuitive result is that the total extinction of the core-shell is decreased relative to the bare core response. The system is simulated using Mie theory with Aden and Kerker extension [1]. The fractal shell parameters are calculated with the Bruggeman effective medium theory (EMT) [23]. We discuss the EMT applicability using the scaling theory approach [12,13]. The model provides a reasonable agreement with experiments in the broad spectral range, from 400 nm to  $20 \,\mu m$ , covering shorter and longer wavelengths relative to the microsphere size. Both experiments and simulations show that the fractal shell with metal filling factor close to 0.5 enables scattering suppression in the visible range along with the increase in total transmission at the wavelength of Mie scattering peak at about 560 nm. The results indicate that this suppression is not just a spectral shift of the resonance in scattering, but suppression of its amplitude without noticeable shift. In the infrared range the gold semicontinuous shell "hides" the absorption resonance of the silica sphere at 9 µm. The effective permittivity for our samples is shown to be epsilon-near-zero for the real part of epsilon. The imaginary epsilon multiplied by the frequency approximately does not depend on the wavelength across the broad spectral range  $0.5-20 \,\mu m$ .

- 1. A. L. Aden, and M. Kerker, "Scattering of Electromagnetic Waves from Two Concentric Spheres," J. Appl. Phys. **22**(10), 1242-1246 (1951).
- 2. M. Kerker, "Invisible Bodies," J. Opt. Soc. Am. 65(4), 376-379 (1975).
- 3. H. Chew, and M. Kerker, "Abnormally Low Electromagnetic Scattering Cross Sections," J. Opt. Soc. Am. **66**(5), 445-449 (1976).
- 4. A. Alù, and N. Engheta, "Achieving Transparency with Plasmonic and Metamaterial Coatings," Phys. Rev. E **75**, 016623 (2005).
- 5. M. Silveirinha, A. Alù, and N. Engheta, "Parallel-Plate Metamaterials for Cloaking Structures," Phys. Rev. E **75**, 036603 (2007).
- 6. S. Oldenburg, R. Averitt, S. Westcott, and N. Halas, "Nanoengineering of Optical Resonances," Chem. Phys. Lett. **288**(2-4), 243-247 (1998).
- F. Tam, A. L. Chen, J. Kundu, H. Wang, and N. J. Halas, "Mesoscopic Nanoshells: Geometry-Dependent Plasmon Resonances Beyond the Quasistatic Limit," J. Chem. Phys. 127(20), 204703 (2007).
- 8. C. Graf, and A. van Blaaderen, "Metallodielectric Colloidal Core–Shell Particles for Photonic Applications," Langmuir **18**(2), 524-534 (2002).
- T. Ji, V. G. Lirtsman, Y. Avny, and D. Davidov, "Preparation, Characterization, and Application of Au-Shell/Polystyrene Beads and Au-Shell/Magnetic Beads," Adv. Mater. 13(16), 1253-1256 (2001).

- C. A. Rohde, K. Hasegawa, and M. Deutsch, "Coherent Light Scattering from Semicontinuous Silver Nanoshells near the Percolation Threshold," Phys. Rev. Lett. 96(4), 045503 (2006).
- 11. D. Stauffer, Introduction to Percolation Theory (CRC Press, 1994).
- 12. Y. Yagil, M. Yosefin, D. J. Bergaman, G. Deutscher, and P. Gadenne, "Scaling theory for the optical properties of semicontinuous metal films," Phys. Rev. B **43**(13), 11342-11352 (1991).
- Y. Yagil, P. Gadenne, C. Julien, and G. Deutscher, "Optical properties of thin semicontinuous gold films over a wavelength range of 2.5 to 500 μm," Phys. Rev. B 46(4), 2503-2511 (1992).
- M. I. Stockman, L. N. Pandey, L.S. Muratov, and T. F. George, "Photon Scanning Tunneling Microscopy Images of Optical Excitations of Fractal Metal Colloid Clusters," Phys. Rev. Lett. 75(12), 2450 (1995).
- 15. M. I. Stockman, L. N. Pandey, L. S. Muratov, and T. F. George, "Optical Absorption and Localization of Eigenmodes in Disordered Clusters," Phys. Rev. B **51**(1), 185-195 (1995).
- 16. M. I. Stockman, L. N. Pandey, and T. F. George, "Inhomogeneous Localization of Polar Eigenmodes in Fractals," Phys. Rev. B **53**(5), 2183-2186 (1996).
- 17. M. I. Stockman, "Inhomogeneous Eigenmode Localization, Chaos, and Correlations in Large Disordered Clusters," Phys. Rev. E 56, 6494 (1997).
- S. Grésillon, L. Aigouy, A. Boccara, J. Rivoal, X. Quelin, C. Desmarest, P. Gadenne, V. Shubin, A. Sarychev, and V. Shalaev, "Experimental Observation of Localized Optical Excitations in Random Metal-Dielectric Films," Phys. Rev. Lett. 82(22), 4520-4523 (1999).
- D. A. Genov, A. K. Sarychev, and V. M. Shalaev, "Metal-Dielectric Composite Filters with Controlled Spectral Windows of Transparency," J. Nonlinear Optic. Phys. Mat. 12(4), 419-440 (2003).
- 20. P. Nyga, V. P. Drachev, M. D. Thoreson, and V. M. Shalaev, "Mid-IR Plasmonics and Photomodification with Ag Films," Appl. Phys. B **93**, 59-68 (2008).
- 21. M. D. Thoreson, J. Fang, A. V. Kildishev, L. J. Prokopeva, P. Nyga, U. K. Chettiar, V. M. Shalaev, and V. P. Drachev, "Fabrication and Realistic Modeling of Three-Dimensional Metal-Dielectric Composites," J. Nanophoton. 5(1), 051513 (2011).
- 22. A. K. Sarychev, and V. M. Shalaev, *Electrodynamics of Metamaterials* (World Scientific, 2007).
- 23. D. A. G. Bruggeman, "Berechnung Verschiedener Physikalischer Konstanten von Heterogenen Substanzen," Ann. Phys. **416**(7), 636-664 (1935).

# 5.14 High-Frequency Homogenization for Layered Hyperbolic Metamaterials (UNT contribution)

Published in A.A. Krokhin, J. Arriaga, L.N. Gumen, and V.P. Drachev, Phys. Rev. B 93, 075418 (2016).

Uniaxial metamaterials with optical anisotropy going beyond the difference in the absolute values of the components of the dielectric tensor  $\epsilon ik(\omega) = diag(\epsilon \parallel, \epsilon \parallel, \epsilon \perp)$  and showing extreme birefringence when  $\epsilon \parallel \epsilon \perp < 0$  are known as hyperbolic metamaterials [1-4]. Due to (formally) infinite values of the wave vector allowed by hyperbolic dispersion relation for propagating electromagnetic mode these materials strongly modify the rate and direction of spontaneous emission [5]. The dielectric function of a periodic structure becomes negative at sufficiently low frequencies when the contribution to polarization from the metallic layers overcomes the

contribution from the dielectric constituent. Since polarizations along the layers and perpendicular to them are different, the elements of the dielectric tensor  $\epsilon \|(\omega)$  and  $\epsilon \perp (\omega)$  vanish at different frequencies, giving rise to the frequency bands with either elliptic ( $\epsilon \|(\omega) \epsilon \perp (\omega) > 0$ ) or hyperbolic ( $\epsilon \|(\omega) \epsilon \perp (\omega) < 0$ ) dispersion. Here the sub-indices  $\|$  and  $\perp$  refer to the propagation parallel or perpendicular to the optical axis, respectively.

In order to clarify the problem of frequency dependence of the tensor of effective permittivity of a layered hyperbolic material we propose a simple homogenization scheme which takes into account the effects of frequency and spatial dispersion directly from Rytov's equation [6].

In this work we presented an analytical solution for the effective dielectric functions  $\epsilon II(\omega)$  and  $\epsilon \perp (\omega)$  and establish the limits of applicability of the Drude model with effective plasma frequencies used in the literature so far. Propagation in plane of periodicity and parallel to the layers are considered separately. In the latter case the bands originated from the evanescent surface plasmon-like mode and from the oscillating waveguide-like mode lead to different results for the effective dielectric function. The proposed method of homogenization is quite general. It is valid not only for 1D superlattice but for any periodic structure. Unlike the quasi-static approach known in the literature, our method accounts for spatial variation of the fields within the unit cell and, thus, may be valid at high frequencies. It gives the parameters of the effective medium for the parts of the dispersion curve close to the edge of the Brillouin zone where group velocity vanishes and each Bloch eigenmode becomes a standing wave with the maximum value of the Bloch vector.

Our analytical approach for calculation of the effective dielectric functions  $\epsilon \parallel(\omega)$  and  $\epsilon \perp(\omega)$  of metal-dielectric superlattices gives asymptotically correct results in the long-wavelength limit. It gives the exact positions for all frequencies of the topological transitions where one of the dielectric functions changes its sign. Near any of the frequencies of the topological transition the accuracy of the proposed theory exceed that of any other known approaches. In particular, it is shown that the widely-used approximations obtained in [6] with the quasi-static approach may be not applicable at all, or their accuracy turns out to be not sufficient for modern optical studies. The accuracy of the quasi-static approach becomes low if the width of the metallic layer exceeds the skin depth. Applications of hyperbolic metamaterials are due to their ability to increase the rate of spontaneous emission. The rate increase depends on the both components of the dielectric tensor and if even one of these components gives a considerable error as a result of quasi-static approximation, the frequency dependence of the rate of spontaneous emission may be incorrect as it is shown in Ref. [7,8]. This is of particular importance for the type 1 hyperbolic metamaterials when the component  $\epsilon \perp(\omega)$  changes its sign, since the quasi-static approach is not applicable near the frequency where  $\epsilon \perp(\omega) = 0$ .

- D. R. Smith and D. Schurig, Phys. Rev. Lett. 90, 077405 (2003); D. R. Smith, D. Schurig, J. J. Mock, P. Kolinko, and P. Rye, Appl. Phys. Lett. 84, 2244 (2004).
- 2. M. A. Noginov, Yu. A. Barnakov, G. Zhu, T. Tumkur, H. Li, and E. E. Narimanov, Appl. Phys. Lett. **94**, 151105 (2009).
- 3. V. P. Drachev, V. A. Podolskiy, and A. V. Kildishev, Opt. Express 21, 15048 (2013).
- A. Poddubny, I. Iorsh, P. Belov, and Yu. Kivshar, Nat. Photonics 7, 948 (2013).
- M. A. Noginov, H. Li, Y. A. Barnakov, D. Dryden, G. Nataraj, G. Zhu, C. E. Bonner, M. Mayy, Z. Jacob, and E. E. Narimanov, Opt. Lett. 35, 1863 (2010); A. N. Poddubny, P. A. Belov, P. Ginzburg, A. V. Zayats, and Yu.S. Kivshar, Phys. Rev. B 86, 035148 (2012); W.D. Newman,

- C.L. Cortes, and Z. Jacob, J. Opt. Soc. Am. B 30, 766 (2013); Lei Gu, J. E. Livenere, G. Zhu, T.U. Tumkur, H. Hu, C.L. Cortes, Z. Jacob, S. M. Prokes, and M.A. Noginov, Sci. Rep. 4, 7327 (2014); T. Galfsky, H.N.S. Krishnamoorthy, W. Newman, E.E. Narimanov, Z. Jacob, and V.M. Menon, Optica 2, 63 (2015).
- 6. S.M. Rytov, Sov. Phys.-JETP 2, 466 (1956).
- 7. O. Kidwai, S. V. Zhukovsky, and J. E. Sipe, Optics Letters, 36, 2530 (2011).
- 8. O. Kidwai, S.V. Zhukovsky, J. E. Sipe, Phys. Rev. A 85, 053842 (2012).