AFRL-OSR-VA-TR-2014-0176

NONLINEAR ELECTROMAGNETICS AND COHERENT ENERGY TRANSFER IN NEGATIVE INDEX METAMATERIALS

Alexander Popov
UNIVERSITY OF WISCONSIN SYSTEM

08/07/2014
Final Report

Air Force Research Laboratory
AF Office Of Scientific Research (AFOSR)/ RTB
Arlington, Virginia 22203
Air Force Materiel Command

REPORT DOCUMENTATION PAGE

Form Approved
OMB No. 0704-0188

1. REPORT DATE *(DD-MM-YYYY)*
31/07/2014

2. REPORT TYPE
Final

3. DATES COVERED *(From - To)*
1June 2012 to 31 May 2014

4. TITLE AND SUBTITLE
NONLINEAR ELECTROMAGNETICS AND COHERENT ENERGY TRANSFER IN NEGATIVE-INDEX METAMATERIALS

5a. CONTRACT NUMBER

5b. GRANT NUMBER
FA9550-12-1-0298

5c. PROGRAM ELEMENT NUMBER

6. AUTHOR(S)
Popov, Alexander K.

5d. PROJECT NUMBER

5e. TASK NUMBER

5f. WORK UNIT NUMBER

7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES)
UNIVERSITY OF WISCONSIN-STEVENS POINT
2100 MAIN ST
STEVENS POINT, WI 54481-3871

8. PERFORMING ORGANIZATION REPORT NUMBER

9. SPONSORING / MONITORING AGENCY NAME(S) AND ADDRESS(ES)
Air Force Office of Science and Research
875 Randolph Street
Suite 325 Room 3112
Arlington, VA 22203

10. SPONSOR/MONITOR'S ACRONYM(S)

11. SPONSOR/MONITOR'S REPORT NUMBER(S)

12. DISTRIBUTION / AVAILABILITY STATEMENT
Publicly available

13. SUPPLEMENTARY NOTES
Performance period of this effort was cut by one year because the PI moved from UW-SP to Birck Nanotechnology Center, Purd

14. ABSTRACT
An understanding was advanced of nonlinear propagation properties of electromagnetic (EM) waves in double-domain negative/positive index metamaterials (MMs) with focus on the coherent nonlinear-optical (NLO) energy transfer between the ordinary and backward waves (BWs), i.e. the waves with contra-directed energy flux and phase velocity. A theory was developed and proof-of-principle computational studies were conducted of the outlined processes in the context of particular MMs and their unique potential applications to photonics. Numerical simulations were carried out of the multi-parametric dependences of the solutions to the set of partial differential wave equations accounting for the backwardness of one of the coupled waves. Frequency conversion, which stems from the NLO coupling of contra-propagating short pulses, was studied. A novel approach was proposed to engineering of the MMs, which support coexistence of phase-matched ordinary and BEM eigenmodes satisfying to three-wave mixing. It is based on negative spatial dispersion. A possibility to mimic the outlined extraordinary processes using stimulated Raman scattering on BW optical phonons was shown, which enables greatly enhanced amplification of ordinary Stokes signals.

15. SUBJECT TERMS
Nonlinear optics, metamaterials, backward electromagnetic and elastic waves, coherent energy exchange between electromagn

16. SECURITY CLASSIFICATION OF: a. REPORT	b. ABSTRACT	c. THIS PAGE	17. LIMITATION OF ABSTRACT	18. NUMBER OF PAGES	19a. NAME OF RESPONSIBLE PERSON
U	U	U	UU		19b. TELEPHONE NUMBER *(include area code)*

Standard Form 298 (Rev. 8-98)
Prescribed by ANSI Std. Z39.18

FINAL PERFORMANCE REPORT

To: technicalreports@afosr.af.mil

Subject: Final Performance Report to *Dr. Arje Nachman*

Contract/Grant Title: NONLINEAR ELECTROMAGNETICS AND COHERENT ENERGY TRANSFER IN NEGATIVE-INDEX METAMATERIALS

Contract/Grant #: FA9550-12-1-0298

Reporting Period: 1 June 2012 to 31 May 2014

Considerable progress was achieved in conceptual understanding of nonlinear propagation properties of electromagnetic waves (EMWs) in the double-domain negative/positive index metamaterials with focus on the coherent energy transfer between the ordinary and backward waves. Corresponding theory was developed and computational studies were conducted of the outlined processes in the context of particular materials and their unique potential applications to photonics. Numerical simulations were carried out of the multi-parametric dependences of the numerical solutions to the set of coupled nonlinear wave equations. The equations described strongly nonlinear frequency conversion processes with account for the counter-directed phase velocity and energy flow in one of the coupled waves. Particularly, frequency conversion processes, which stem from the nonlinear-optical coupling of contra-propagating short pulses, were studied. A set of corresponding coupled partial differential equations was numerically solved and multi-parameter dependences of the solutions were numerically analyzed.

Energy flux and phase velocity of an EMW become contra-directed in negative-index meta-materials (NIMs). Such waves are referred to as *backward waves*. Given extraordinary property enables phase matching of ENWs contra-propagating in frequency double-domain positive/negative index nonlinear-optical (NLO) meta-slabs. The latter give rise to counterintuitive coherent (i.e., phase dependent) NLO propagation processes and requires a revision of major concepts generally accepted in NLO. It also requires the development of novel theoretical and computational approaches to investigation of such photonic processes in particular media. Among them is three-wave mixing, which can be viewed as a split of fundamental photons into two entangled "siblings" – pairs of signal and idler photons satisfying energy conservation law. Each of them stimulates further chain of splits. In ordinary isotropic nonlinear-optical materials, the momentum conservation law (phase matching) dictates all three beams to be co-directed and signal and idler to exponentially grow along the propagation path. The indicated dependence was predicted to change dramatically in the case of coupled ordinary and backward waves with matching phases. Here, all wave vectors must be codirected, whereas one of the Poynting vectors appears contra-directed to others. This gives rise to the extraordinary nonlinear resonances in the numbers of generated entangled photons, which emerge in respect to intensity of the fundamental wave. A huge enhancement of the output in the resonances indicates the appearance of distributed nonlinear-optical feedback. The latter is equivalent to effective coupling length tending to infinity. Such a singularity enables *greatly enhanced* energy conversion efficiency, whereas the required size of the convertor may reduce down to microscopic values. However, the investigations and the estimates revealed that intensities of fundamental radiation required for the realization of microscopic convertors in the continuous-wave regime may exceed the optical breakdown threshold. Short-pulse regime seems to offer a solution. However, strengths of EM field changes through the pulse which raises many open questions regarding the outcomes and the optimization. *In the course of the given effort*, encouraging predictions were made with the aid of particular proof-of-principles examples and significant progress in overall understanding of the outlined processes was achieved.

The possibility was shown of conversion of short ordinary EM pulses to contra-propagating pulses of backward EM wave at doubled frequency and some of the basic extraordinary features of the process were investigated. Such metaslab can be viewed as a *frequency-doubling* NLO *metamirror* and data-processing chip, which can be also used as a nonlinear-optical sensor.

Considerable attention was paid to the particular realizations of the predicted extraordinary processes. Current mainstream in fabricating bulk NIMs relies on engineering of LC nanocircuits - plasmonic mesoatoms with negative optical magnetic response. A different paradigm was proposed in the course of given effort, which relied on negative spatial dispersion of waves, to realize the outlined processes. As a proof-of-principles example of the possibility of nanoengineering of the required negative dispersion, the metamaterial made of standing carbon nanotubes was proposed. It was shown that with proper adjustment of heights and spacing between the nanotubes, the eigenmodes pertinent to the given tampered nanowaveguide can be tailored in the way that such *carbon "nanoforest"* enables coexistence of ordinary and backward eigenmodes, i.e. EMWs with positive and negative dispersion/group velocities at different frequencies. Numerical analysis of dispersion properties and losses attributed to such modes was carried out. It demonstrated that the eigenmodes can be tailored to achieve *phase matching* (i.e., equal phase velocities) for the ordinary and backward waves which satisfy the energy conservation law for three-wave mixing. For the investigated models, the frequencies fall in the THz and thermal IR wavelength ranges. As outlined above, phase matching is a requirement of critical importance for coherent NLO. The possibilities of slow-wave regime and the analogs of epsilon-near-zero propagation regime in such carbon "nanoforest" were shown too.

Besides that, a class of readily available Raman crystals was proposed to mimic the outlined extraordinary three-wave mixing propagation processes which are commonly attributed to the plasmonic NIMs. The underlying idea was to replace one of the coupled EMWs by the elastic waves having negative spatial dispersion. Negative dispersion and, consequently negative group velocity is inherent to optical phonons. However, it appeared that fast damping of optical phonons imposes a severe detrimental obstacle to realization of the idea. The possibility to eliminate that obstacle by making use of femtosecond pulses was proposed and proved through numerical simulations. Particularly, the models of diamond and calcite crystals where used for proof-of-principle numerical simulations. Unparalleled properties of enhanced Raman amplification of ordinary Stokes signals by stimulated scattering on backward phonons were investigated. Besides the possibility of huge enhancement of its quantum conversion efficiency, the possibility of tailoring duration and shapes of the generated Stokes and transmitted fundamental laser pulses were shown. It was found out that relatively low group velocity of backward optical phonons, which need to be phase matched, dictates relatively high threshold intensities of the fundamental lasers to attain the predicted extraordinary regime. It explains why it was not observed so far. However, the required pulse power was estimated as achievable with now commercially available lasers. Diamond crystals of good optical quality, which are sized at about 1 cm, have emerged recently on the market and have been advertised by major photonic journals because of their excellent thermal conductivity needed for high power Raman lasers.

A concept of remotely actuated and all-optical controlled nonlinear optical sensor was proposed based on the above described processes and materials. The sensors was supposed to frequency up-convert IR radiation emitted in the vicinity of the sensor, frequency scan and amplify the up-converted signal and send it back towards a remote detector**.**

Overall, the major outcome of this forward looking effort is a considerable progress in the fundamental understanding summarized above of unparalleled electromagnetic processes and counterintuitive nonlinear optical effects originated from the exotic electromagnetic properties of negative index/negative spatially dispersive metamaterials and from the proposed specific realization schemes. Novel coherent nonlinear-optical technique for compensating strong losses intrinsic to plasmonic metamaterials was further developed. Advanced concepts of unique nanophotonic devices as well as development of the modeling/simulation approaches to nonlinear nanophotonics are also among the outcomes of the research.

The effort may have impact on the DoD and, specifically, on the Air Force capabilities through the development of novel transformative concepts of a new generation of ultracompact photonic devices and components with enhanced functionalities. The devices hold a potential for use in the telecommunication industry, for nonlinear optical sensing and image processing as well as for the variety of biological and medical applications.

Results obtained, including recent proof-of-principle preliminary results to be further developed, were discussed with peers at *seminars and major topical international conferences* as listed below:

1. Alexander K. Popov, ``Nonlinear Optics and Negative Phase Velocity," **invited talk** at International Workshop "***Novel Ideas in Optics: From Advanced Materials to Revolutionary Applications***," May 31 - June 2, 2012, Purdue University, West Lafayette, IN, USA https://engineering.purdue.edu/~shalaev/workshop/InternationalW_rev11.pdf
2. Alexander K. Popov, "Nonlinear Optics with Backward Waves," ***invited talk at seminar*** in the Metamaterial Department of the National Research University of

Information Technologies, Mechanics and Optics, St. Petersburg, Russia, 10 July, 2012.

3. Alexander Popov, “Merging Nonlinear Nanophotonics and Negative-Index Metamaterials,” ***invited talk at seminar, Purdue University***, 8 October, 2012.
4. Alexander Popov, “Coherent Nonlinear Optics, Backward Waves and Negative Spatial Dispersion,” ***invited talk at seminar, Purdue University***, 5 May, 2014.
5. Alexander Popov, “Nonlinear optics with backward waves: enhanced tailored reflectivity and transparency extraordinary second harmonic generation, stimulated Raman scattering and parametric amplification in short pulse regimes,” Paper presented at ***Nonlinear Optics Contractors Meeting*** held by the Air Force Office of Scientific Research at the Joint Technology Office, University of New Mexico, Albuquerque, NM, 18-20 September 2012.
6. Alexander Popov, “Nonlinear Optics in Negative-index Materials: Extraordinary Features, (meta)Materials and Applications,” A talk presented at ***Nonlinear Optics Contractors Meeting*** held by the Air Force Office of Scientific Research at Basic Research Innovation Collaboration Center, Arlington, VA, 4-5 September 2013.
7. Alexander K. Popov, Igor. S. Nefedov, Sergey A. Myslivets, Mikhail I. Shalaev and Vitaly V. Slabko, “Nonlinear-optical up and down frequency-converting backward-wave metasensors and metamirrors,” Paper 8725-93, International Conference ***SPIE Defense, Security, and Sensing 2013***, Baltimore, MD, 29 April – 3 May, 2013, http://spie.org/defense-security-sensing.xml
8. A. K. Popov, M. I. Shalaev, S. A. Myslivets, V. V. Slabko, “Unidirectional amplification and shaping of optical pulses by three-wave mixing with negative phonons,“ META'13, the ***4th International Conference on Metamaterials, Photonic Crystals and Plasmonics***, Sharjah, United Arab Emirates, 18-22 March 2013. http://meta13.metaconferences.org.
9. A. K. Popov, S. A. Myslivets and I. S. Nefedov, “Generation of Short Contrapropagating Pulses of Second Harmonic on Frequency Double-Domain Positive/Negative Index Metamaterials,” Paper c12a430, Proceedings of ***Photonics Global Conference (PGC) 2012***, Singapore, December 13-16, 2012. http://www.photonicsglobal.org/download/Final_Programme.pdf .
10. A. K. Popov, S. A. Myslivets and I. S. Nefedov, “Frequency Doubling Nonlinear-optical Metamirror Made of Aligned Carbon Nanotubes,” Paper PA-SA-II-(1)-10, Proceedings of ***Optics & Photonics Taiwan, International Conference (OPTIC 2012)***, Taipei, Taiwan, December 6- 8, 2012. http://optic2012.ntu.edu.tw/agenda.php .
11. A. K. Popov, M. I. Shalaev, S. A. Myslivets, V. V. Slabko aand I. S. Nefedov, “Nonlinear Optics With Backward Waves,” Paper LM3A.4, ***Latin America Optics and Photonics Congress***, São Sebastião, Brazil, 10-13 November, 2012. http://www.osa.org/osaorg/media/osa.media/Meetings/PDFSupportingDoc/LAOP-Program-Guide-2012.pdf .

12. Alexander K. Popov, Mikhail I. Shalaev, Sergey A. Myslivets and Vitaly V. Slabko, “Photonic Devices with Negative Phonons: a Concept, Extraordinary Properties and Materials,” ***8 th International Conference on Optics-photonics Design & Fabrication*** “ODF' 12,” St. Petersburg, Russia, July 2 - 5th, 2012, Paper 4S4-02. http://odf2012.ru/file/stat/4/odf12_advance_program.pdf
13. A. K. Popov, M. I. Shalaev; S. A. Myslivets; V. V. Slabko and I. S. Nefedov, “Nonlinear backward-wave photonic metamaterials,” The 4th International Conference "***Smart Materials, Structures and Systems***," Montecatini Therme, Tuscany, Italy, June 10-14, 2012, paper A-12.3:L06. p. 46. http://www.cimtec-congress.org/2012/pdf/abstracts_cimtec_2012.pdf.
14. Alexander K. Popov, “Nonlinear Optics with Backward Waves: Extraordinary Features, Materials and Applications,” Proceedings of **The International Conference on Coherent and Nonlinear Optics** (ICONO), June 18-23, 2013, Moscow, Russia, p. 38. http://iconolat13.phys.msu.ru/ICONO_LAT-2013/Proceedings.html.
15. A. K. Popov, M. I. Shalaev, S. A. Myslivets and V. V. Slabko, “Simulating NIMs with Raman Crystals,” Proceedings of ***The International Conference on Coherent and Nonlinear Optics*** (ICONO), June 18-23, 2013, Moscow, Russia, p. 79. http://iconolat13.phys.msu.ru/ICONO_LAT-2013/Proceedings.html.
16. A. K. Popov, S. A. Myslivets, V. V. Slabko, M. I. Shalaev and I. S. Nefedov, “Photon Management by Nonlinear Coupling in Nanostructures,” ***The XXII International Workshop on Optical Wave & Waveguide Theory and Numerical Modelling OWTNM14***, Nice, France, June 27-28, 2014
17. Alexander K. Popov, Vitaly V. Slabko, Sergey A. Myslivets, and Mikhail I. Shalaev “Mimicking Frequency-mixing in Nonlinear Optical Negative-index Metamaterials: Unidirectional Raman Amplification and Shaping of Optical Pulses,” (**invited paper**) ***The 16 th International Conference*** «***Laser Optics 2014***», 30 June – 4 July 2014, St.Petersburg, Russia.
18. A. K. Popov, S. A. Myslivets and I. S. Nefedov, “Nonlinear photonics in carbon nanoforests and graphene multifences,” ***XII International Conference on Nanostructured Materials (NANO 2014),*** Moscow, July 13-18, 2014. http://www.nano2014.org/thesis/view/4362